

Transition metals in organic synthesis. Annual Survey covering the year 1992 *

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1. General comments

This Annual Survey covers the literature for 1992 dealing with the use of transition metal intermediates for organic synthetic transformations. It is not a comprehensive review but is limited to reports of discrete

systems that lead to at least moderate yields of organic compounds, or that allow unique organic transformations, even if low yields are obtained. Catalytic reactions that lead cleanly to a major product and do not involve extreme conditions are also included. This is not a critical review, but rather a listing of the papers published in the title area.

The papers in this survey are grouped primarily by reaction type rather than by organometallic reagent,

* Reprints are not available. For previous Annual Survey see *J. Organomet. Chem.*, 457 (1993) 167.

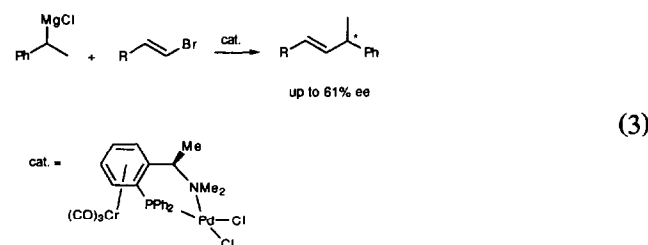
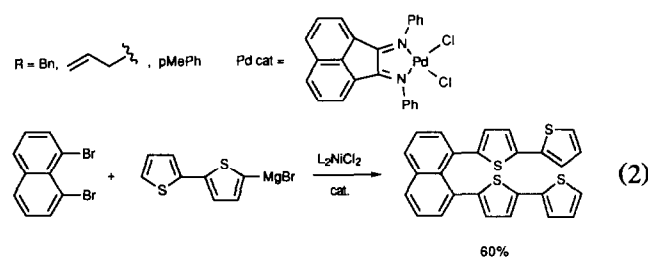
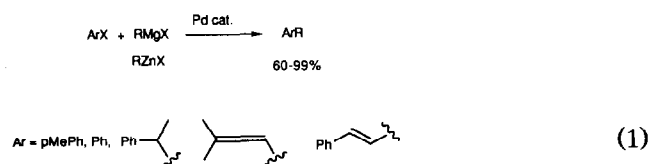
since the reader is likely to be more interested in the organic transformation effected than the metal causing it. Oxidation, reduction, and hydroformylation reactions are specifically excluded, and will be covered in a different annual survey. Also excluded are structural and mechanistic studies of organometallic systems unless they present data useful for synthetic application. Finally, reports from the patent literature have not been surveyed since patents are rarely sufficiently detailed to allow reproduction of the reported results.

2. Carbon-carbon bond-forming reactions

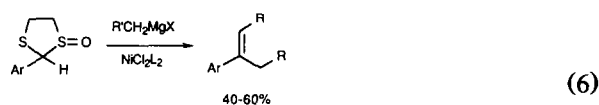
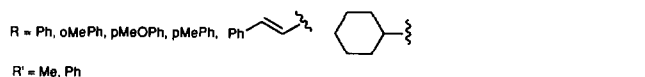
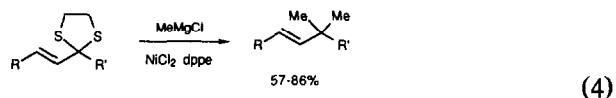
2.1. Alkylations

2.1.1. Alkylation of organic halides, tosylates, triflates, acetates and epoxides

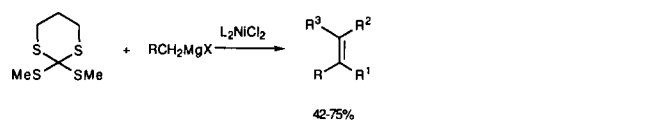
Metal-catalyzed Grignard reactions continue to be developed and applied in organic synthesis. A review entitled "Carbon-carbon bond formation in heterocycles using nickel and palladium-catalyzed reactions" has appeared [1], and deals with metal-catalyzed Grignard reactions as well as the Castro-Stevens reaction. Palladium diazine complexes catalyzed the coupling of aryl halides with Grignard and organozinc derivatives (eqn. (1) [2]). Nickel phosphine complexes catalyzed the coupling of 1,8-dibromo-naphthalene with thiophene Grignard reagents (eqn. (2) [3]). Chromium complexes of optically active aryl aminophosphines catalyzed the asymmetric coupling of phenethyl Grignard reagents with vinyl halides (eqn. (3) [4]).



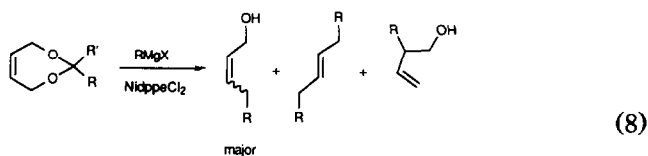
Nickel phosphine complexes catalyzed the alkylation of allylic dithianes (eqn. (4) [5]), benzylic dithianes (eqn. (5) [6] and eqn. (6) [7]), the coupling of tetrathiomethanes (eqn. (7) [8]) and the alkylation of 1,3-dioxep-4-enes by Grignard reagent (eqn. (8) [9]). Acid anhydrides were alkylated by halides in the presence of reduced nickel salts (eqn. (9) [10]).



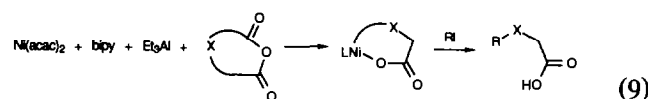
Ar = 1-Naphthyl, 2-Naphthyl, pMePh, 3,4-(MeO)₂Ph, 3,5-(MeO)₂Ph



R	R ¹	R ²	R ³
Me	Me	H	H
Bu	Bu	H	H
C ₆	C ₆	-(CH ₂) ₅	H
tBuCH ₂	tBuCH ₂	tBu	H
TMSCH ₂	TMSCH ₂	TMS	H
Ph	H	Me	H
Bu	Me	H	H

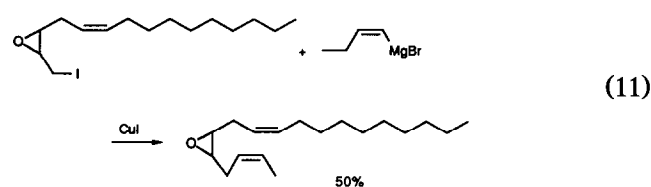
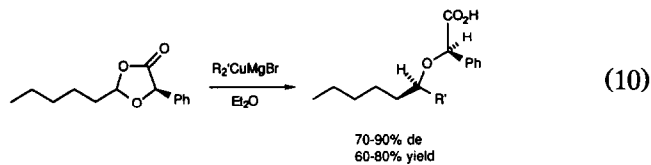


R = Et, nPr, nBu, iBu, iPr, tBu, Ph, nC₆,

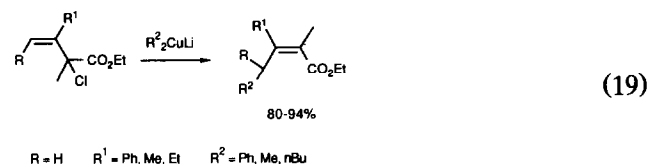
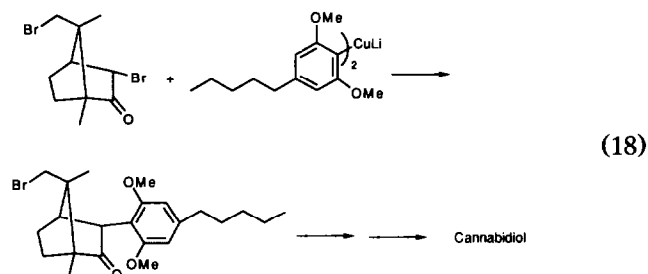
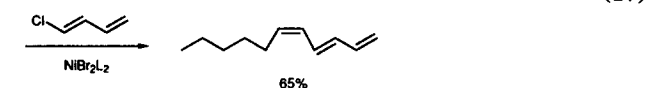
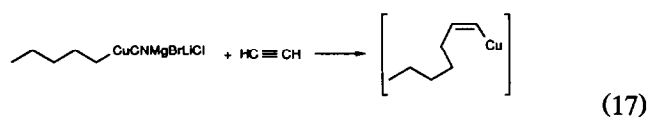
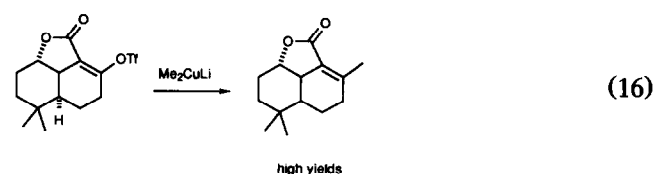
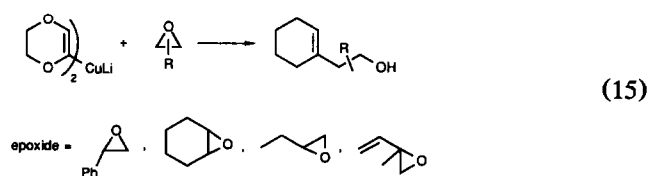
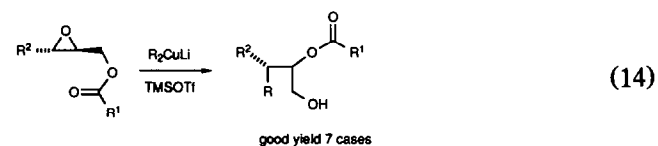
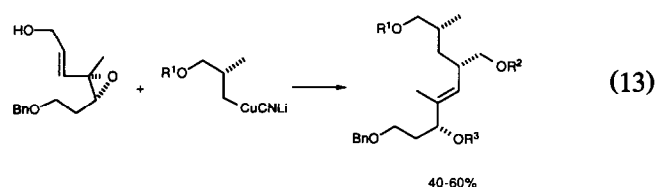
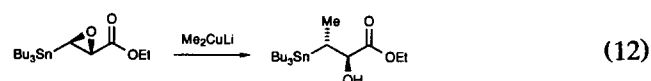


X = CH₂, (CH₂)₂, , ,

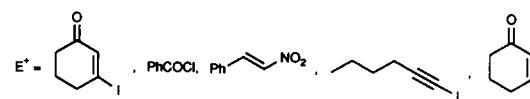
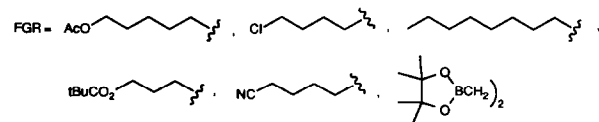
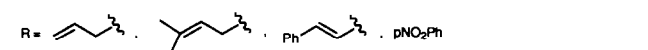
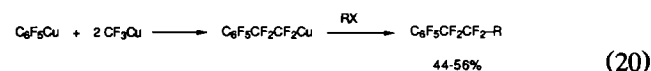
Copper catalyzed the alkylation of acetals (eqn. (10) [11]) and α -iodoepoxides (eqn. (11) [12]). Titanocene dichloride catalyzed the alkylation of gem dihaloalkanes by Grignard reagents [13].

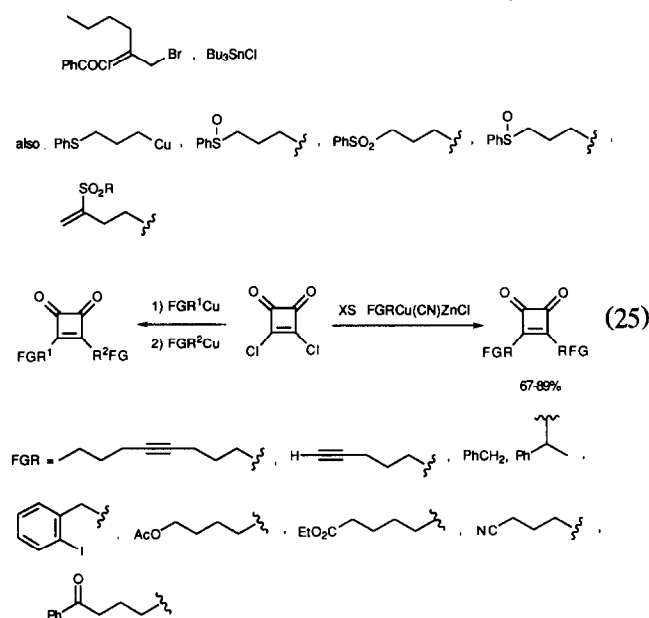
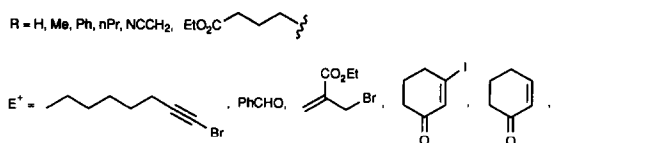
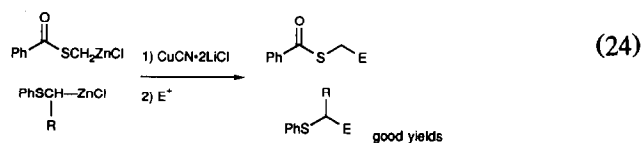
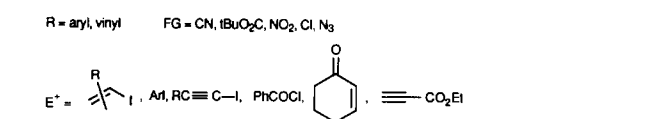
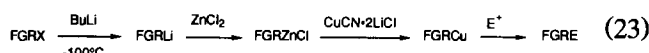
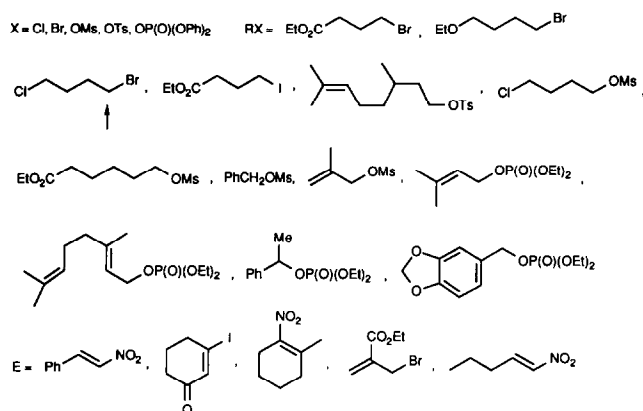
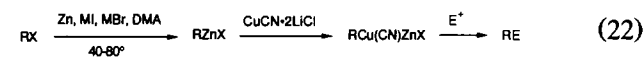


Organocuprates efficiently alkylated epoxides (eqn. (12) [14], eqn. (13) [15], eqn. (14) [16], eqn. 15 [17]) and vinyl triflates (eqn. (16) [18]). Trienes were synthesized by the coupling of chlorodienes with vinylcuprates from carbocupration of alkynes (eqn. (17) [19]). The arylation of an α -haloketone by a vinylcuprate was a key step in the synthesis of cannabidiol (eqn. (18) [20]). α -Haloallylic esters underwent alkylation by cuprates with clean S_N2' regioselectivity (eqn. (19) [21]).



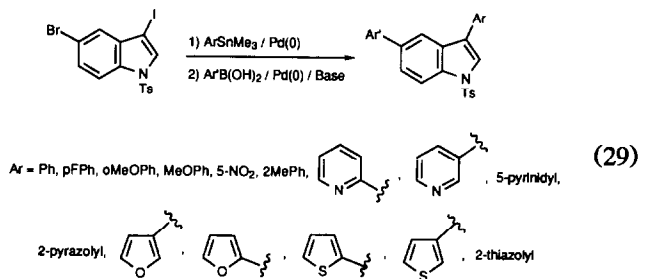
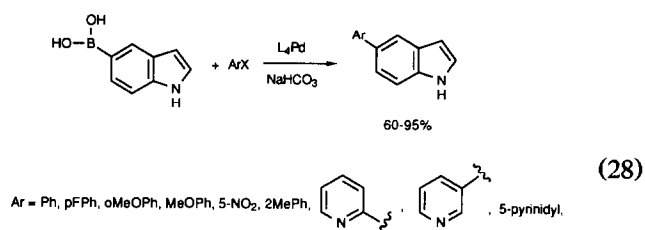
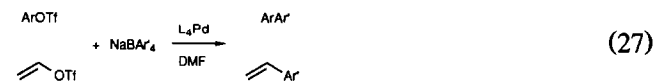
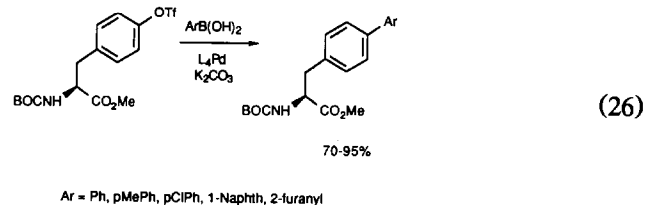
Pyrrole and imidazole cyanocuprates had typical "higher order cuprate" reactivity [22]. Perfluorophenyl copper combined with perfluoromethyl copper to insert two CF_2 groups, and this homologated fragment transferred to halides (eqn. (20) [23]). Functionalized organocuprates continued to be developed and exploited in synthesis (eqn. (21) [24], eqn. (22) [25], eqn. (23) [26], eqn. (24) [27], and eqn. (25) [28]).

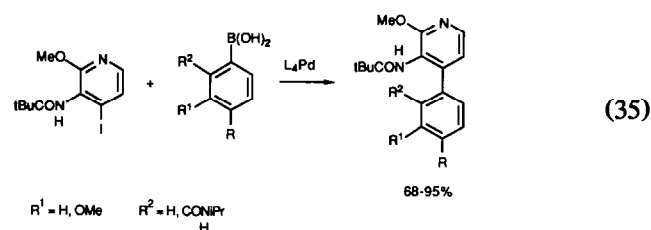
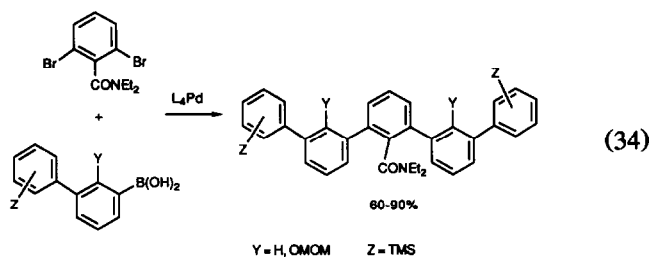
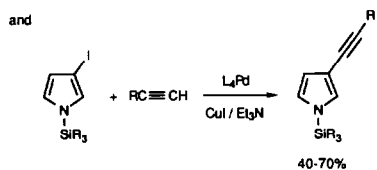
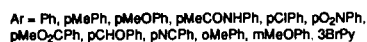
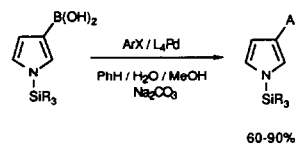
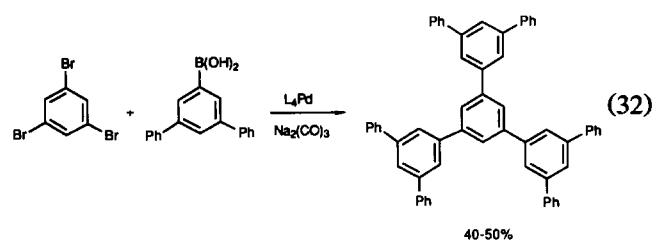
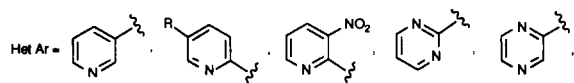
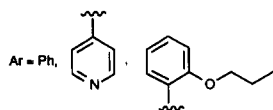




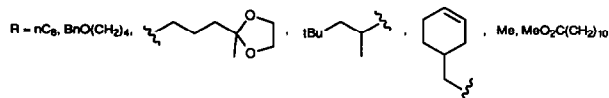
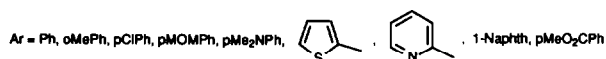
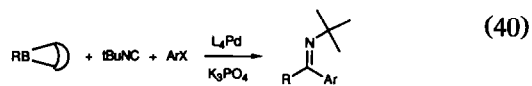
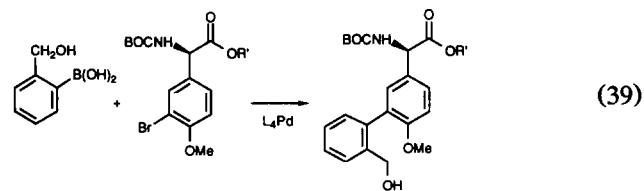
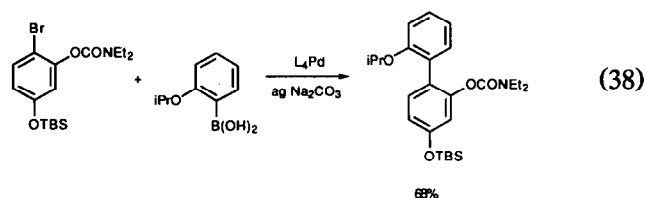
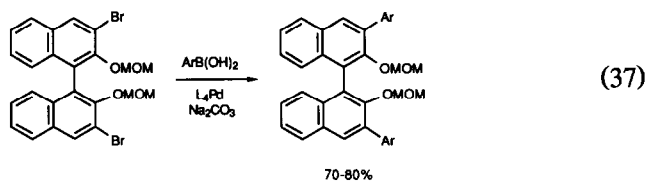
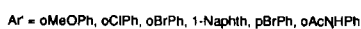
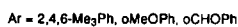
The reagents Me₄CoLi₂ and Me₄FeLi₂ were claimed to be superior to Me₂CuLi for the methylation of vinyl halides [29]. β-Haloalkenylketones were alkylated by alkyl organomanganese compounds [30]. Allylic manganese compounds alkylated γ-bromo-α,β-unsaturated carboxylic acid esters [31].

Palladium catalyzed coupling of halides and triflates to main group organometallics *via* oxidative addition/transmetalation sequences continues to be a growth industry, an indication that good ideas are easier to recognize than to get. Boron is fast becoming the main group metal of choice. Examples of coupling aryl triflates or halides to arylboronic acids abound (eqn. (26) [32], eqn. (27) [33], eqn. (28) [34], eqn. (29) [35], eqn. (30) [36], eqn. (31) [37], eqn. (32) [38], eqn. (33) [39], eqn. (34) [40], eqn. (35) [41], eqn. (36) [42], eqn. (37) [43], eqn. (38) [44], eqn. (39) [45], and eqn. (40) [46].

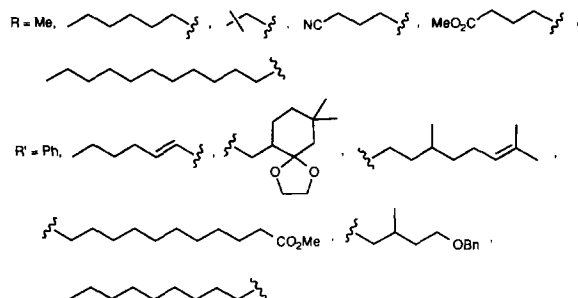
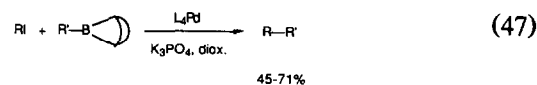
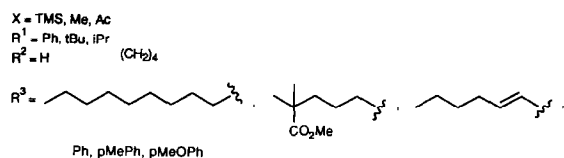
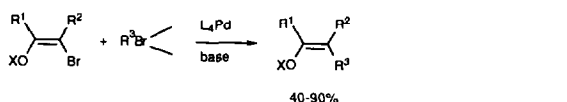
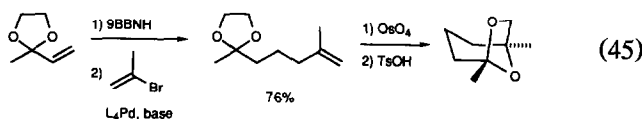
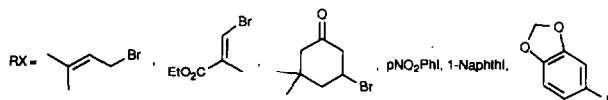
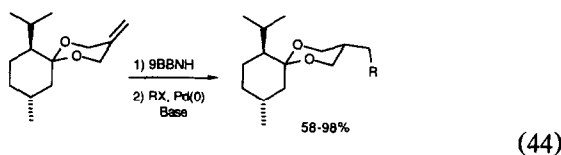
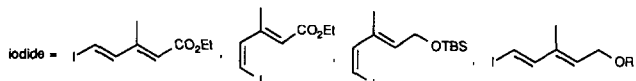
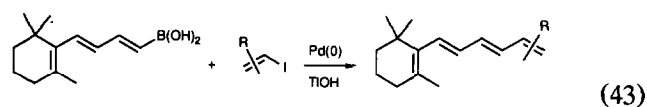
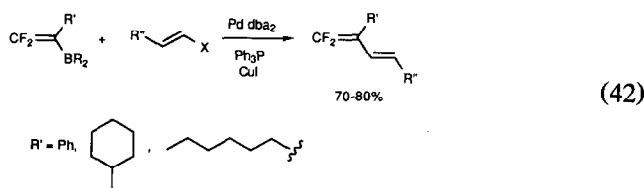
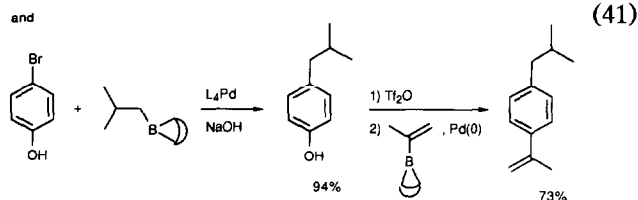
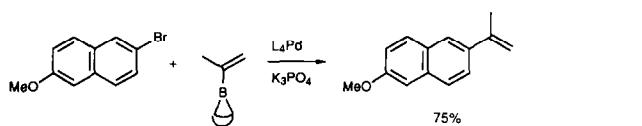




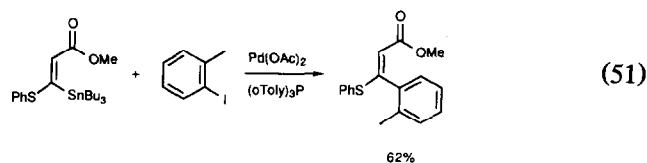
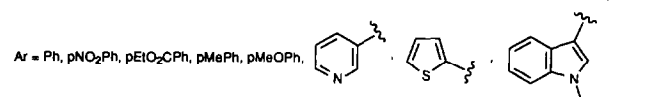
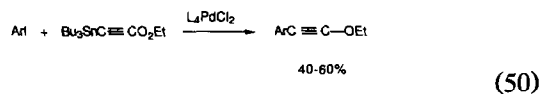
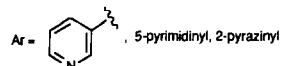
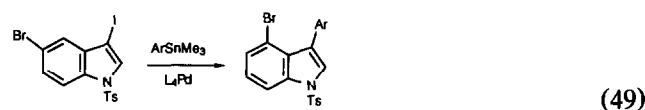
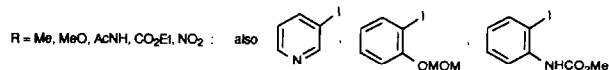
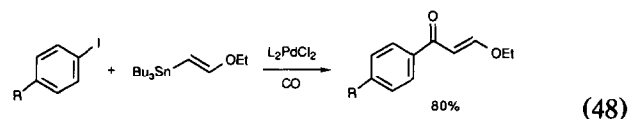
(suppresses deboronation)

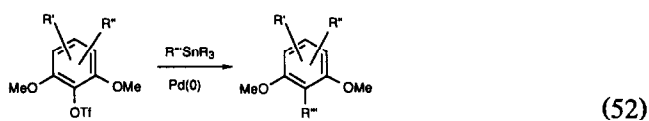


Vinyl boronates (eqn. (41) [47], eqn. (42) [48], and eqn. (43) [49]) and even alkyl boronates (eqn. (44) [50], eqn. (45) [51], and eqn. (46) [52]) couple. Remarkably, halides having β-hydrogens can also be made to undergo the palladium catalyzed coupling process with only minimal β-elimination (eqn. (47) [53]).



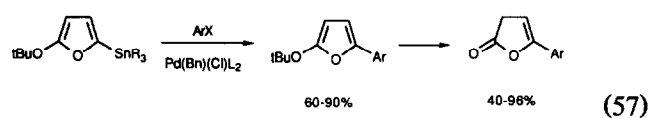
Palladium catalyzed couplings involving transmetalation from tin remained very popular and were the subject of an in depth review (174 references) [54]. Aryl halides were coupled efficiently to vinyl tin reagents (eqn. (48) [55]), aryl tin reagent (eqn. (49) [56]), acetylene tin reagents (eqn. (50) [57]) and β -stannyleneones (eqn. (51) [58]). Aryl triflates (eqn. (52) [59]) and eqn. (53) [60] and *p*-fluorophenyl sulfonates (eqn. (54) [61]) were also efficiently coupled to stannanes.



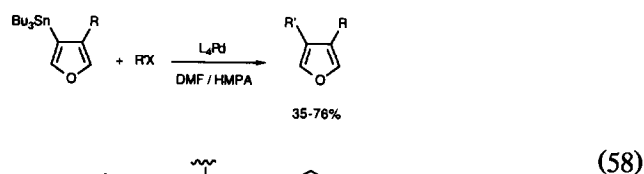
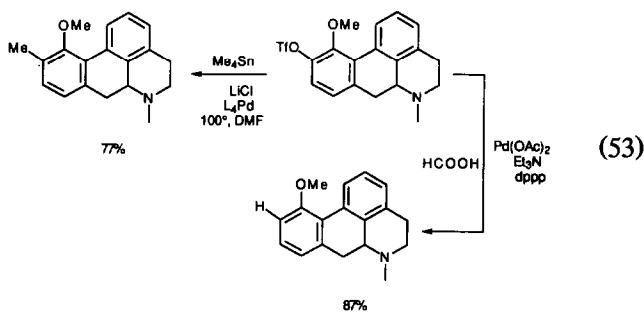


(1) Ph_3P required for electron rich systems.
 (2) >10% Pd required

R'' can be Me, Ph, $\text{PhC}\equiv\text{C}$, Ar, $\text{CH}_2\text{CH}_2\text{CH}_2$, but not nbutyl

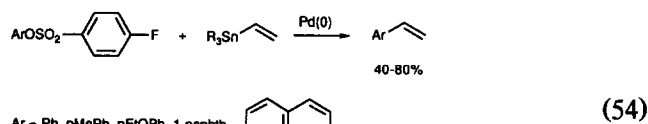
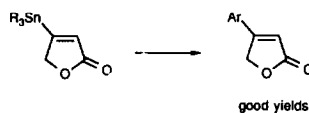
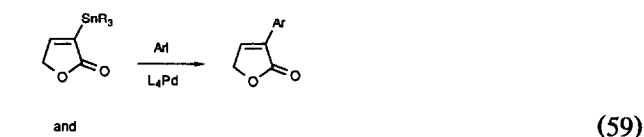


Ar = pNO_2Ph , oCF_3Ph , oMePh , pPhPh , 3-py, pClPh , CHOPh , Br , $\text{2,6-Me}_2\text{Ph}$



R = Me, HO-C(CH₃)₂-CH₂-Ph, Ph-CH(OH)-CH₂-Ph, C_6H_{11}

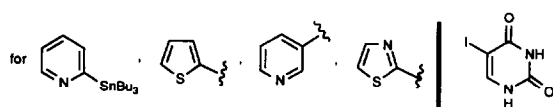
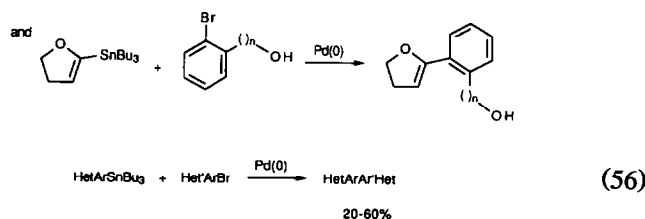
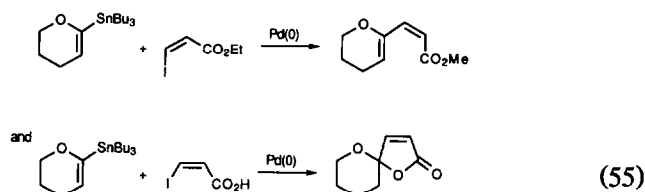
R' = pMeOPh , Ph , EtO_2C , $\text{CH}_2\text{CH}_2\text{CH}_2$



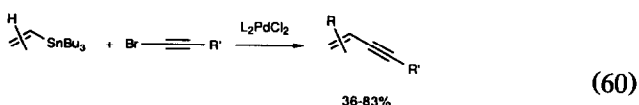
Ar = Ph, pMePh , pEtOPh , 1-naphthyl, C_6H_4

also $\text{R}_3\text{SnCH}_2\text{CH}_2$, R_3SnPh , $\text{R}_3\text{SnCH}_2\text{CH}_2\text{Ph}$ couple

Palladium also catalyzed a variety of coupling reactions with heterocyclic tin reagents (eqn. (55) [62], eqn. (56) [63], eqn. (57) [64], eqn. (58) [65], and eqn. (59) [66]).

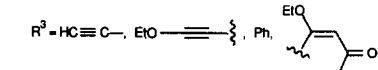
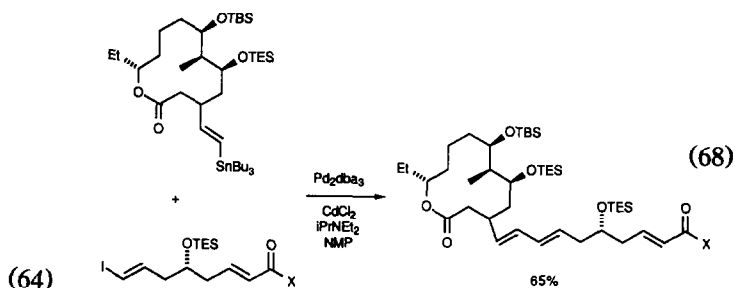
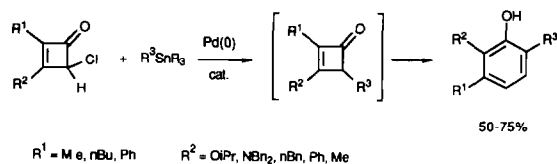
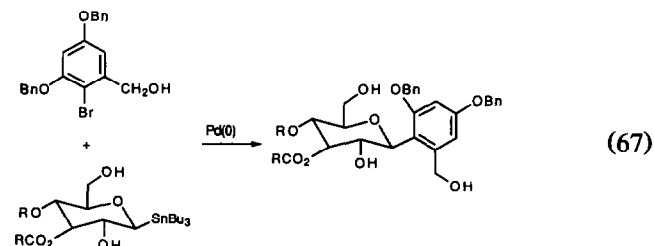
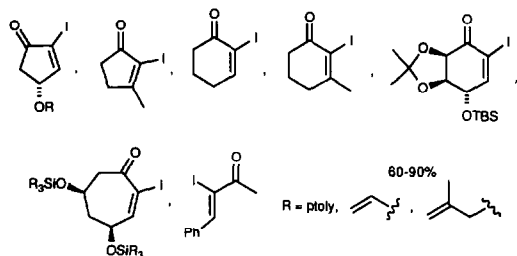
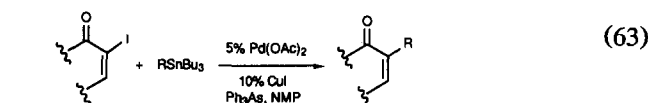
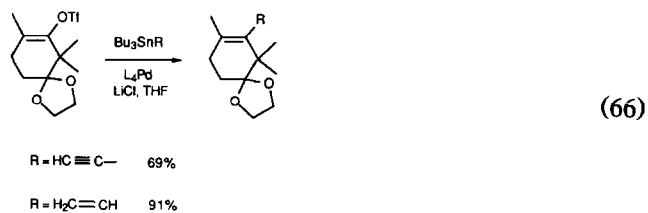
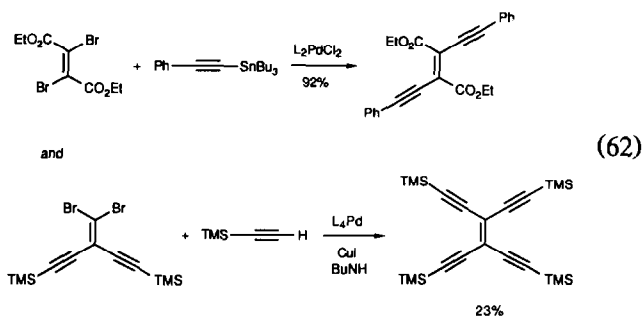


Vinyl stannanes coupled to bromoalkynes (eqn. (60) [67]), β -iodostyrenes (eqn. (61) [68]) and acetylenic stannanes coupled to vinyl bromide (eqn. (62) [69]). α -Iodoenones were alkylated by vinyl and allyl stannanes in the presence of palladium(0) catalysts (eqn. (63) [70]) as were α -halocyclobutenones (eqn. (64) [71]) and α -halo ethers (eqn. (65) [72]).

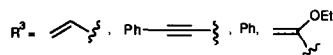
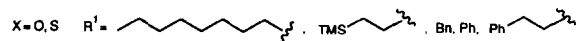
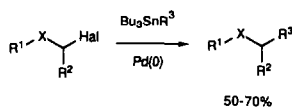


R = H, $\text{CH}(\text{OEt})_2$, $\text{CH}_2\text{CH}(\text{OEt})_2$, $(\text{CH}_3)_2$

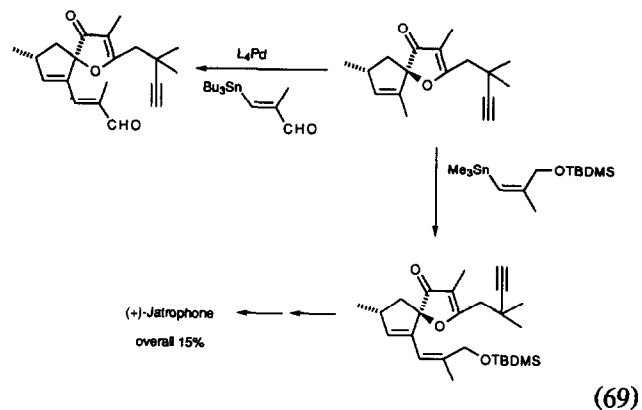
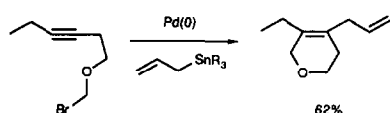
R' = $\text{CH}(\text{OEt})_2$, $\text{CH}_2\text{CH}(\text{OEt})_2$, Bu, Ph, CO_2Me



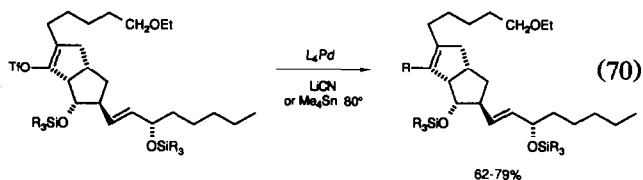
(65)

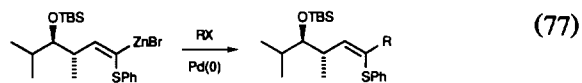
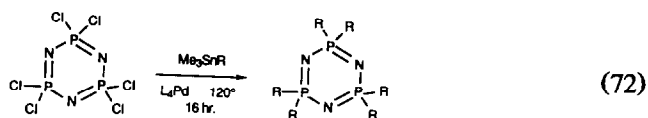
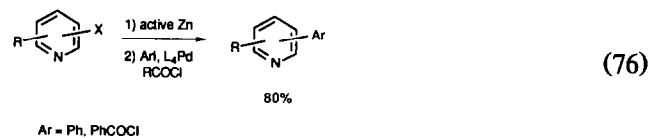
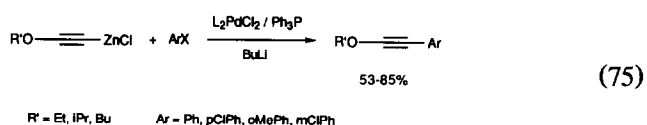
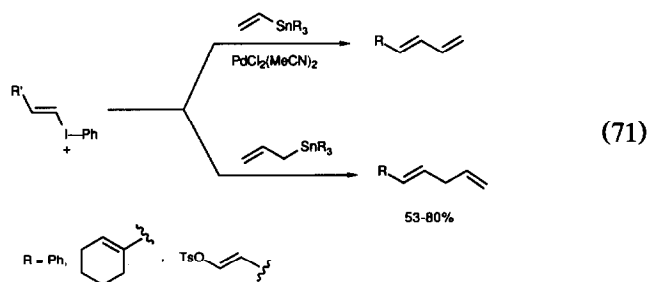


and



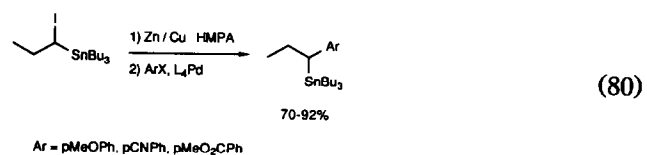
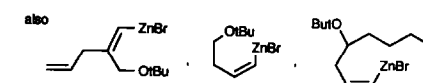
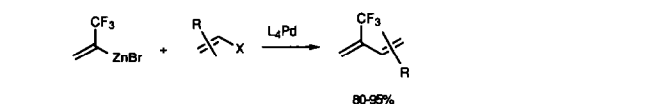
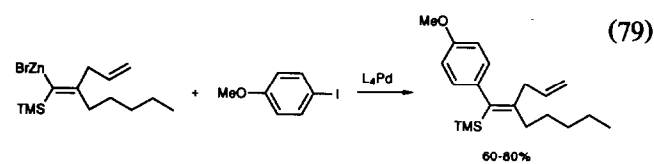
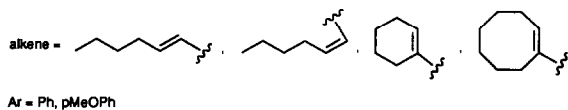
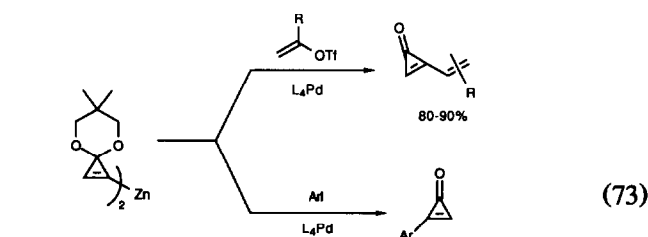
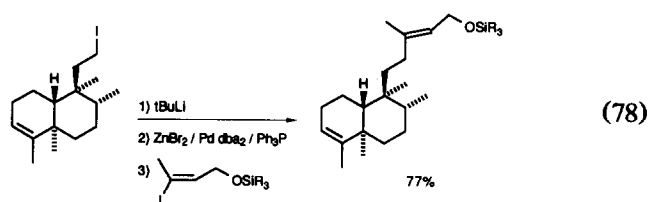
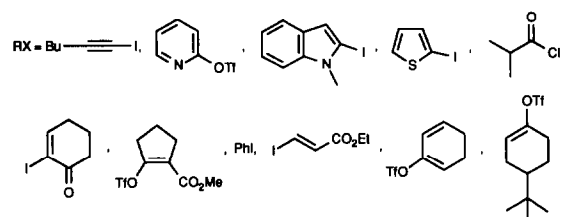
Palladium catalyzed tin coupling reactions have been used extensively in the synthesis of complex molecules (eqn. (66) [73], eqn. (67) [74], eqn. (68) [75], eqn. (69) [76], and eqn. (70) [77]).

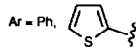
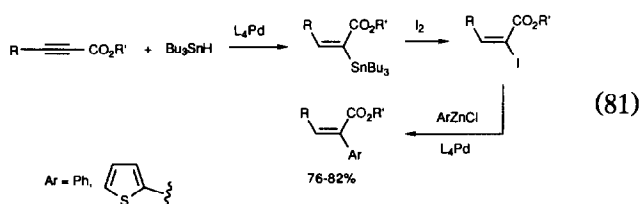




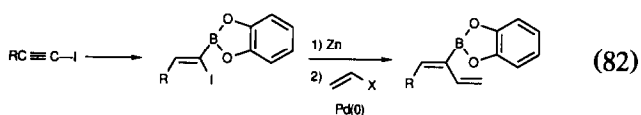
Other less conventional halides also underwent palladium catalyzed coupling with organostannanes (eqn. (71) [78] and eqn. (72) [79]).

Zinc also effectively transmetallates to palladium, and many palladium catalyzed couplings of organozinc reagents with triflates and halides have been developed (eqn. (73) [80], eqn. (74) [81], eqn. (75) [82], eqn. (76) [83], eqn. (77) [84], eqn. (78) [85], eqn. (79) [86], eqn. (80) [87], eqn. (81) [88], and eqn. (82) [89]).

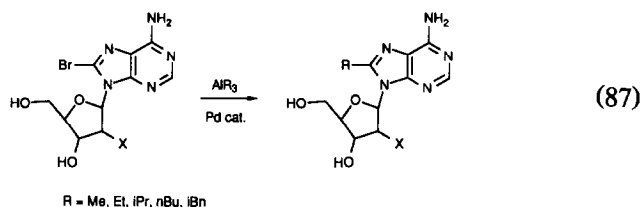
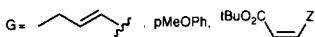
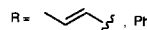
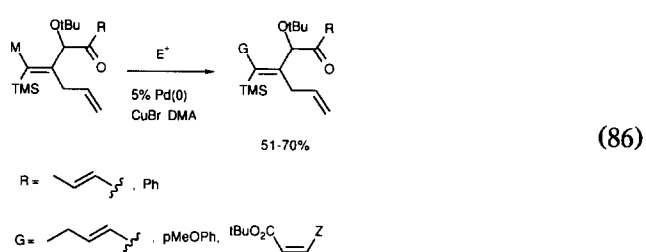
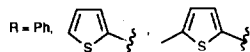
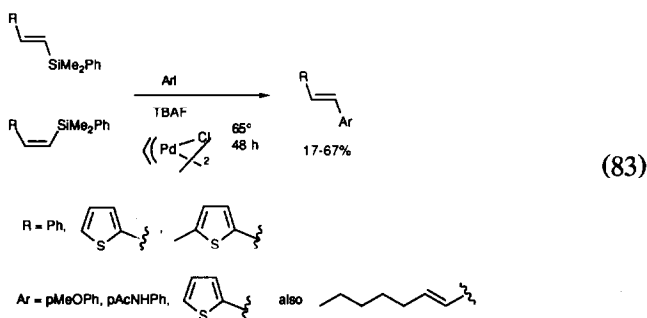




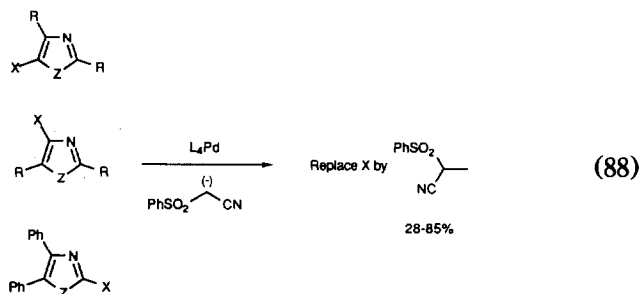
76-82%



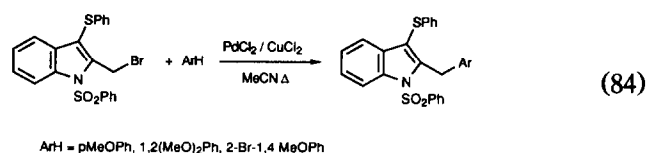
“Highly selective cross-coupling reactions of organosilicon compounds mediated by fluoride ion and a palladium catalyst” is the title of a recent review (48 references) [90]. Examples of this kind of reaction are shown in eqn. (83) [91].



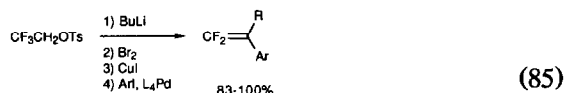
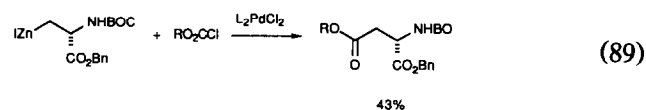
R = Me, Et, iPr, nBu, tBu



Palladium also catalyzed the coupling of halides to organocopper species (eqn. (84) [92], eqn. (85) [93], and eqn. (86) [94]), aluminum alkyls (eqn. (87) [95]) and to stabilized carbanions (eqn. (88) [96]).

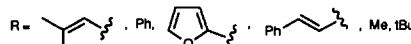
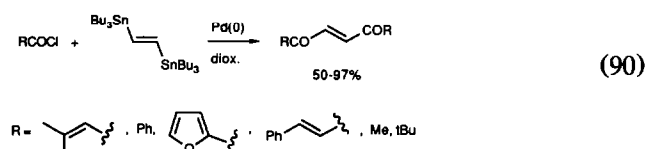


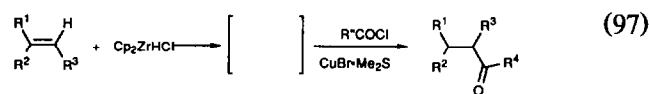
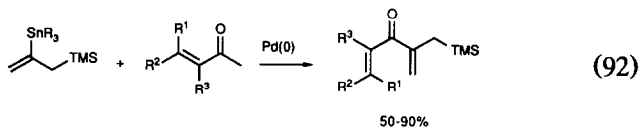
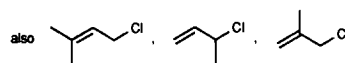
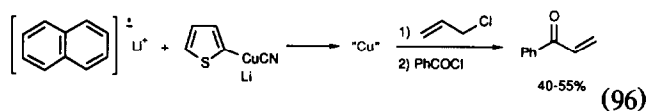
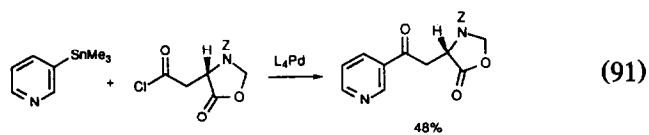
ArH = pMeOPh, 1,2(MeO)2Ph, 2-Br-1,4 MeOPh



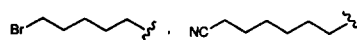
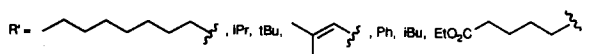
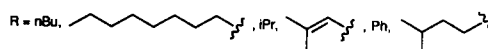
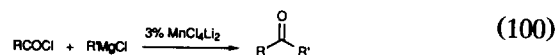
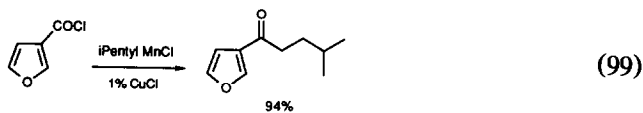
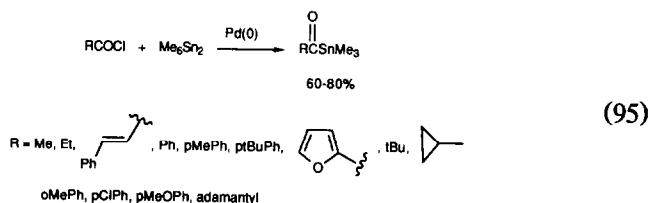
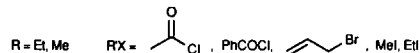
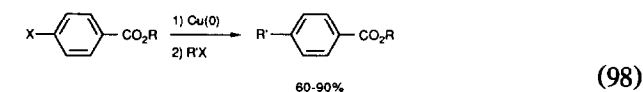
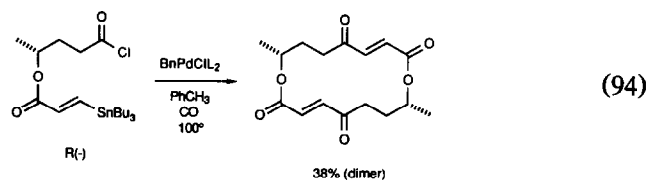
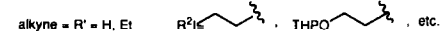
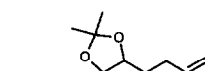
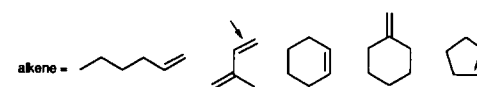
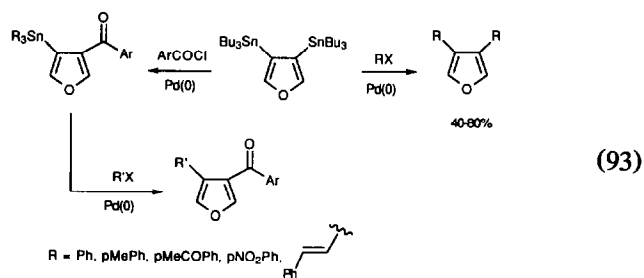
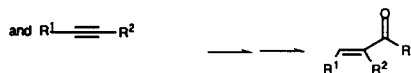
R = Bu, secBu

Ar = Ph, pNO2Ph, mNO2Ph, pAcPh, pEtO2CPh, pClPh, pMeOPh, 1-Naphthyl





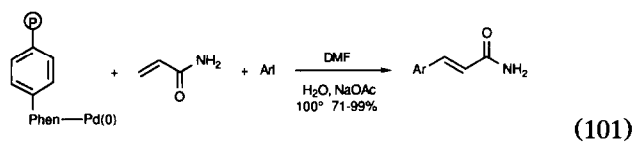
good yields



Organocopper complexes are waning in popularity for the conversion of acid halides to ketones, but a few cases have been reported (eqn. (96) [104], eqn. (97) [105], and eqn. (98) [106]). Organomanganese complexes also alkylate acid chlorides (eqn. (99) [107] and eqn. (100) [108]).

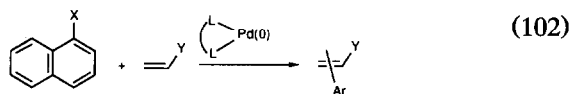
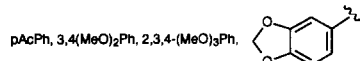
2.1.3. Alkylation of Olefins

The Heck reaction – palladium catalyzed oxidative addition/olefin insertion/ β -elimination – continued to be the most popular method to alkylate olefins, and many aspects of this reaction have been studied. Heck reactions in aqueous solutions were achieved by using sulfonated phosphines as ligands. The reactions went at mild temperatures in aqueous ethanol or acetonitrile, and gave high yields [109]. Polymer supported phenanthroline ligand/palladium complexes catalyzed the arylation of acrylamide (eqn. (101) [110]). Several other heterogeneous supports were also developed [111,112]. The effects of ligands on the regioselectivity of the arylation of a variety of alkenes by 1-naphthyl iodide or triflate were studied (eqn. (102) [113] and eqn. (103) [114]).



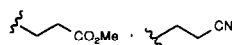
(101)

Ar = Ph, pClPh, pBrPh, oClPh, 2,6-Cl₂Ph, pNO₂Ph, pMePh, pMeOPh, mNO₂Ph, pCO₂HPh,

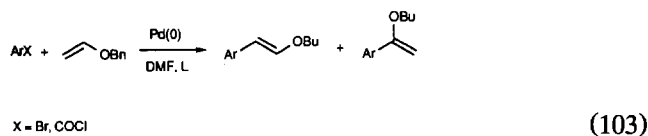


(102)

Y = OBn, , Cl₂OH, , , OH, , OAc, Ph, CO₂Me, CN, H, COMe,



L = DPPP, DMF, DPPB, DPPE



(103)

X = Br, COCl

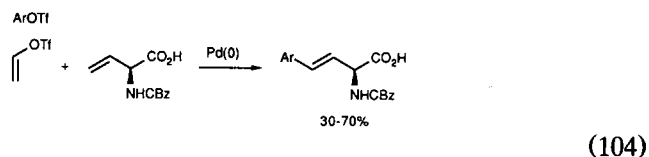
Ar = 1-Naphthyl, oMePh, oMeOPh, oCH₃COPh, oNCPh, oO₂NPh, oMeO₂CPh, mMePh, mMeOPh, mCH₃COPh, mCNPh, mO₂NPh, pMePh, pMeOPh, pCH₃COPh, pNCPh, pClPh, pO₂NPh

TfOAc favors α , Bidentate L favors α .

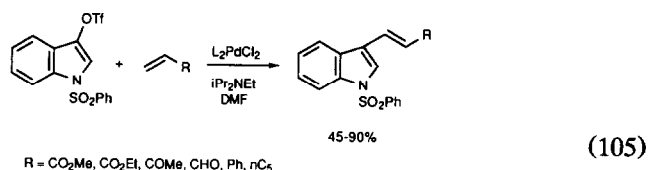
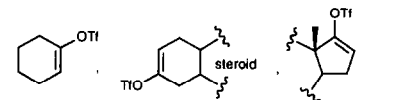
Catalyzed asymmetric Heck reactions have been reviewed (15 references) [115] and (24 references) [116].

Palladium(0) catalyzed the arylation and vinylation of vinyl glycine by triflates (eqn. (104) [117]), the vinylation of indole triflates (eqn. (105) [118]) and naphthyl

triflates (eqn. (106) [119]), and the arylation of acetamido acrylates (eqn. (107) [120]) and acrylamides (eqn. (108) [121]). Palladium(0) also catalyzed the vinylation of cyclopentene diols (eqn. (109) [122]) and the arylation of styrenes (eqn. (110) [123]).

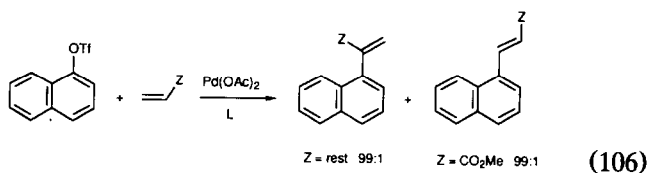
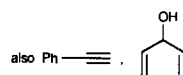


(104)



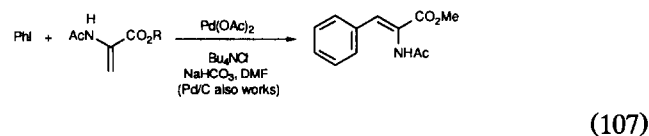
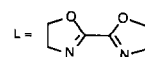
(105)

R = CO₂Me, CO₂Et, COMe, CHO, Ph, nC₅

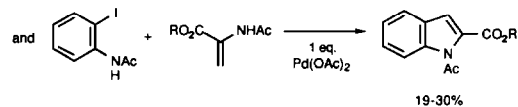


(106)

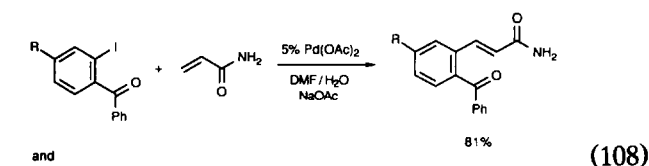
Z = OBn, Ph, , ,



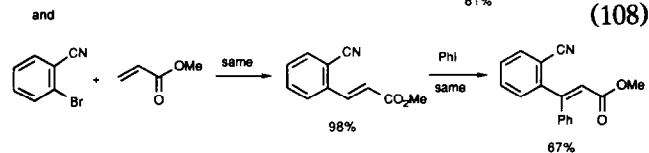
(107)



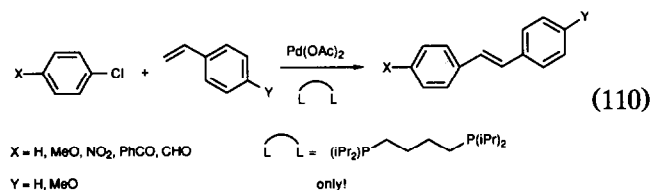
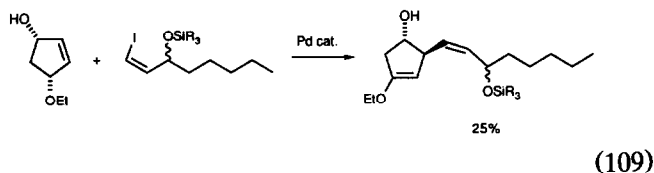
19-30%



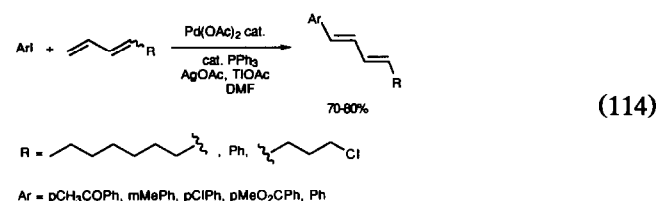
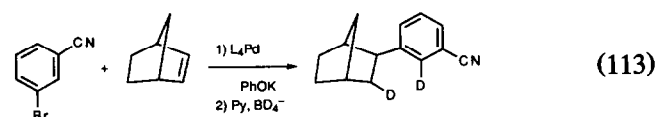
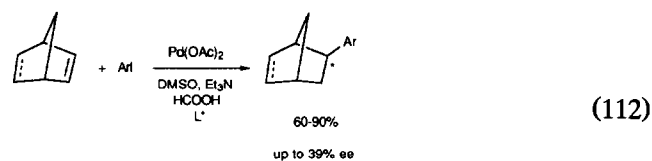
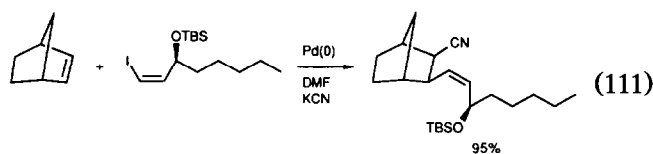
(108)



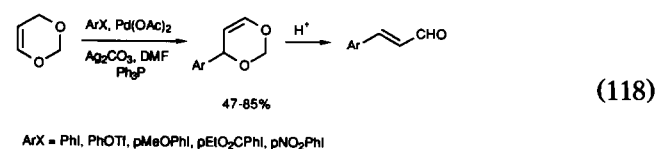
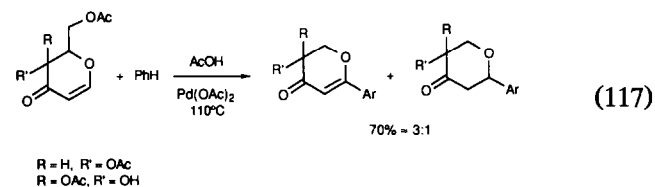
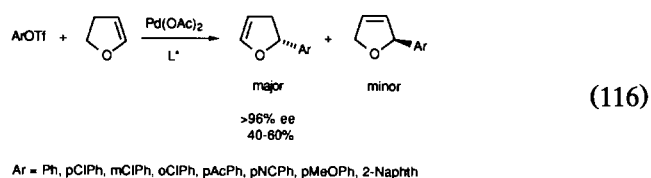
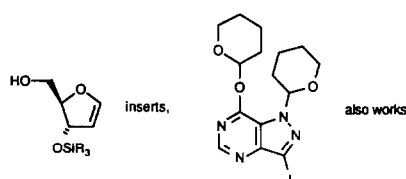
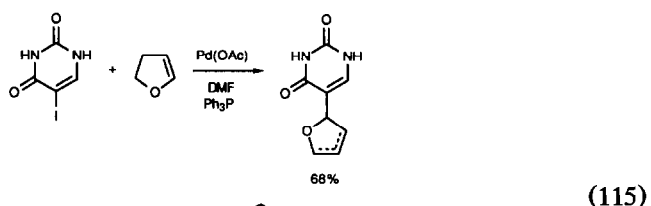
67%

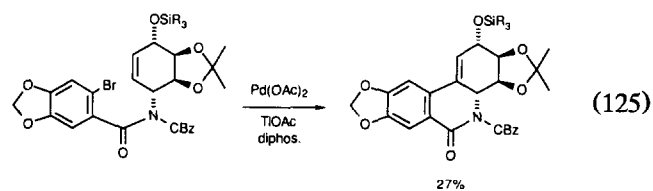
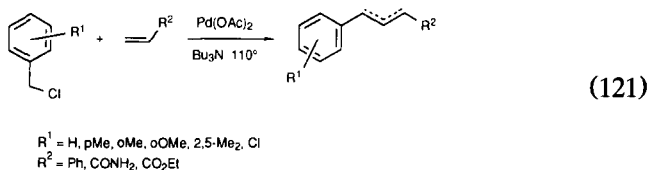
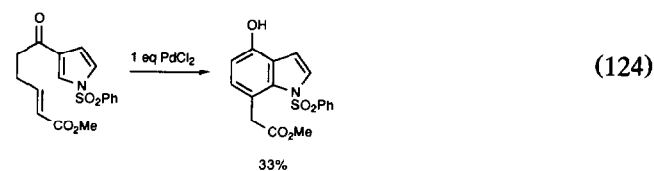
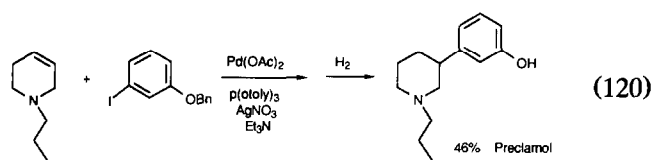


Norbornene (eqn. (111) [124], eqn. (112) [125], eqn. (113) [126]) and 1,3-dienes (eqn. (114) [127]) were alkylated under Heck reaction conditions.

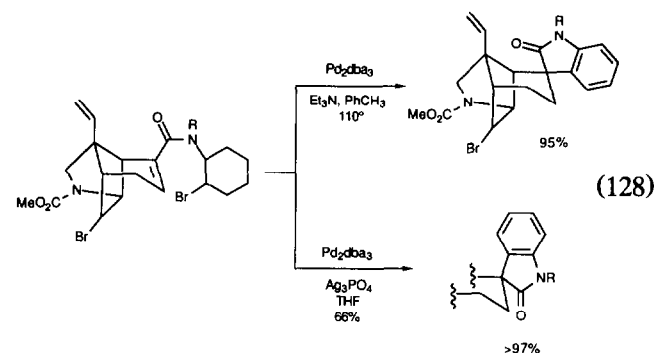
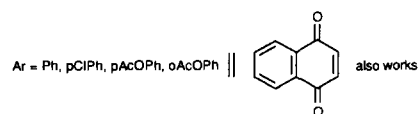
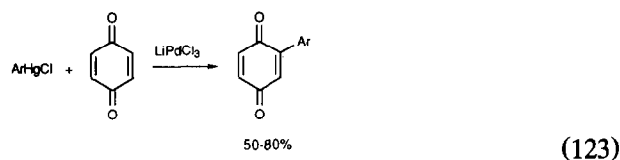
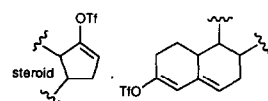
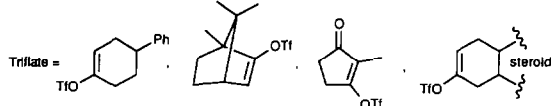
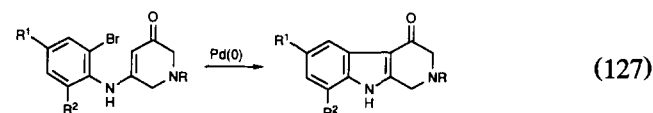
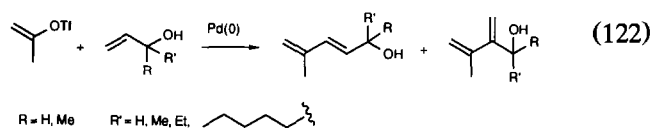
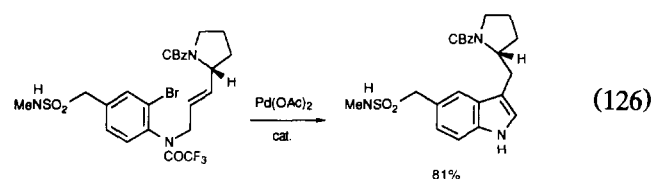


Oxygen (eqn. (115) [128], eqn. (116) [129], eqn. (117) [130], eqn. (118) [131]) and nitrogen (eqn. (119) [132], eqn. (120) [133]) heterocyclic alkenes were alkylated under Heck conditions. Benzyl chlorides also alkylated olefins under Heck conditions (eqn. (121) [134]).

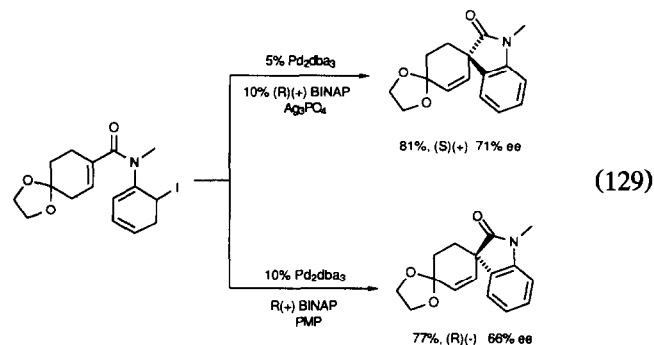


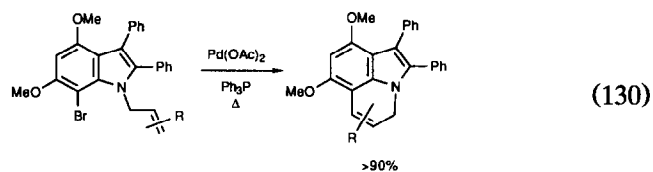


Tertiary allylic alcohols were alkylated by vinyl triflates under Heck conditions (eqn. (122) [135]). Quinones were arylated by aryl mercuric chlorides in the presence of palladium(II) catalysts (eqn. (123) [136]).

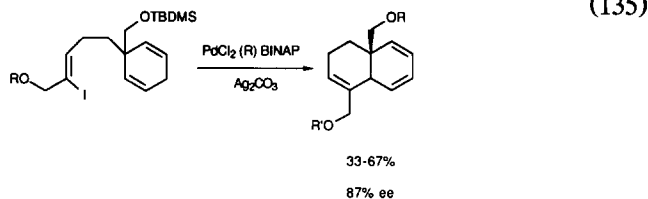
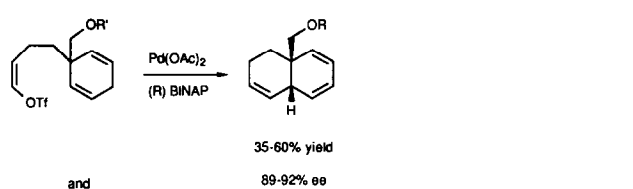
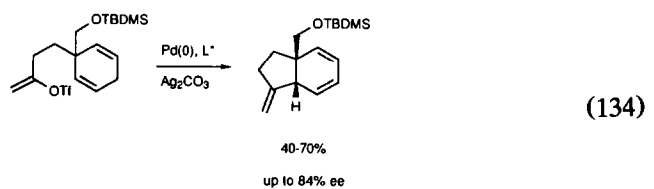
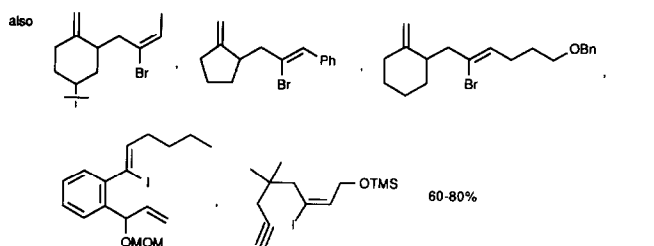
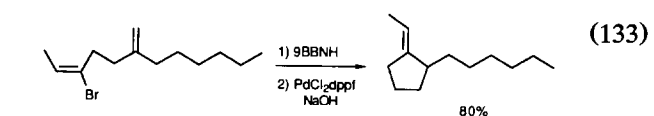
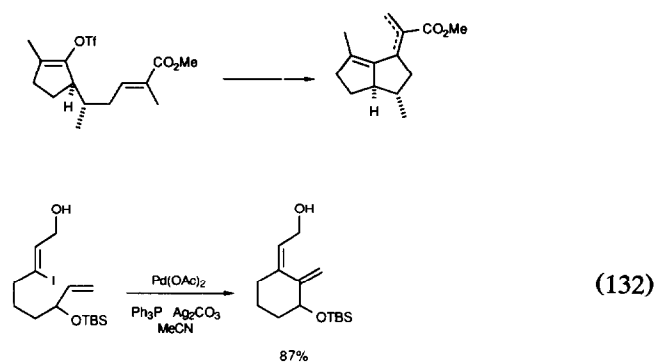
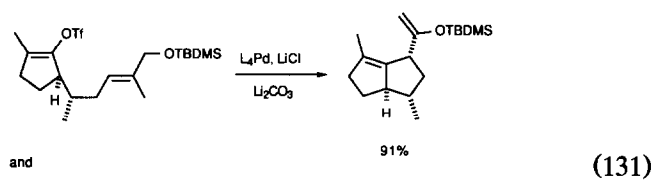


Intramolecular Heck reactions have been extensively used to form cyclic systems and have been the subject of several reviews [137] (23 references), [138] (25 references) [139]. It has proved very useful for the synthesis of a wide variety of nitrogen heterocycles (eqn. (124) [140], eqn. (125) [141], eqn. (126) [142], eqn. (127) [143], eqn. (128) [144], eqn. (129) [145], and eqn. (130) [146]).

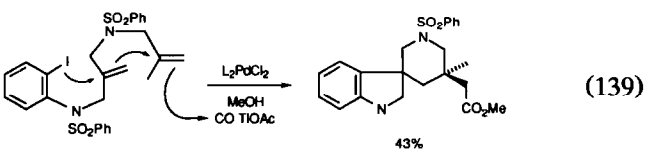
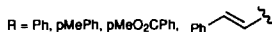
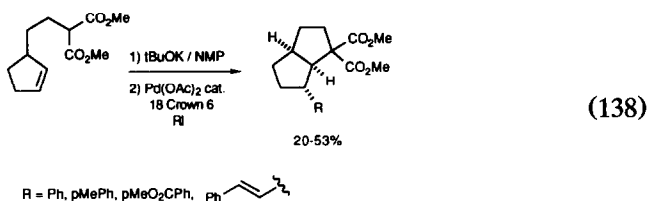
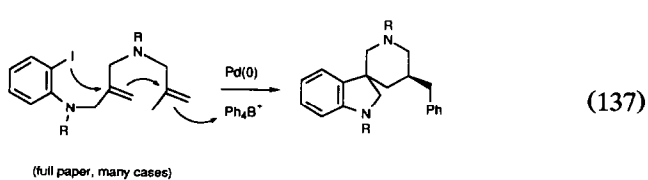
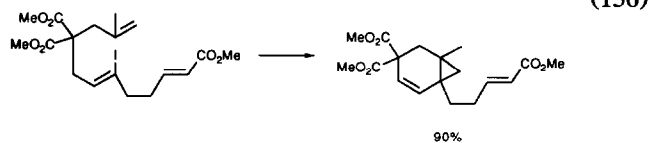
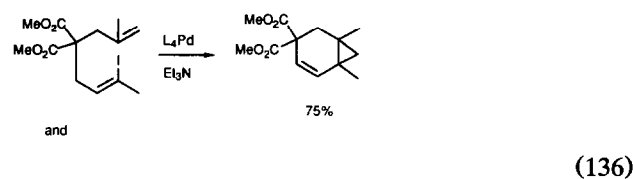




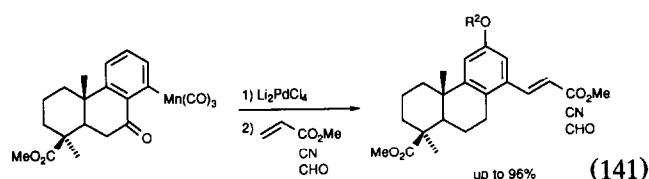
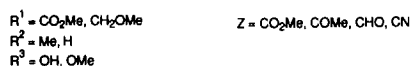
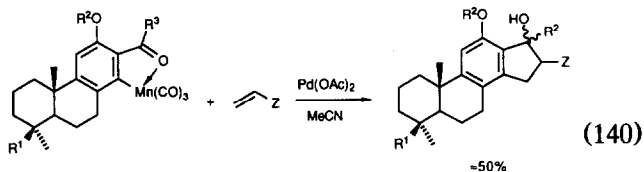
Carbocycles were also produced by intramolecular Heck alkylations of alkenes (eqn. (131) [147], eqn. (132) [148], eqn. (133) [149], eqn. (134) [150], and eqn. (135) [151]).



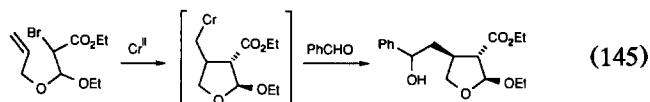
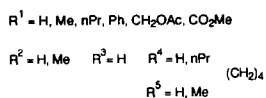
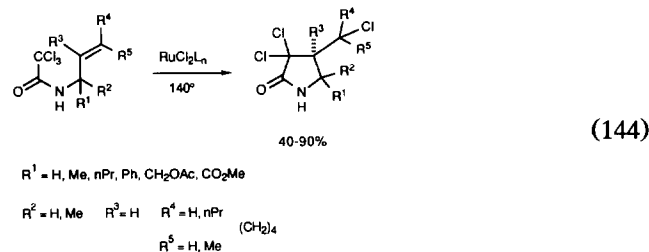
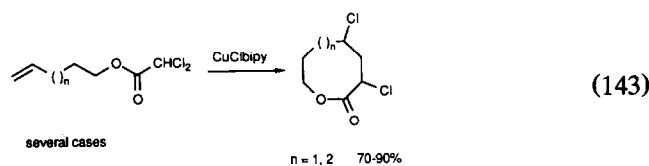
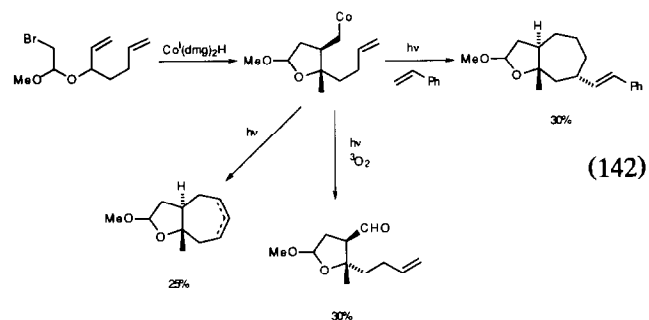
By intercepting the initial σ -alkylpalladium intermediate in the Heck reaction, multiple bonds can be formed, generating cascade cyclizations, a topic of a recent review (15 references) [152]. Examples of this process are seen in eqn. (136) [153], eqn. (137) [154], eqn. (138) [155], and eqn. (139) [156].



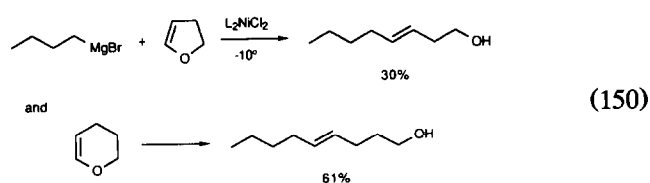
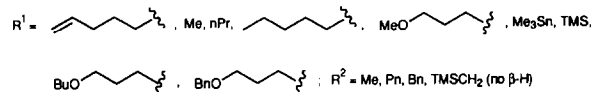
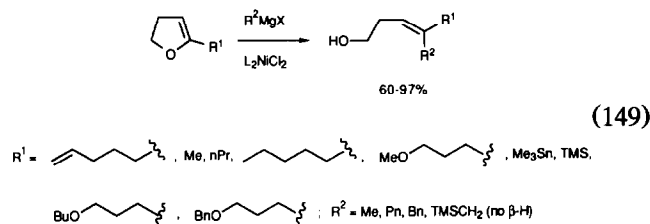
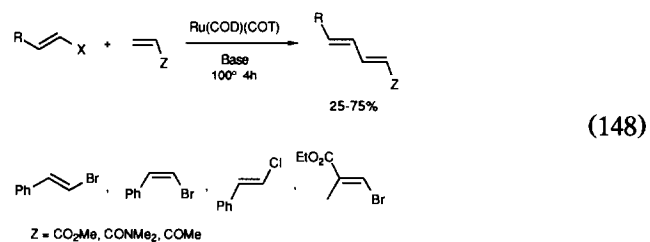
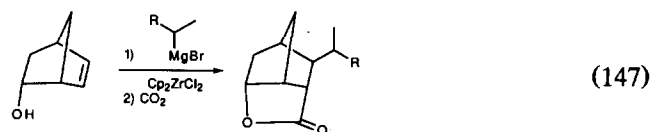
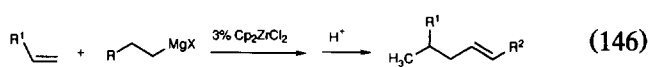
Ortho-manganated arenes underwent palladium catalyzed olefin insertion (eqn. (140) [157] and eqn. (141) [158]).

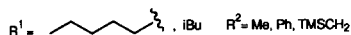
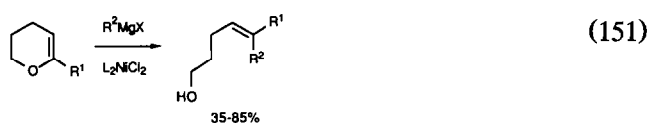


Alkenes were alkylated by free radical reactions initiated by cobalt(I) (eqn. (142) [159]), copper(I) (eqn. (143) [160]), ruthenium(II) (eqn. (144) [161]) or chromium(II) (eqn. (145) [162,163]).

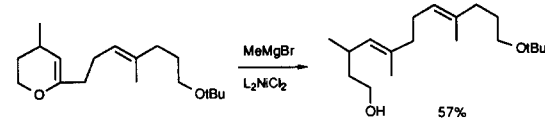


Zirconocene dichloride catalyzed the alkylation of olefins by Grignard reagents (eqn. (146) [164] and eqn. (147) [165]). Ruthenium complexes catalyzed the alkylation of alkenes by vinyl halides (eqn. (148) [166]). Nickel(II) phosphine complexes catalyzed the alkylation of dihydrofurans (eqn. (149) [167], eqn. (150) [168]) and dihydropyrans (eqn. (151) [169]).

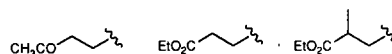
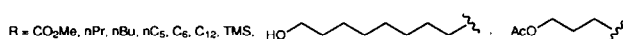
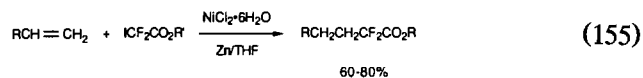
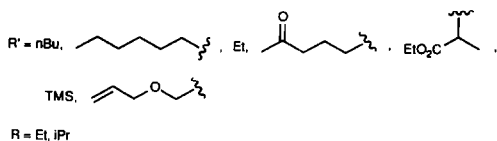
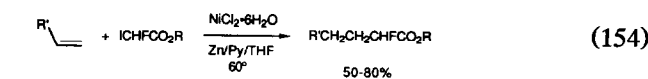
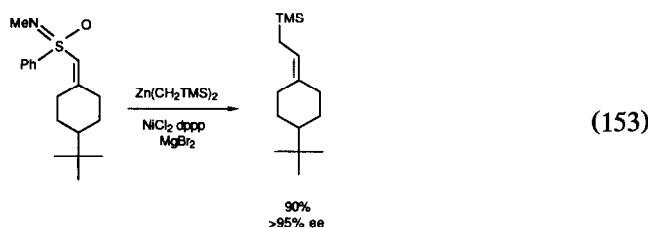
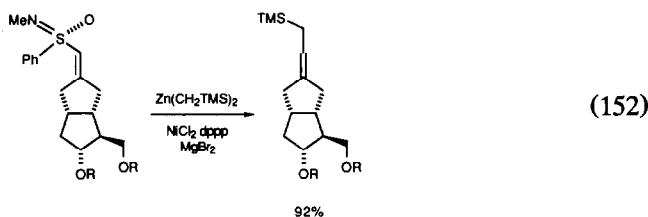




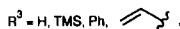
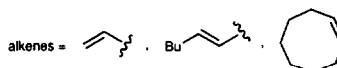
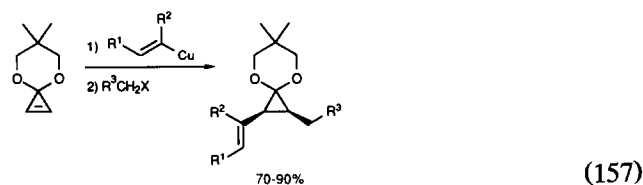
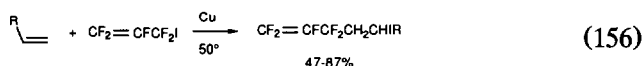
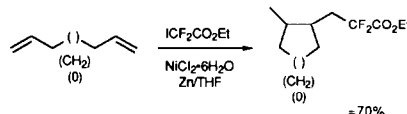
and



Nickel(II) phosphine complexes catalyzed the alkylation of vinyl sulfinamides by organozinc reagents (eqn. (152) [170] and eqn. (153) [171]). Nickel(0) complexes catalyzed the alkylation of olefins by α -iodoesters (eqn. (154) [172] and eqn. (155) [173]). Copper catalyzed the alkylation of olefins with perfluoroallyl iodides (eqn. (156) [174]) and cyclopropene acetals by halides (eqn. (157) [175]).

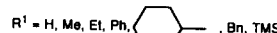
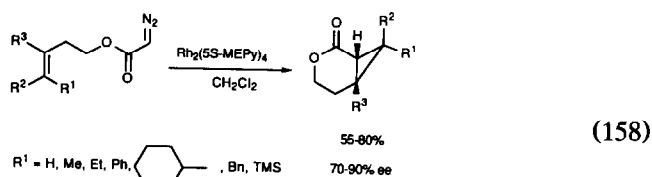


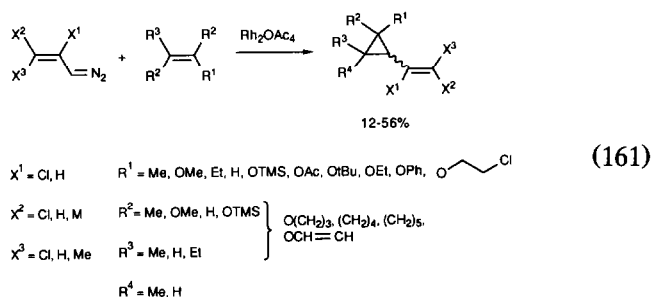
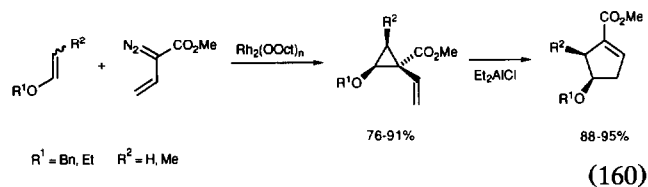
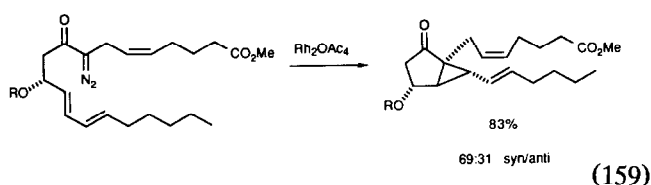
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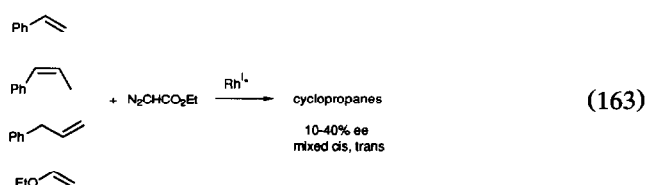
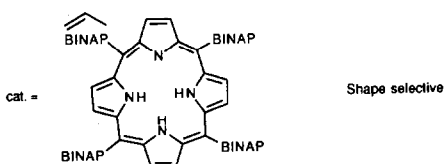
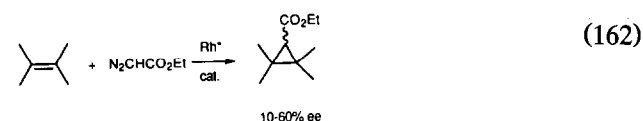
2.1.4. Decomposition of diazoalkanes and other cyclopropanation reactions

Metal catalyzed decomposition of diazoalkanes is another fairly standard class of organometallic reaction that has been "discovered" by organic chemists and has been explored with a vengeance. Perhaps the best-known process is the Rh(II) catalyzed cyclopropanation of alkenes by α -diazoketones and esters. The chiral rhodium(II) catalysis "MEPyH"₄Rh₂ has been bound to soluble polyethylene to aid in its recovery from completed cyclopropanation reactions [176]. Rhodium(II) catalyzed intramolecular cyclopropanation was an efficient process (eqn. (158) [177] and eqn. (159) [178]). Cyclopentenes were prepared by rhodium(II) catalyzed cyclopropanations followed by ring expansion (eqn. (160) [179]). Vinyl cyclopropanes were made by rhodium(II) catalyzed decomposition of vinyl diazo compounds (eqn. (161) [180]).

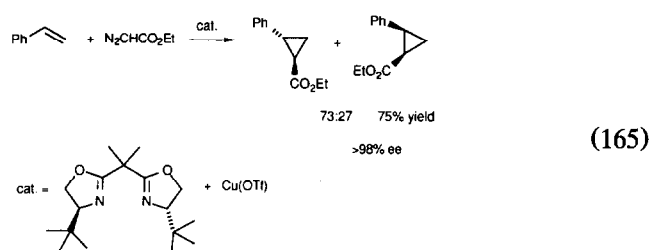
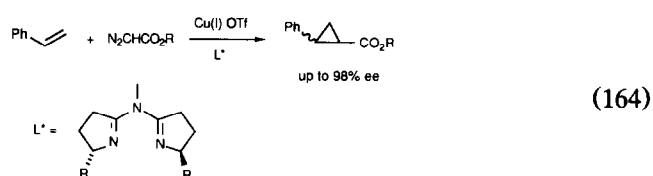




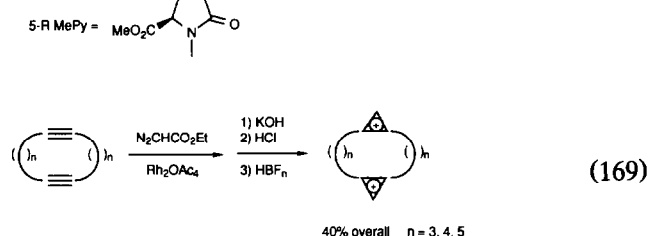
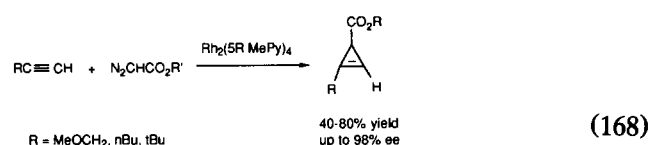
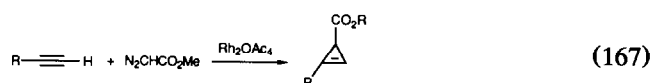
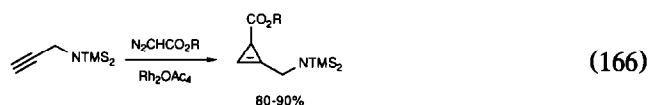
The mechanism of rhodium–porphyrin catalyzed cyclopropanation of alkenes has been studied [181]. Chiral BINAP–porphyrin rhodium complexes catalyzed olefin cyclopropanation with modest ee (eqn. (162) [182] and eqn. (163) [183]).



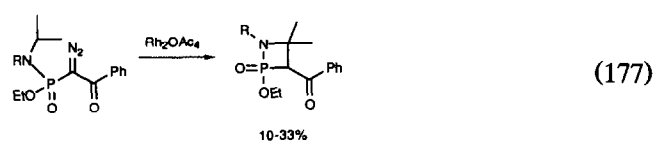
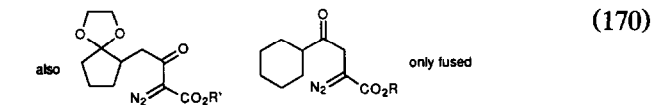
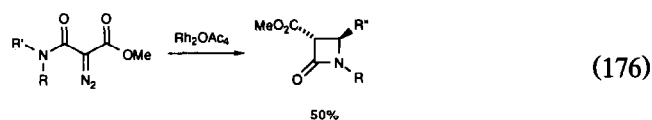
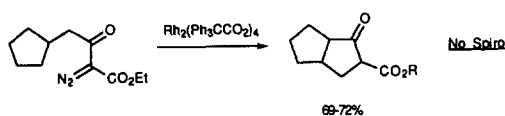
Chiral copper complexes also catalyzed the asymmetric cyclopropanation of alkenes (eqn. (164) [184] and eqn. (165) [185,186]).



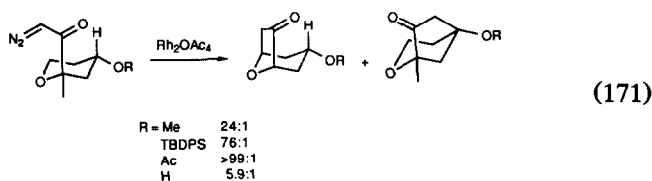
The cyclopropanation of alkenes by diazoesters was also catalyzed by ruthenium carborane clusters [187], copper(II) acetylacetonate [188], palladium(II) acetate [189] and copper salen complexes [190]. Rhodium(II) complexes also catalyzed the cyclopropanation of alkynes by diazoalkanes (eqn. (166) [191], eqn. (167) [192], eqn. (168) [193], and eqn. (169) [194]).



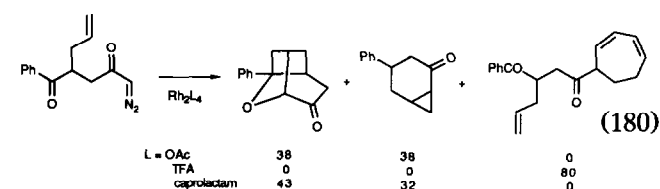
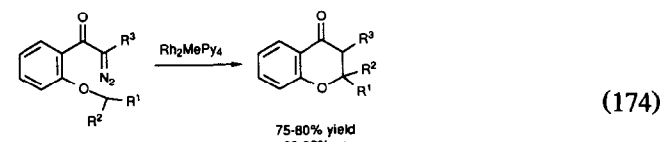
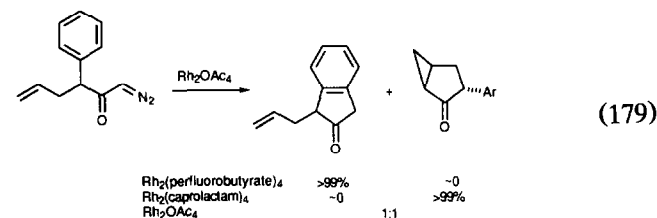
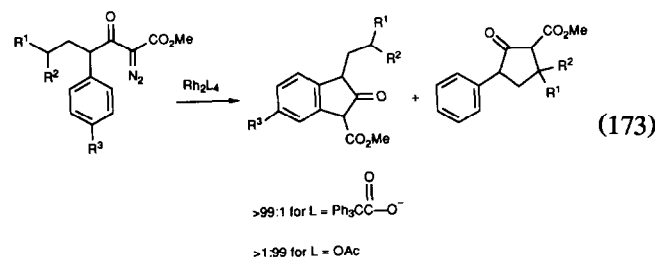
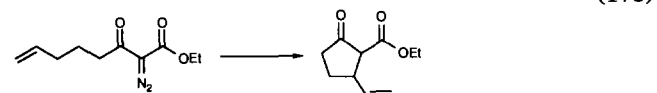
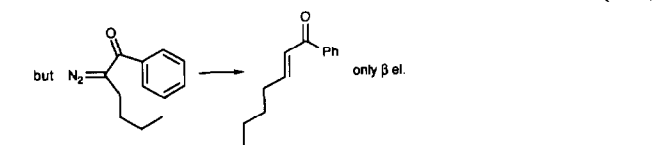
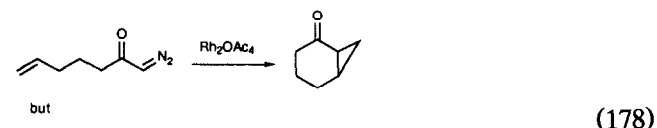
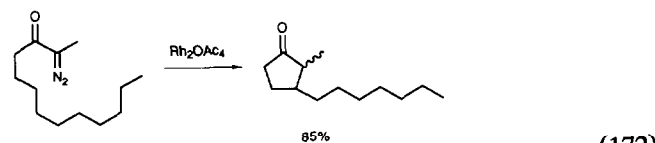
Rhodium(II) complexes also catalyzed C–H insertion reaction of diazoesters, but regiochemical predictions were difficult and strongly dependent on the parameters of the reaction (eqn. (170) [195], eqn. (171) [196], eqn. (172) [197], and eqn. (173) [198]). However, many reactions were straightforward (eqn. (174) [199], eqn. (175) [200], eqn. (176) [201], and eqn. (177) [202]).



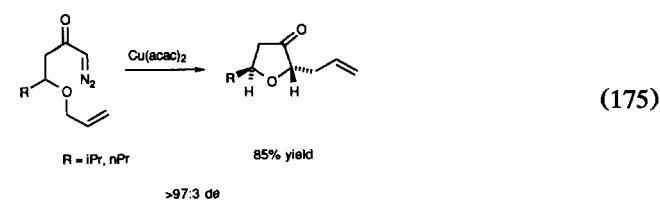
$\text{Rh}_2\text{OAc}_4 \Rightarrow$ Mainly Spiro



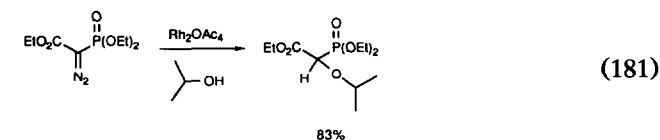
In substrates that can partition between cyclopropanation and CH insertion, several parameters were observed to effect the outcome of the reaction. In eqn. (178) [203] diazoketones preferred to cyclopropanate whereas diazoesters underwent CH insertion. This partitioning was also influenced by the nature of the catalyst (eqn. (179) [204] and eqn. (180) [205]).



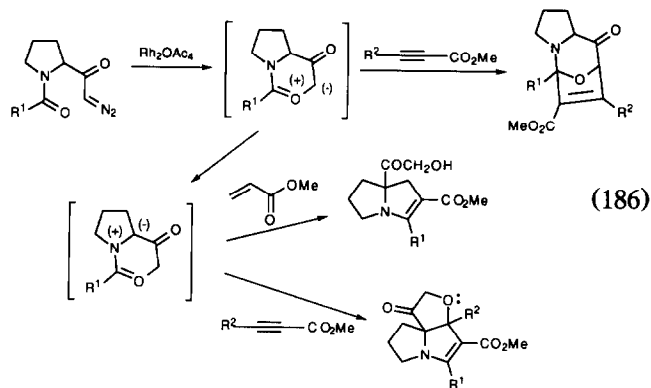
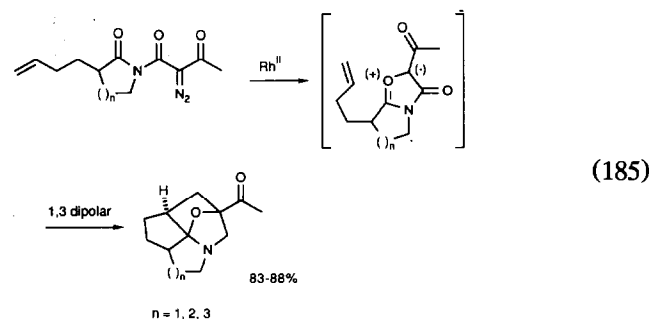
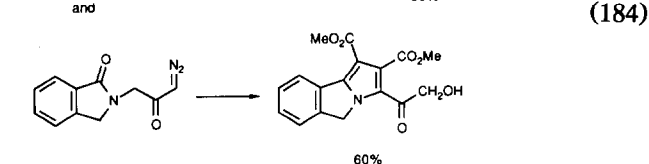
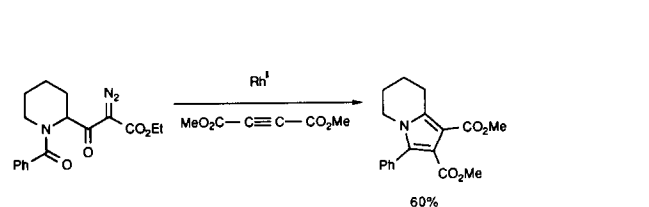
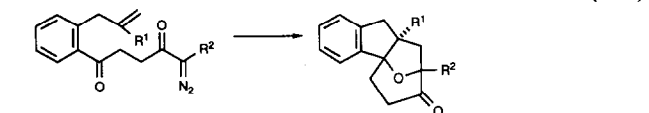
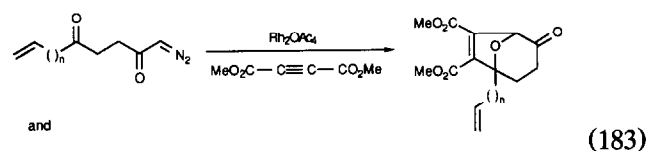
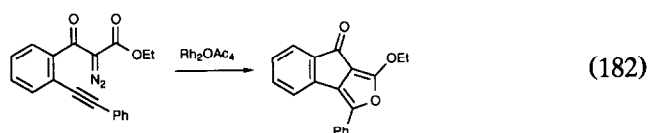
$\text{R}^1 = \text{H, Me}$ $\text{R}^2 = \text{Me, } \text{CH}_2=\text{CH}_2$ $\text{R}^3 = \text{Me, Ph}$



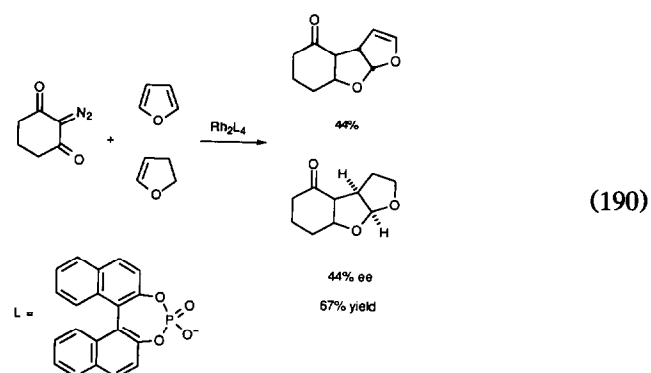
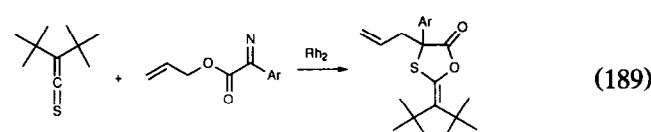
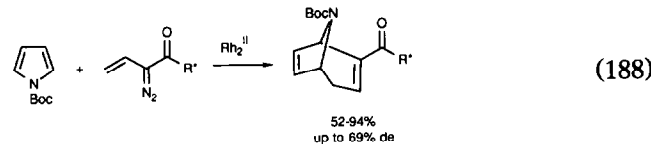
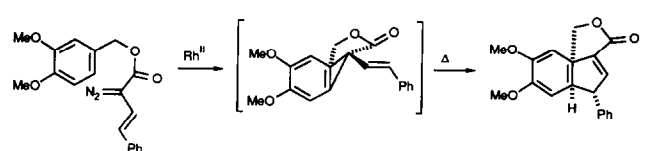
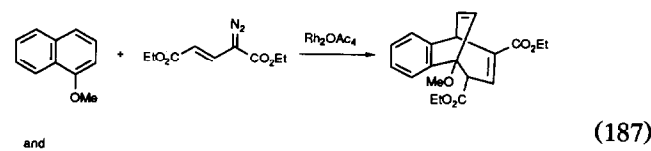
(Binaphthol phosphate)rhodium(II) complexes were efficient catalysts for CH insertion reactions for decomposition of diazo compounds, but enantioselectivities were low [206]. Rhodium(II) acetate catalyzed O-H insertion reactions of diazoketones, as well (eqn. (181) [207]).



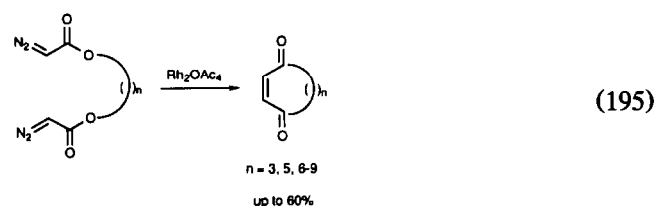
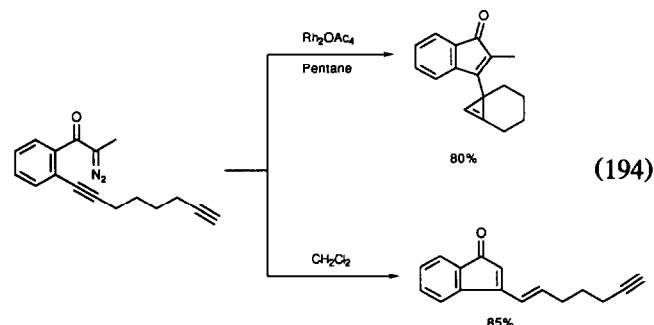
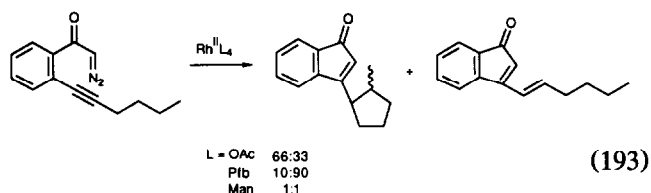
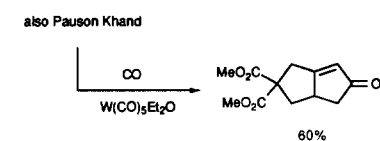
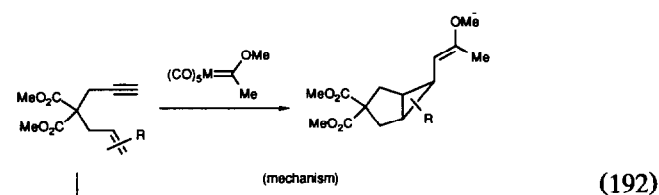
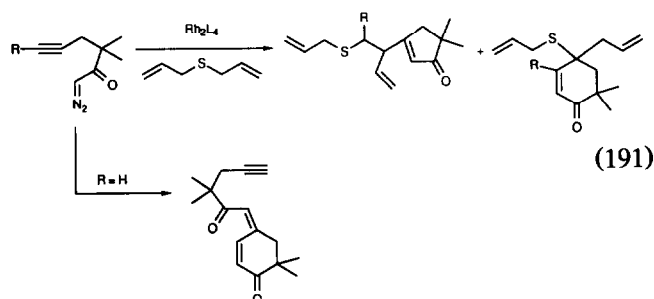
“Application of intramolecular carbenoid reactions in organic synthesis” was the title of a review (228 references) [208]. Rhodium(II) complexes catalyzed a bewildering array of cycloaddition reactions based on the formation of ylides by addition of heteroatoms to carbenoids generated by diazodecomposition reactions (eqn. (182) [209], eqn. (183) [210], eqn. (184) [211], eqn. (185) [212], and eqn. (186) [213]).



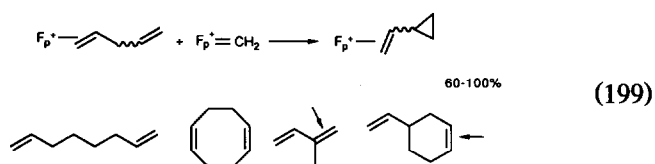
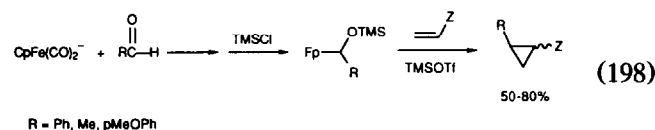
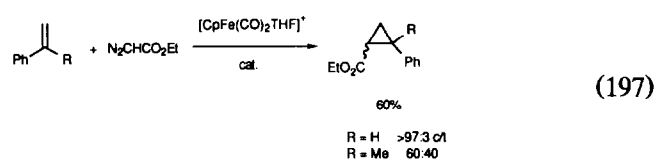
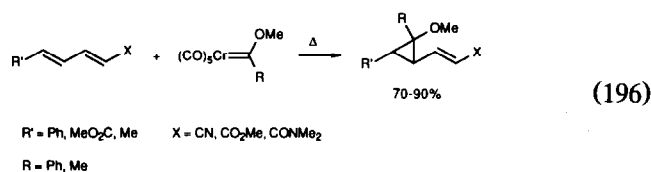
Rhodium(II) complexes catalyzed a number of unusual cycloaddition reactions of diazoalkanes (eqn. (187) [214], eqn. (188) [215], eqn. (189) [216], and eqn. (190) [217]).



Rhodium(II) complexes catalyzed a number of unusual "cascade" processes which involved metal carbenes as intermediates (eqn. (191) [218], eqn. (192) [219], eqn. (193) [220], and eqn. (194) [221]). The same complexes catalyzed the intramolecular dimerization of carbenoids (eqn. (195) [222]).

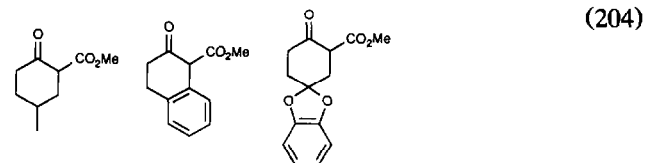
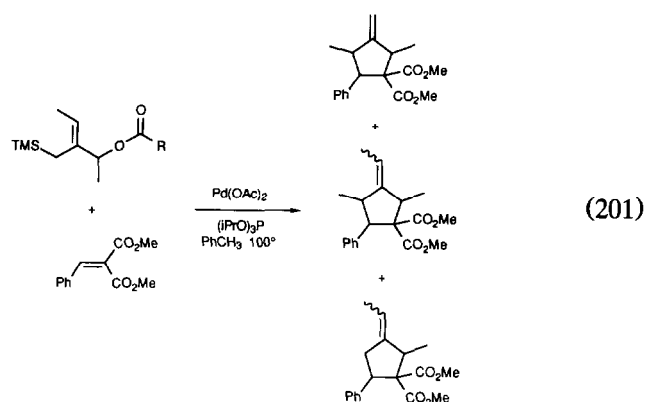
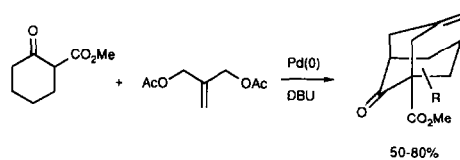
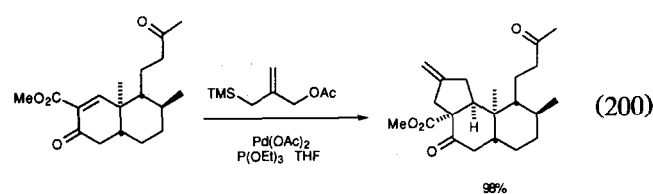


Electron poor dienes were cyclopropanated on the remote alkene by chromium carbene complexes (eqn. (196) [223]). Cationic iron complexes catalyzed the cyclopropanation of alkenes by diazoesters (eqn. (197) [224]). A route to iron carbene complexes, for use in cyclopropanation, from aldehydes has been developed (eqn. (198) [225]). Dienes were monocyclopropanated by iron carbene complexes, by protecting one alkene as the iron complex (eqn. (199) [226]). Nickel(0) catalyzed the cyclopropanation of electron deficient alkenes by gem-dihalo-alkanes [227].

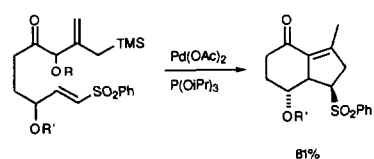


2.1.5. Cycloaddition reactions

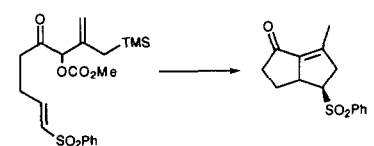
Cycloaddition reactions using transition metal trimethylene methane derivatives have been reviewed (6 references) [228]. A convenient synthesis of the requisite *Z*-2-trimethylsilylmethyl-2-butene-1-ol starting material for palladium catalyzed 3 + 2 cycloadditions has been reported [229]. This process was useful for the synthesis of a variety of complex products (eqn. (200) [230], eqn. (201) [231], and eqn. (202) [232]). By changing conditions, the cycloaddition to conjugated enones can be diverted from addition to the alkene to addition to the carbonyl group (eqn. (203) [233]). Allylic diacetates added to both α -positions of cyclic keto esters, giving bridged bicyclic intermediates (eqn. (204) [234]).



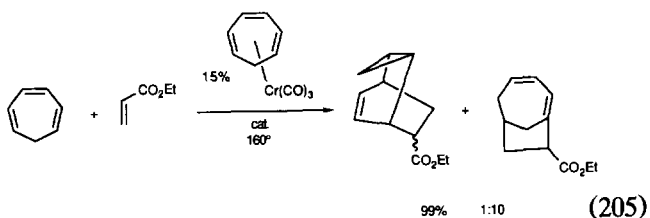
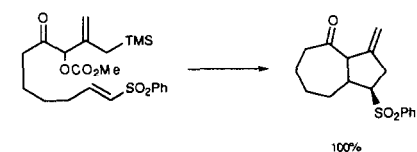
Chromium cycloheptatriene complexes catalyzed a number of [2 + 6] and [4 + 6] cycloadditions (eqn. (205) [235], eqn. (206) [236], and eqn. (207) [237]). Reduced cobalt complexes catalyzed an intramolecular homo Diels Alder reaction (eqn. (208) [238]). Ruthenium hydrides catalyzed [2 + 2] and other unusual cycloadditions (eqn. (209) [239]). Reduced titanium species catalyzed [4 + 2] and [6 + 2] cycloadditions (eqn. (210) [240]). Oxidation of cyclobutadiene iron complexes in the presence of norbornadiene led to several cycloadducts (eqn. (211) [241]). Polysubstituted phenols were synthesized from cyclobutenones *via* cobalt vinyl ketene intermediates (eqn. (212) [242]).



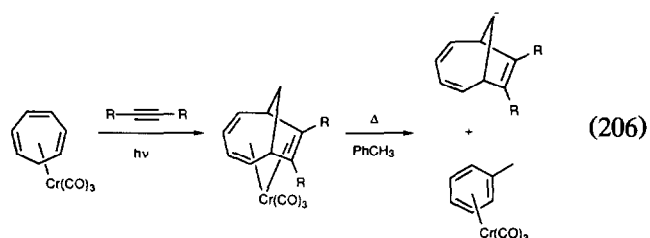
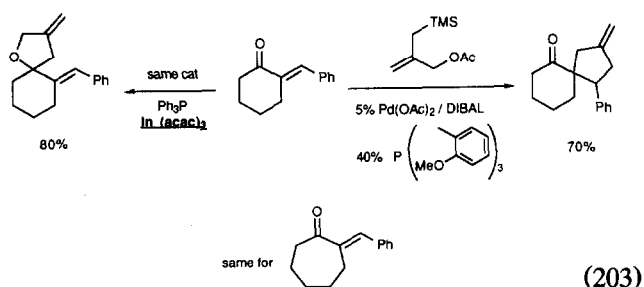
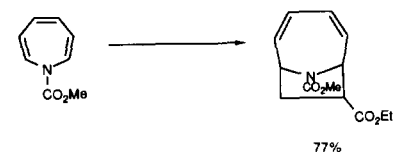
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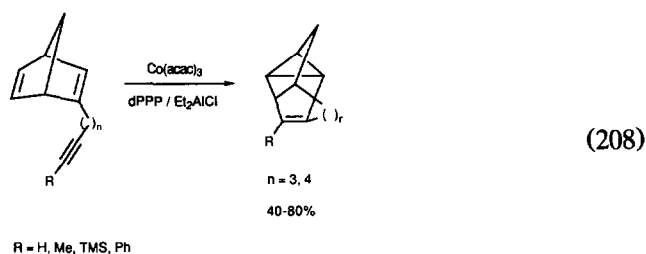
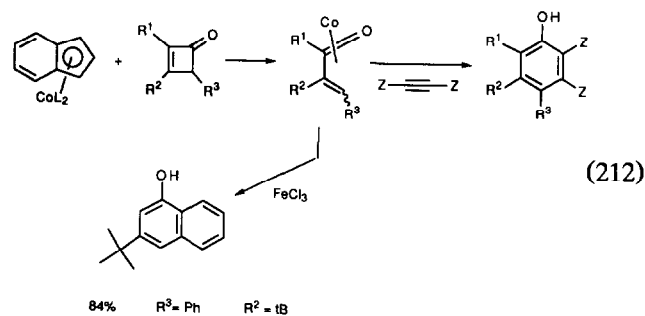
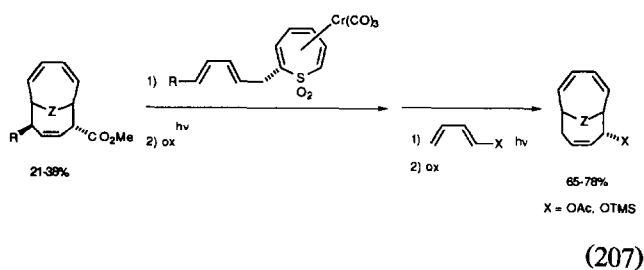


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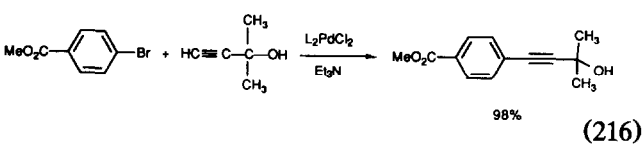
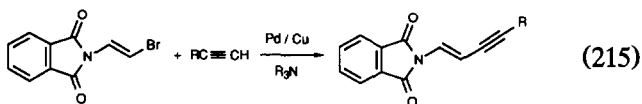
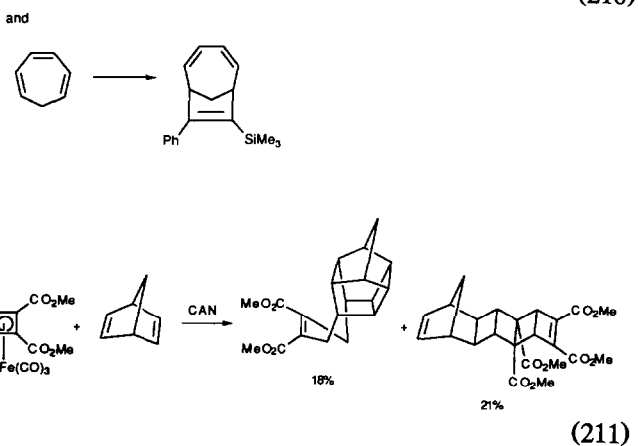
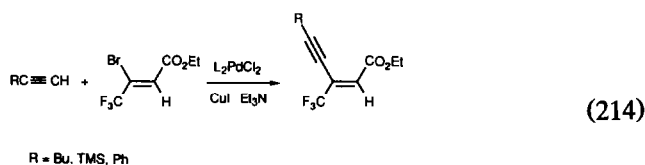
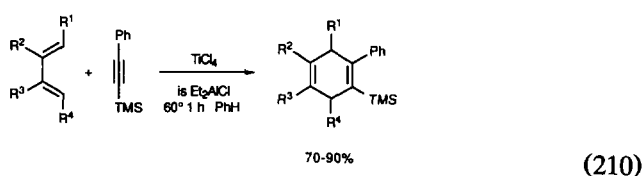
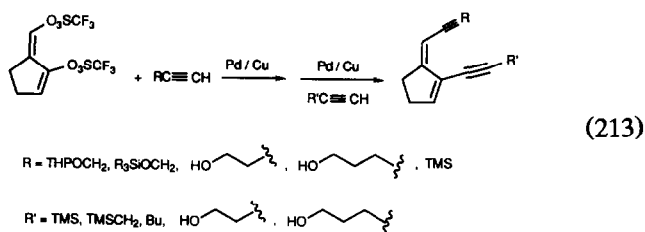
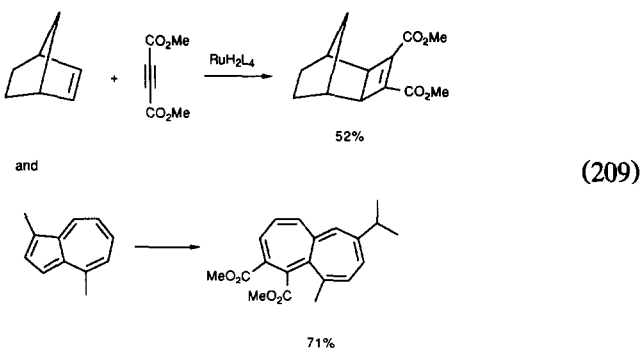
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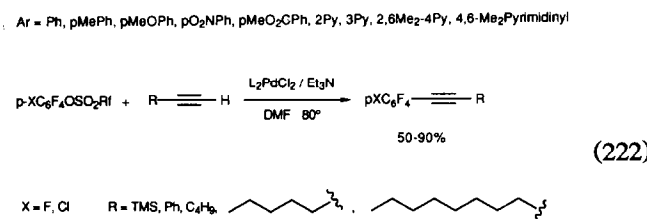
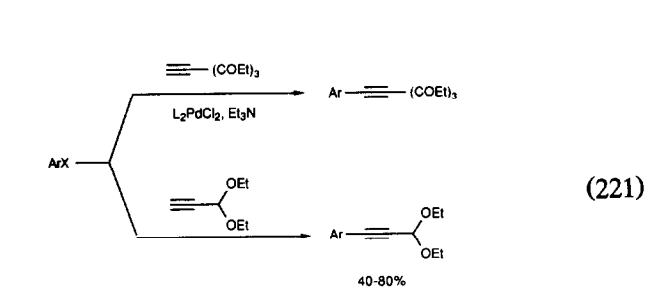
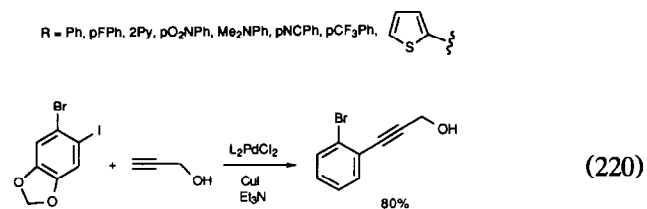
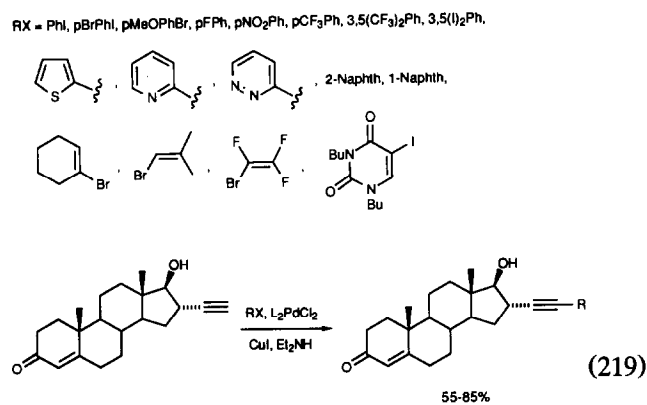
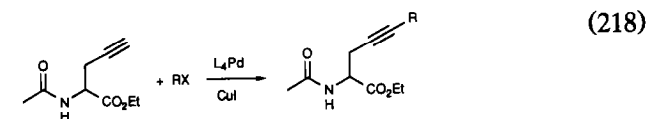
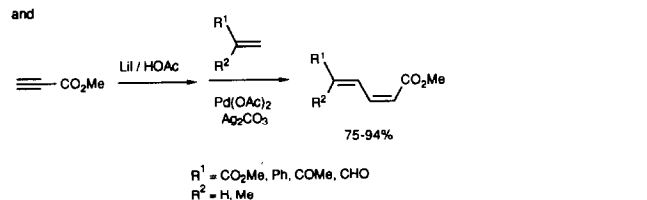
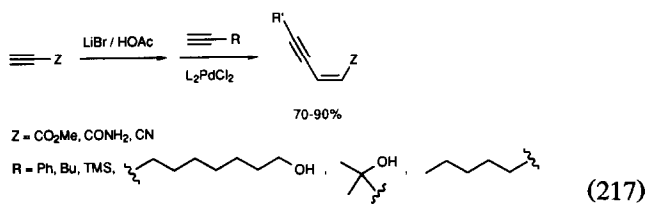




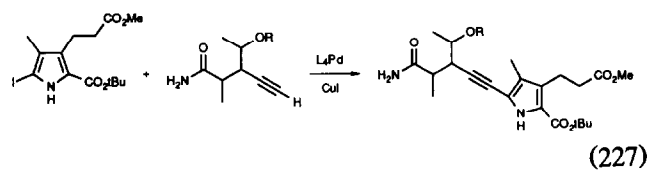
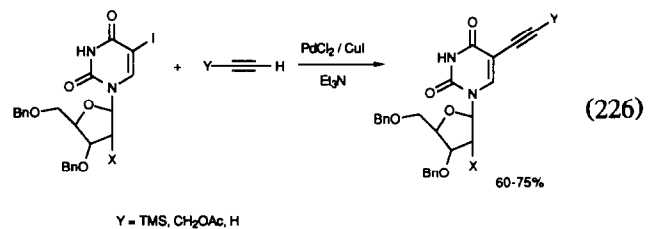
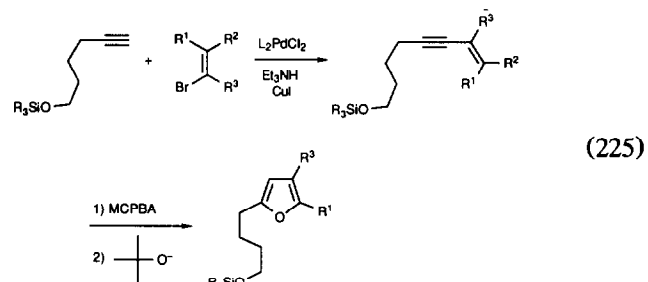
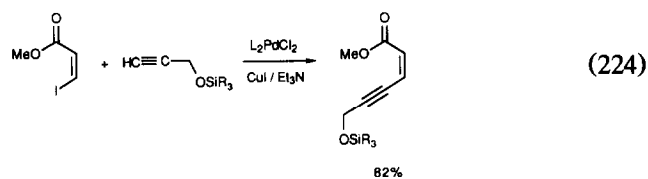
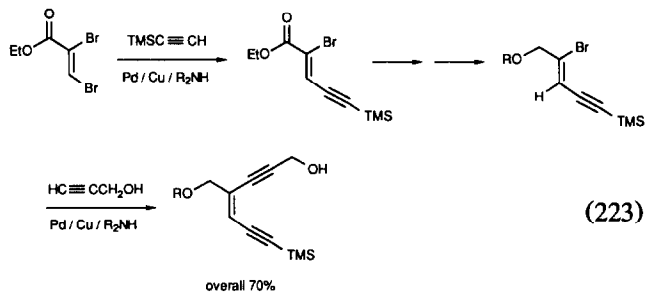
2.1.6. Alkylation of alkynes

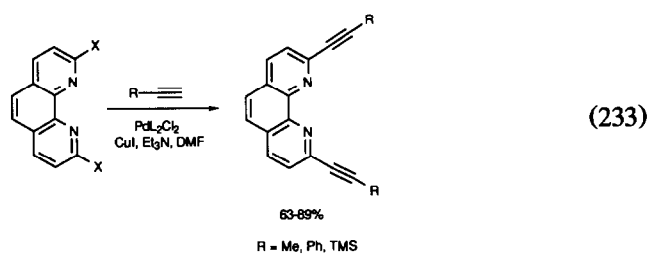
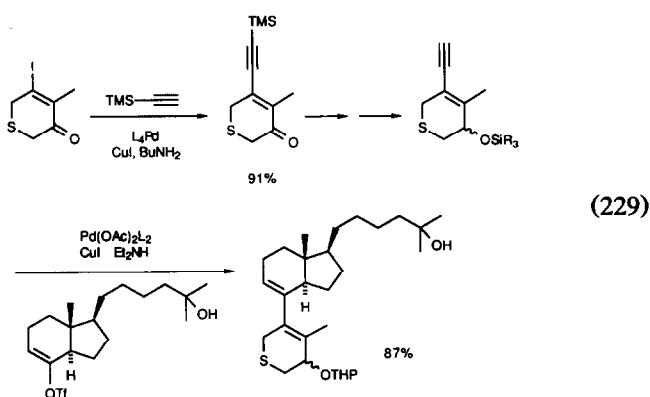
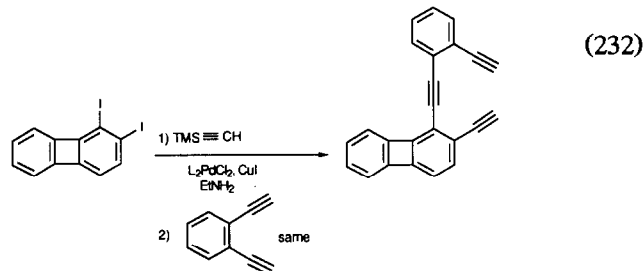
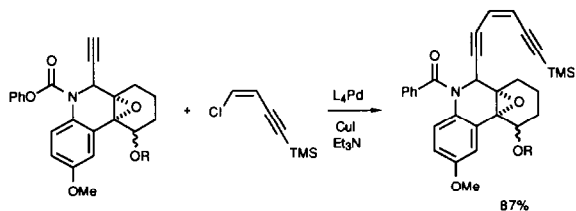
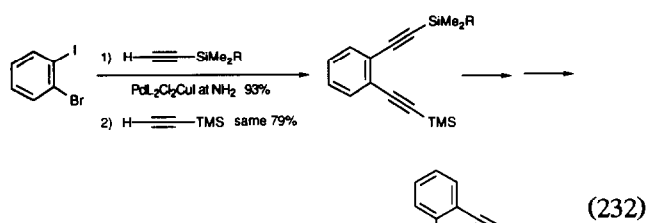
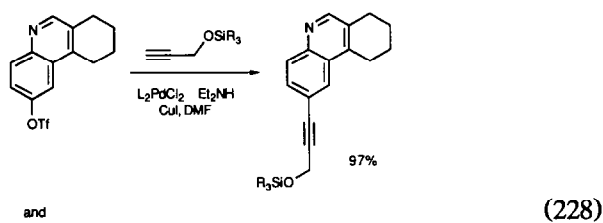
The Castro–Stevens reaction (Pd/Cu/amine) has become the premier way to couple alkynes to aryl and vinyl halides and triflates, and a large number of simple cases have been reported (eqn. (213) [243], eqn. (214) [244], eqn. (215) [245], eqn. (216) [246], eqn. (217) [247], eqn. (218) [248], eqn. (219) [249], eqn. (220) [250], eqn. (221) [251], and eqn. (222) [252]).



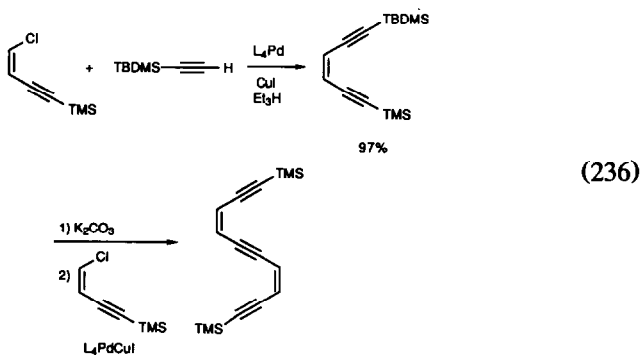
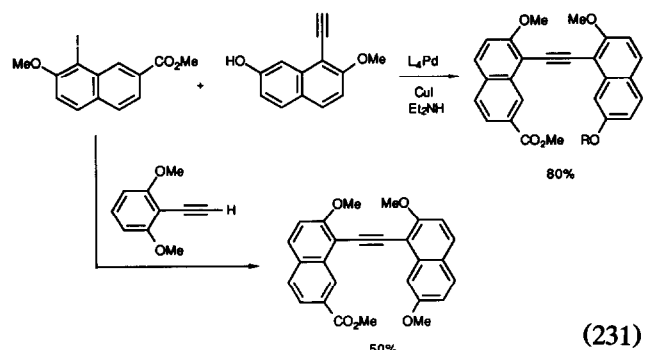
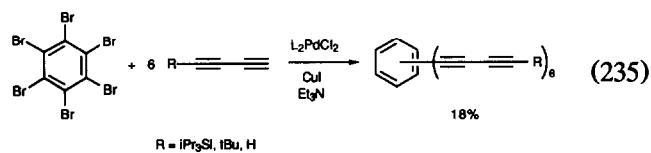
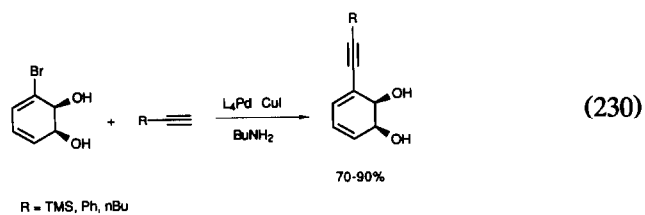
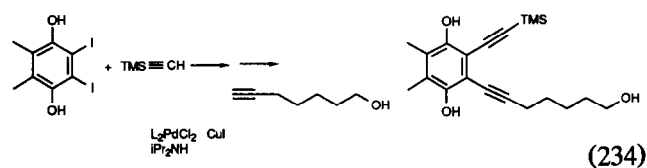


Additional examples are seen in eqn. (223) [253], eqn. (224) [254], eqn. (225) [255], eqn. (226) [256], eqn. (227) [257], eqn. (228) [258], and eqn. (229) [259].

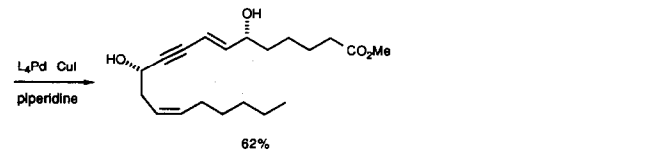
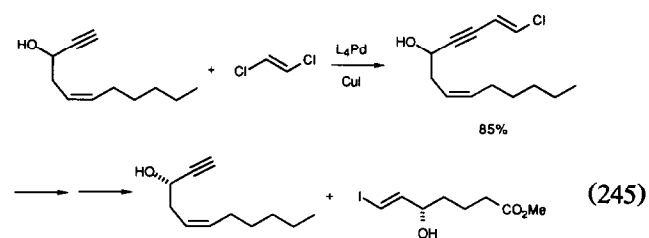
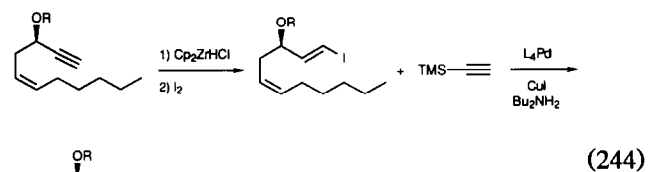
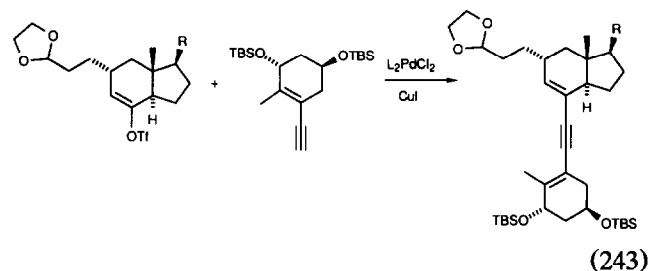
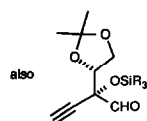
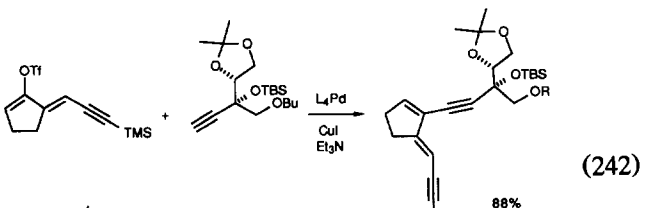
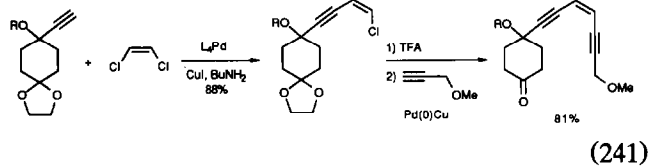
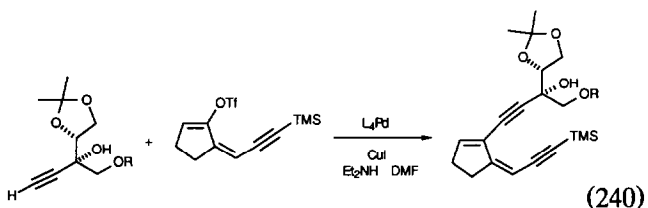
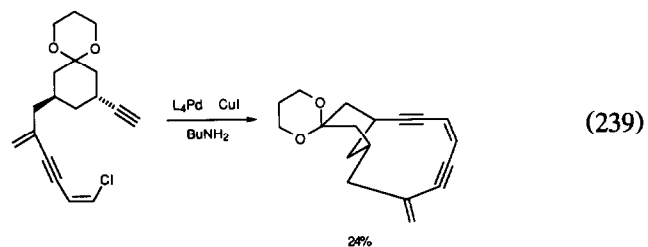
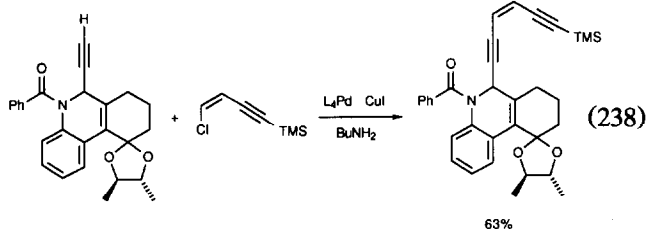
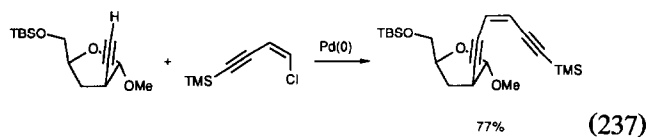




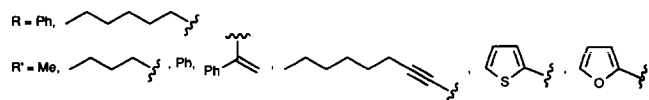
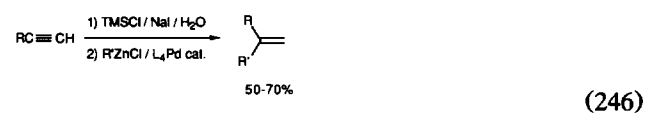
This process was efficient for alkylation of functionalized arenes (eqn. (230) [260] and eqn. (231) [261]), and for multiple incorporation of alkynes into arenes (eqn. (232) [262], eqn. (233) [263], eqn. (234) [264], eqn. (235) [265], and eqn. (236) [266]).

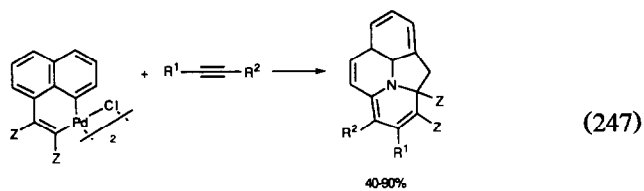


The Castro-Stevens coupling was extensively used in the synthesis of ene-diyne natural products (eqn. (237) [267], eqn. (238) [268], eqn. (239) [269], eqn. (240) [270], eqn. (241) [271], and eqn. (242) [272]) as well as other natural products (eqn. (243) [273], eqn. (244) [274], and eqn. (245) [275]).

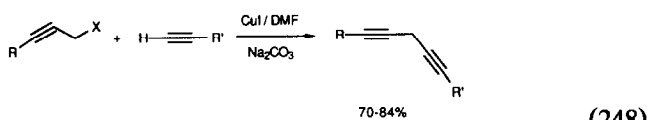


Alkynes were alkylated to give alkenes by treatment with organozinc reagents in the presence of palladium catalysts (eqn. (246) [276]). Orthopalladated species inserted alkynes to give polycyclic materials (eqn. (247) [273]). Alkynes were alkylated by propargyl halides in the presence of copper(I) salts (eqn. (248) [278]). Allyl halides behaved in a similar manner (eqn. (249) [279]), as did aryl and vinyl halides (eqn. (250) [280]). Alkynes were readily carbocuprated (eqn. (251) [281]), and stannylcuprated (eqn. (252) [282]). Copper(I) catalyzed the intramolecular alkylation of alkynes by stabilized carbanions (eqn. (253) [283]).

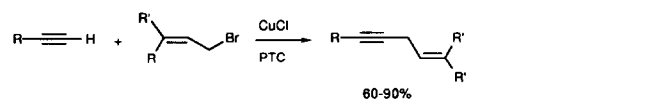




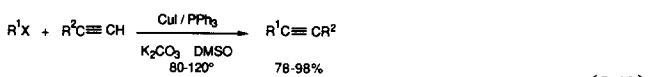
Z = CO₂Me, CF₃ R¹ = CO₂Me, CO₂Et, pNO₂Ph, mCF₃Ph
R² = same



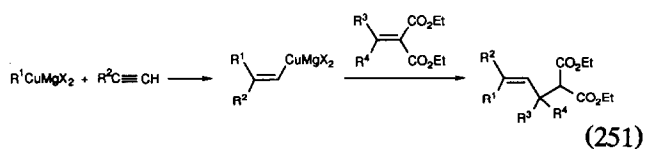
R = TMS, Et, ,
R' = , CO₂Me, CH₂OH, OH



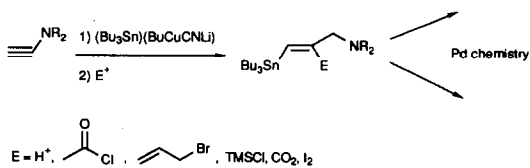
R = , , ,
R' = H, Me



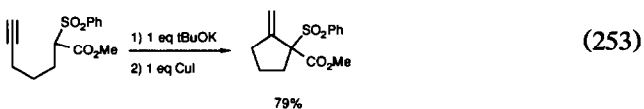
R¹ = Ph, pMePh, pClPh, Ph, , , , ,
R² = Ph,



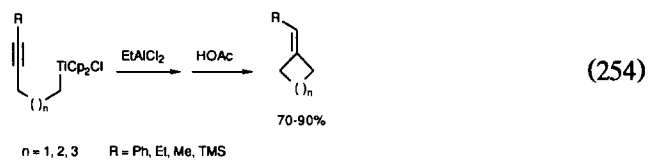
R¹ = , Et, Bu R² = Et, ,
R³ = Me, iPr, H R⁴ = H, Me



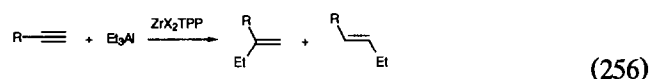
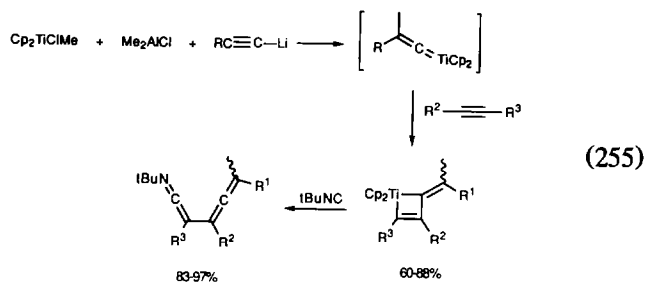
E = H⁺, , , TMSCl, CO₂, I₂



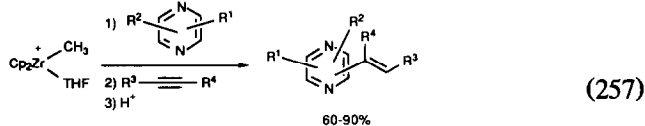
Titanocene complexes alkylated alkynes (eqn. (254) [284], eqn. (255) [285]). Zirconium porphyrin complexes catalyzed the alkylation of alkynes by triethyl aluminum (eqn. (256) [286]). η^2 -Ketimine complexes of zirconium inserted alkynes (eqn. (257) [287] and eqn. (258) [288]). Ruthenium(I) complexes catalyzed the alkylation of alkynes by allyl alcohols (eqn. (259) [289]). Alkynes inserted into arylmanganese complexes (eqn. (260) [290]), and into orthopalladated arenes (eqn. (261) [291]).



n = 1, 2, 3 R = Ph, Et, Me, TMS

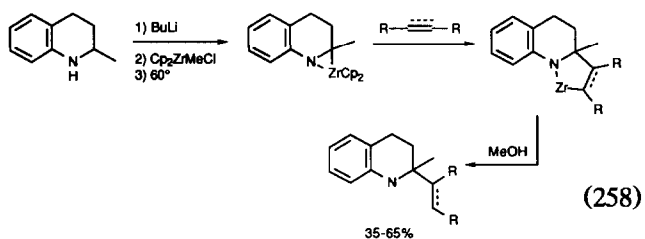


R = nPr, iBu, tBu, , Ph

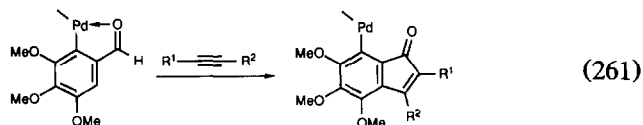
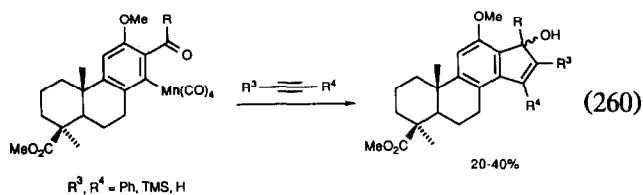
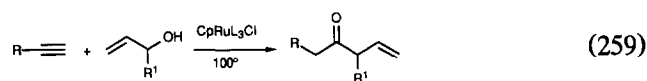


R¹, R², R³ = H, Me

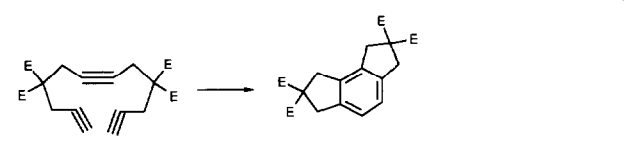
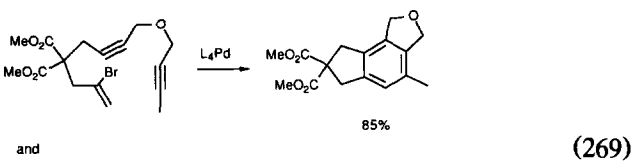
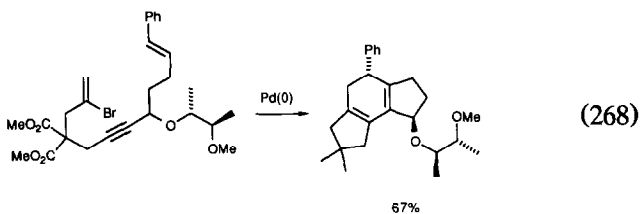
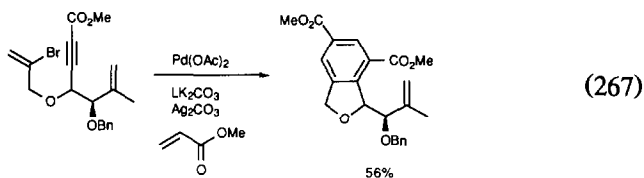
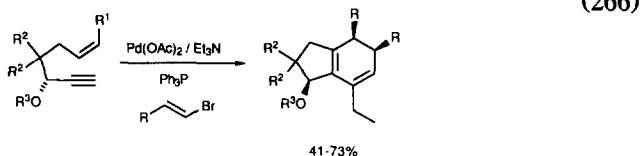
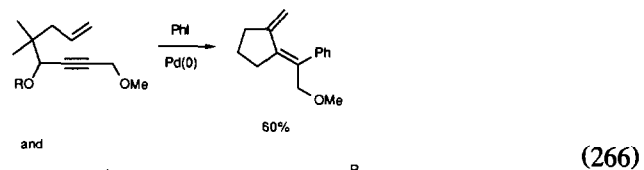
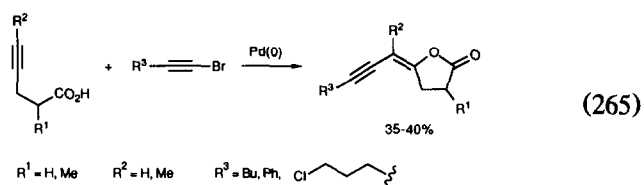
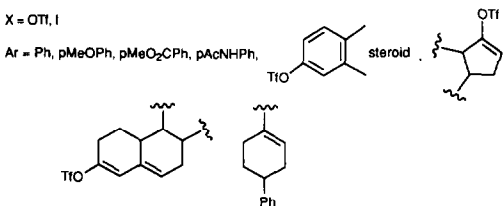
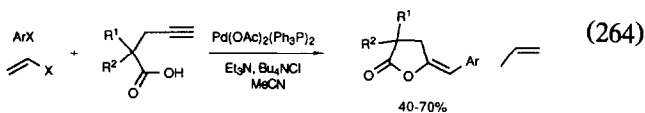
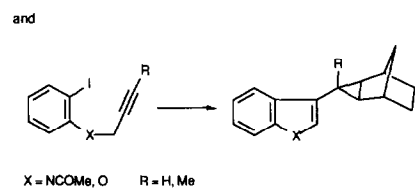
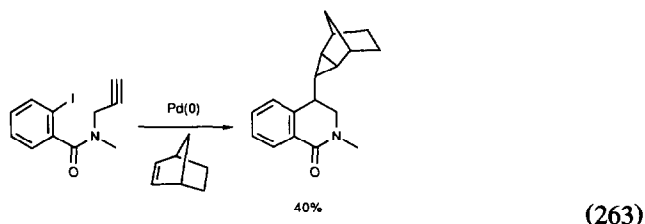
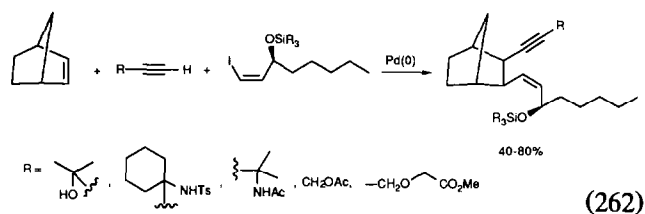
R⁴ = TMS, nPr

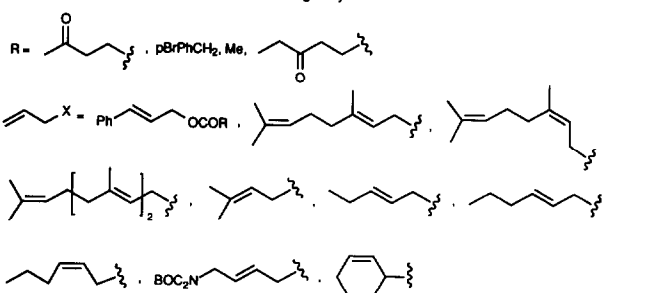
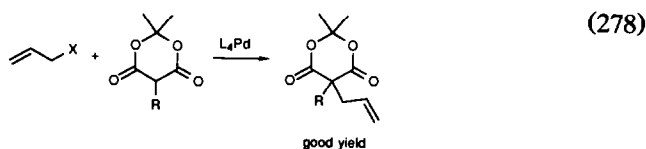
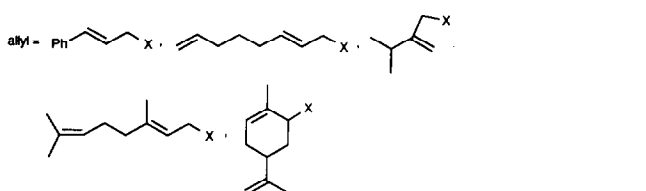
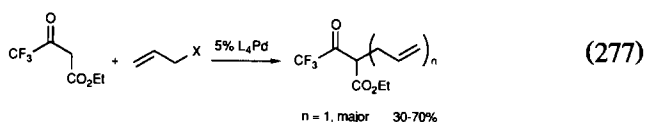
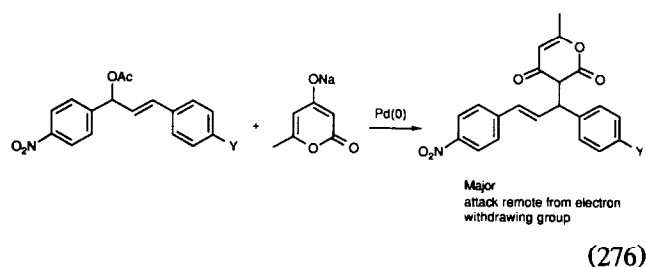
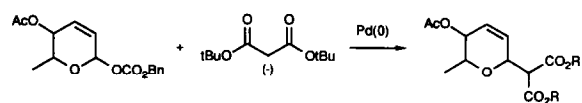
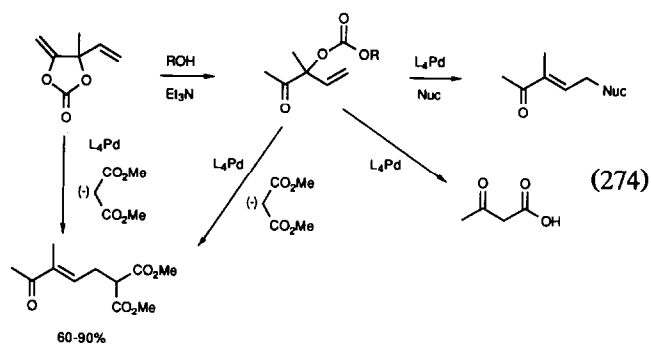
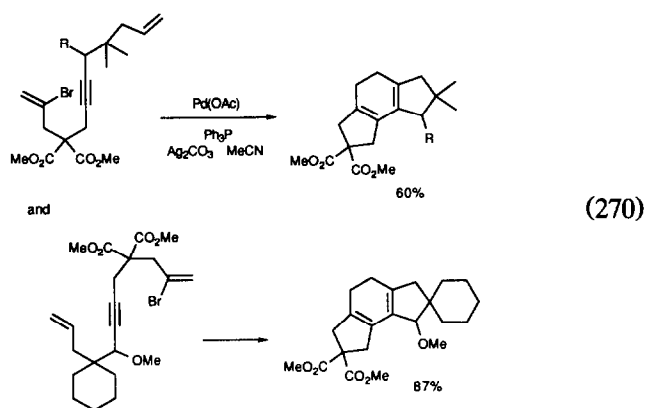


for , TMS-C≡C-



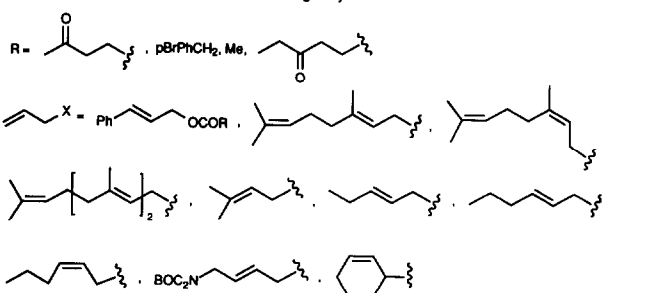
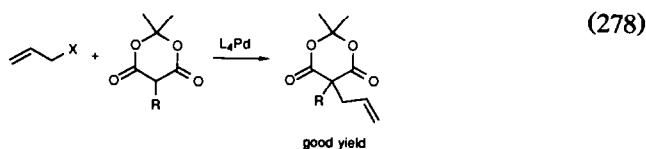
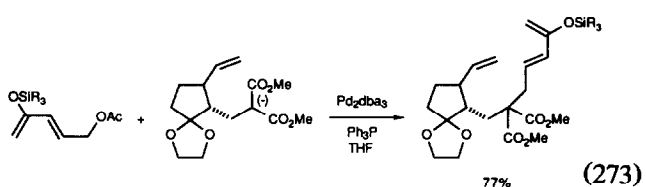
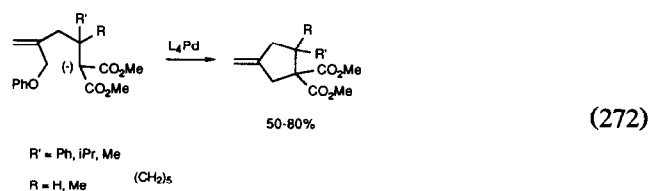
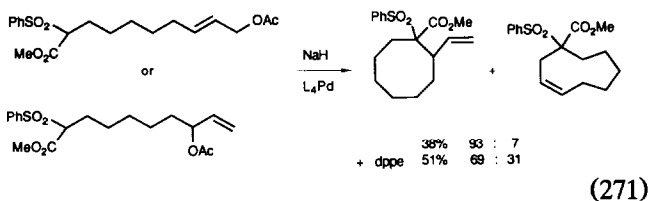
Alkynes participated in a number of “cascade” cyclization processes, involving palladium catalyzed oxidative addition insertion processes (eqn. (262) [292], eqn. (263) [293], eqn. (264) [294], eqn. (265) [295], eqn. (266) [296], eqn. (267) [297], eqn. (268) [298], eqn. (269) [299], and eqn. (270) [300]).



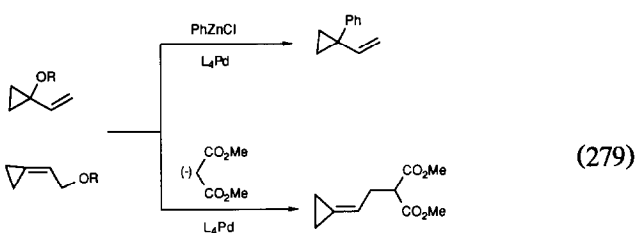


2.1.7. Alkylation of allyl, propargyl and allenyl systems

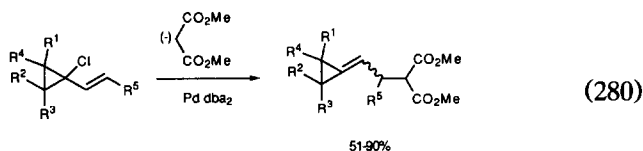
“Selectivity in palladium catalyzed allylic substitutions” was the title of a review (130 references) [301]. Palladium catalyzed alkylation of allyl acetates by stabilized carbanions has been used extensively, both intramolecularly (eqn. (271) [302], eqn. (272) [303]), and intermolecularly (eqn. (273) [304], eqn. (274) [305], eqn. (275) [306], eqn. (276) [307], eqn. (277) [308], and eqn. (278) [309]).



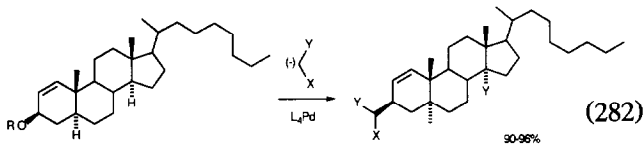
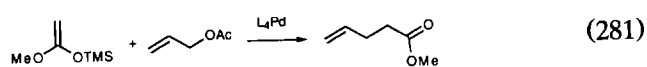
Palladium(0) catalyzed the alkylation of allylic systems of vinyl cyclopropanes (eqn. (279) [310], eqn. (280) [311]), the alkylation of allyl acetates by ketene silyl acetals (eqn. (281) [312]), the alkylation of cholestenyl esters by carbon nucleophiles (eqn. (282) [313]), and the alkylation of allyl acetates by α -hydroxy acid derivatives (eqn. (283) [314]). Vinyl mercurials (eqn. (284) [315]) and zinc reagents (eqn. (285) [316]) alkylated allyl acetates under palladium catalysis.



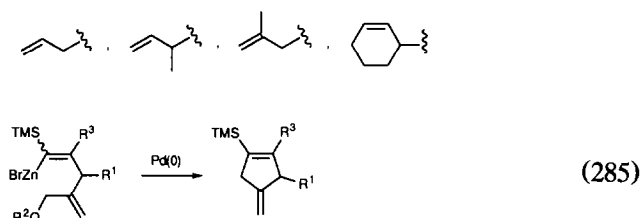
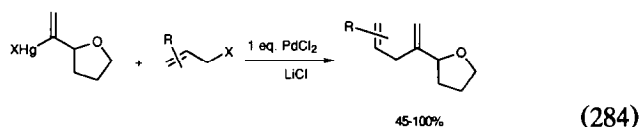
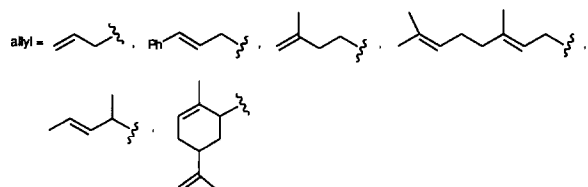
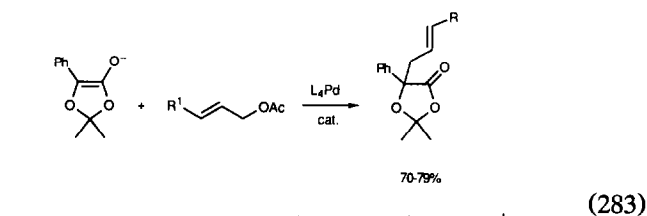
R = Ac, Tf, CO₂Et, Ts



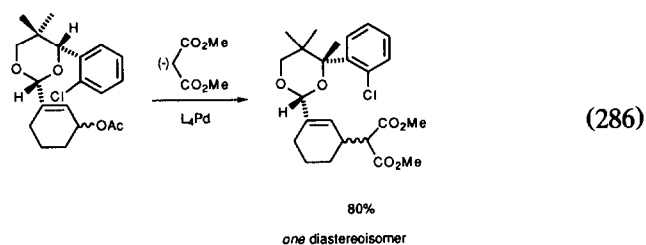
R¹ = H, Me, TMSCH₂ R², R³, R⁴, R⁵ = H, Me



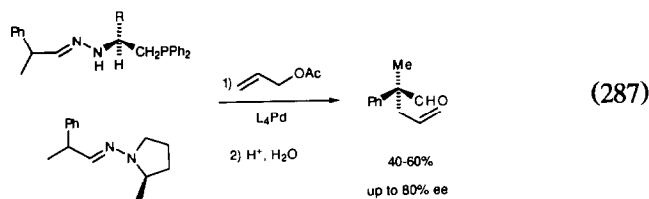
X = Y = CO₂Me, CN
X = CO₂Me, Y = COMe



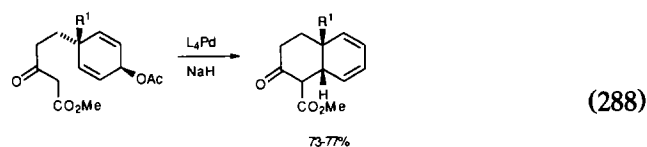
Asymmetric induction in the palladium catalyzed allylic alkylation reaction has been achieved both by using chiral substrates (eqn. (286) [317], eqn. (287) [318]), and chiral ligands (eqn. (288) [319], eqn. (289) [320], eqn. (290) [321], eqn. (291) [322], and eqn. (292) [323]).



one diastereoisomer

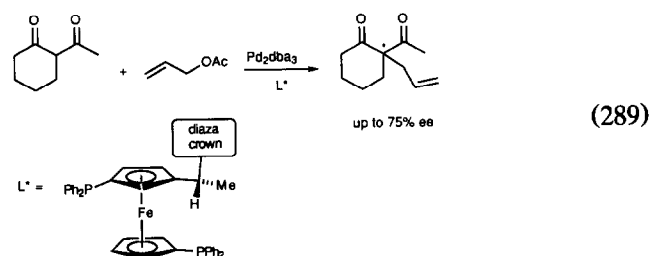


40-60%
up to 80% ee

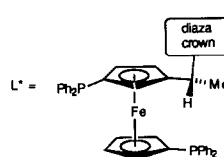


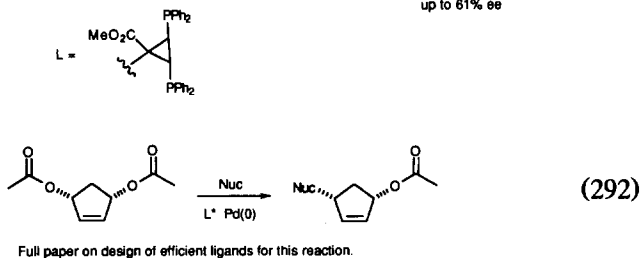
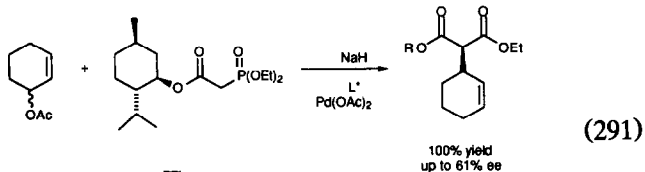
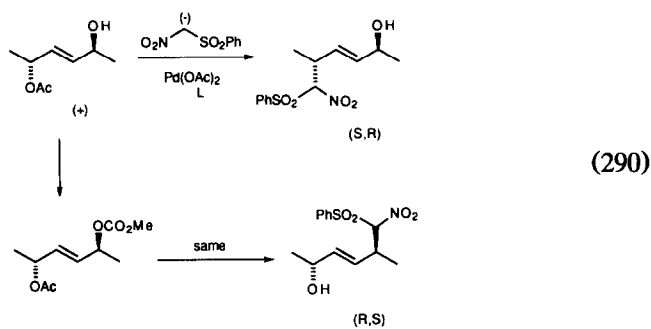
73-77%

with L = (R)(S)BPPFA
34% yield, 83% ee

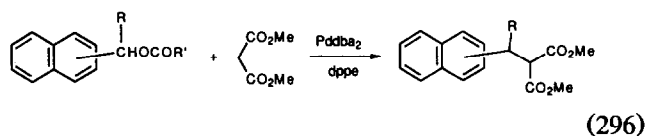
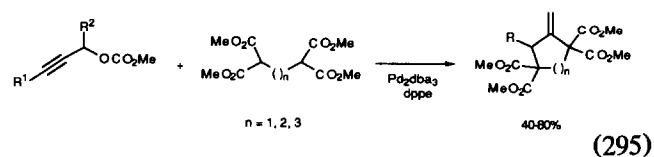
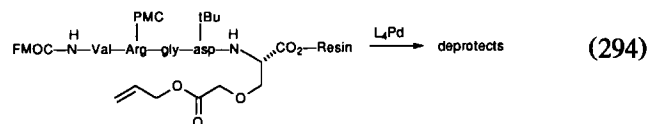
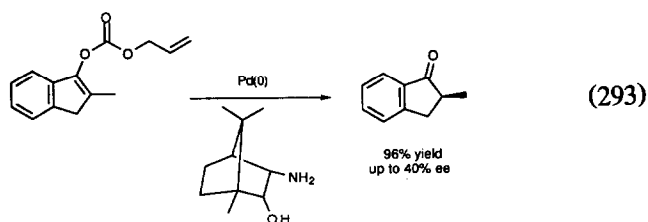


up to 75% ee

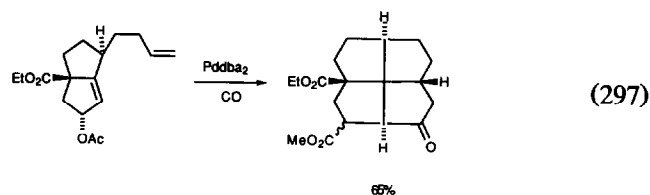




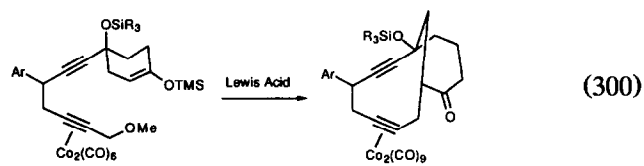
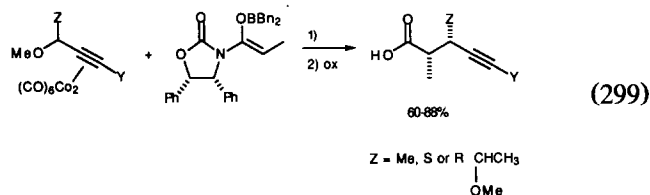
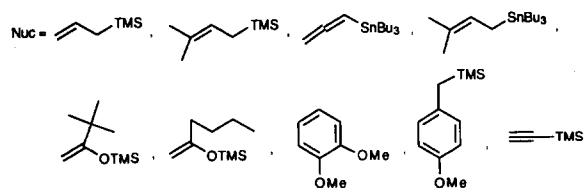
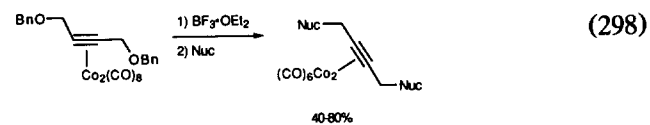
Palladium(0) complexes catalyzed the removal of allyl carbonate protecting groups (eqn. (293) [324] and eqn. (294) [325]). Palladium catalyzed reactions of propargyl carbonates have been reviewed (27 references) [326]. Propargyl carbonates were alkylated by stabilized carbanions in the presence of palladium(0) catalysts (eqn. (295) [327,328]). Benzyl carbonates underwent similar alkylations (eqn. (296) [329]).



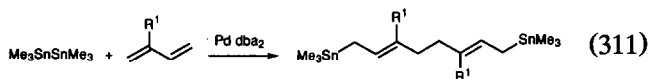
Palladium(0) complexes catalyzed "cascade" cyclizations which were initiated by oxidative addition to an allyl acetate (eqn. (297) [330,331]).



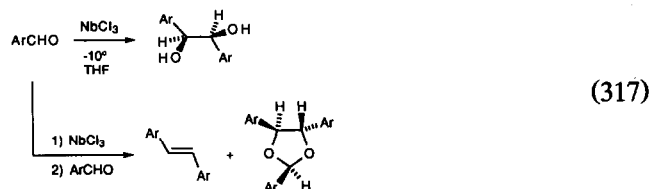
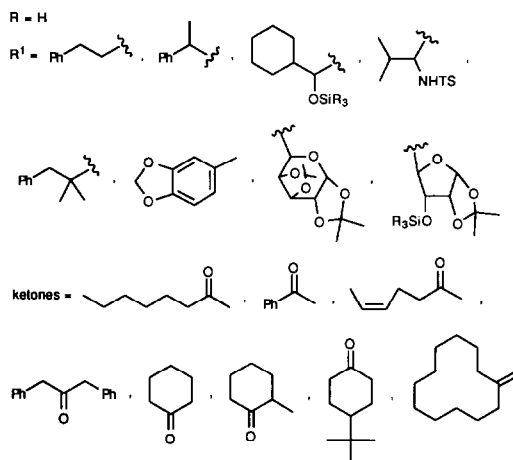
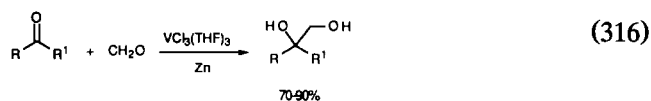
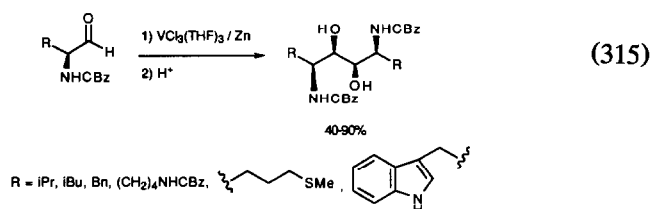
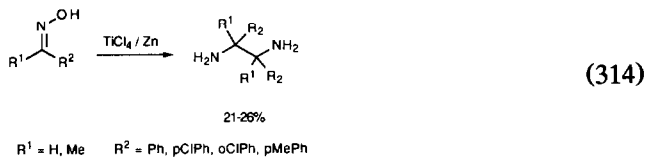
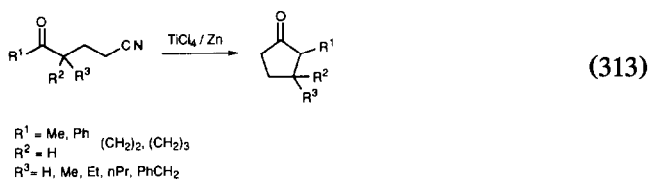
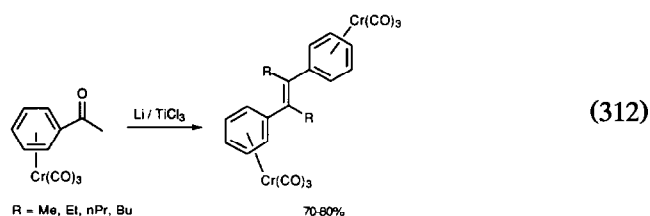
Cobalt-stabilized propargyl cations were alkylated by a variety of carbon nucleophiles (eqn. (298) [332], eqn. (299) [333], and eqn. (300) [334]).



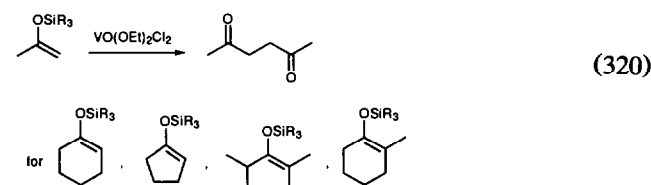
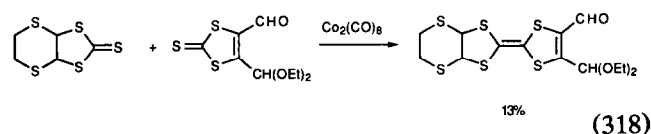
Organocuprates alkylated allylic (eqn. (301) [335] and eqn. (302) [336]) and propargyl systems (eqn. (303) [337] and eqn. (304) [338]). The stereo- and regiochemistry of displacement of allyl benzothiazines by alkyl copper species was the subject of an extensive investigation [339].

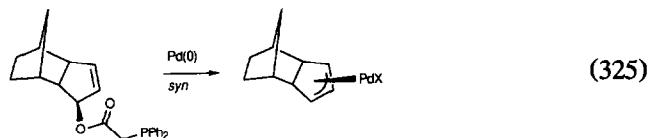
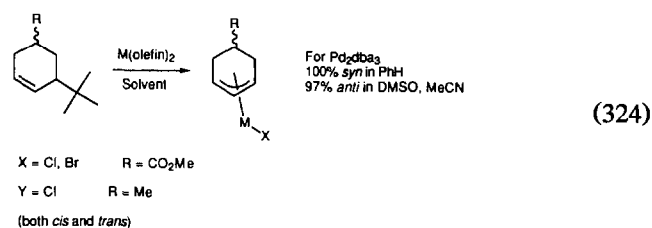
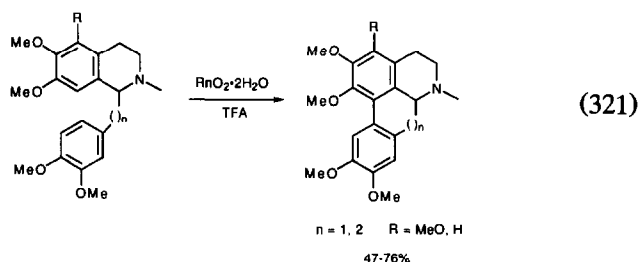


Low valent titanium coupled chromium-complexed aryl ketones (eqn. (312) [348]), α -ketonitriles (eqn. (313) [349]), and oximes (eqn. (314) [350]). Low valent vanadium species coupled α -amino aldehydes (eqn. (315) [351]), and cross coupled ketones with formaldehyde (eqn. (316) [352]). Niobium(III) coupled aldehydes (eqn. (317) [353]).



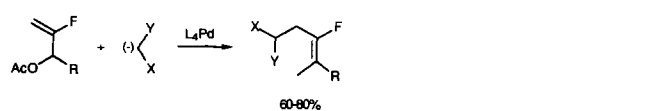
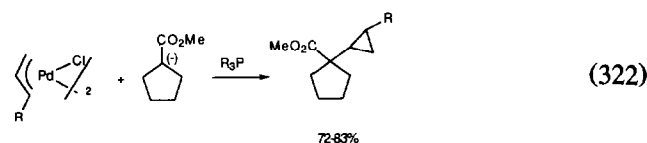
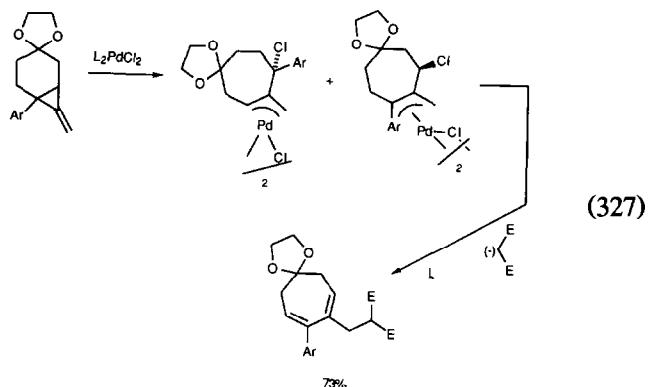
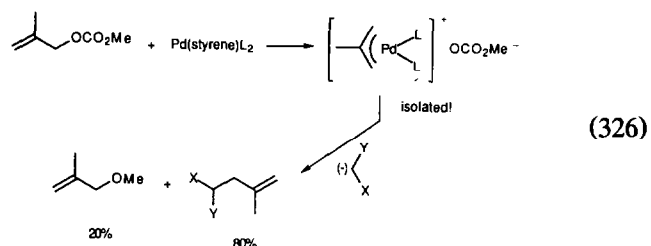
Dicobalt octacarbonyl coupled thiocarbonates (eqn. (318) [354]). Osmium porphyrin complexes catalyzed the coupling of diazocompounds (eqn. (319) [355]). Silylenol ethers were coupled by vanadium oxychlorides (eqn. (320) [356]). Ruthenium oxide catalyzed oxidative biaryl coupling (eqn. (321) [357]).





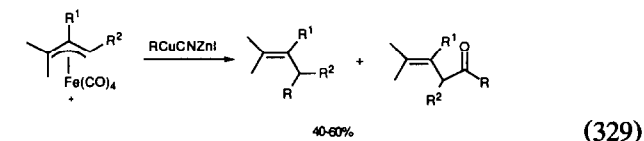
2.1.9. Alkylation of π -allyl complexes

A number of unusual facets of reactivity of π -allylpalladium complexes have been reported this year. The fact that α -branched ester enolates attacked the *central* carbon of π -allylpalladium complexes, giving cyclopropanes, a reaction originally reported over ten years ago, was again reported (eqn. (322) [358]). Complexes from fluorinated allyl acetates were alkylated at the less substituted position (eqn. (323) [359]). The stereochemistry of oxidative addition of palladium(0) complexes to allyl acetates depended strongly on the solvent (eqn. (324) [360]), and on the steric bias of the starting material (eqn. (325) [361]). Evidence for the intermediacy of cationic allyl palladium phosphine complexes in palladium-catalyzed allylic alkylations was obtained by isolating and studying just such intermediates (eqn. (326) [362]). π -Allylpalladium complexes from methlenecyclopropanes were alkylated by carbanions (eqn. (327) [363]). Palladium(0) complexes *isomerized* optically active π -allyl complexes, accounting for loss of stereoselectivity in some reactions (eqn. (328) [364]). π -Allyliron complexes were alkylated by organocuprates (eqn. (329) [365]) and organolithium (eqn. (330) [366]).



$X = \text{COMe}, \text{COPh}, \text{CO}_2\text{Me}, \text{CO}_2\text{Et}, \text{SO}_2\text{Ph}$

$Y = \text{CO}_2\text{Me} \quad R = \text{Ph}, \text{iPr}, \text{cyclohexyl}, \text{nBu}, \text{cyclohexyl}$

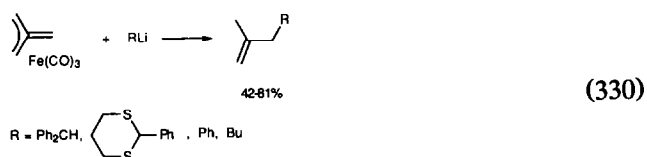


$R^1 = \text{H}, \text{Me}$

$R^2 = \text{H}, \text{Me}$

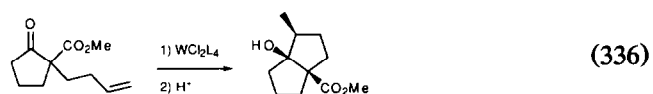
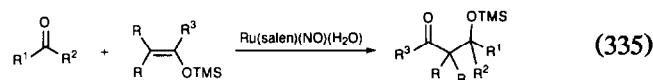
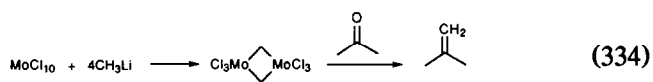
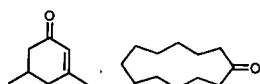
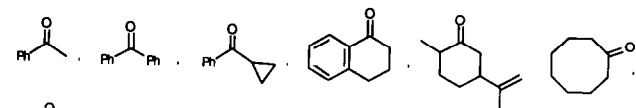
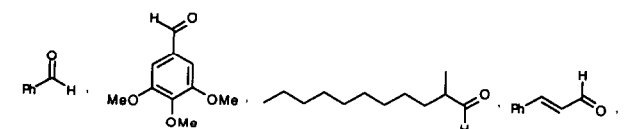
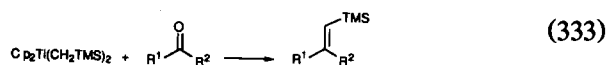
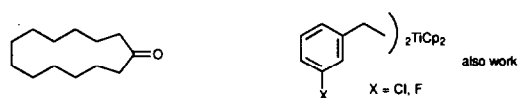
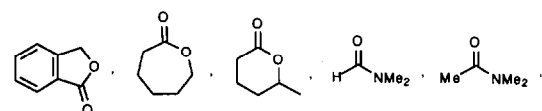
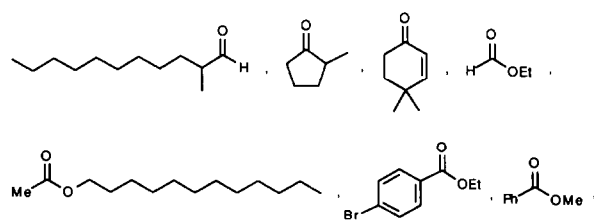
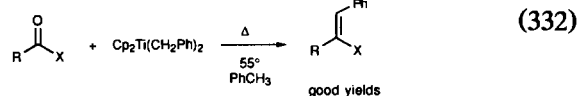
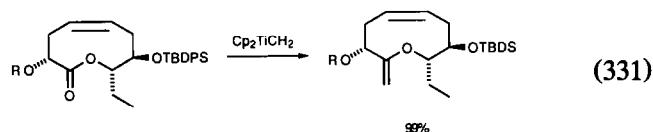
$R = \text{EtO}_2\text{C(CH}_2)_5\text{S}^{\text{t}}, \text{AcO(CH}_2)_5\text{S}^{\text{t}}$

$\text{NC(CH}_2)_5\text{S}^{\text{t}}, \text{PhCH}_2$

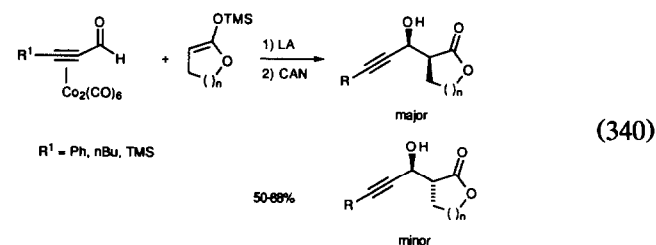
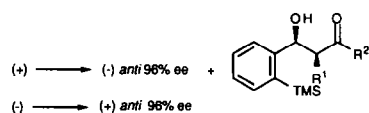
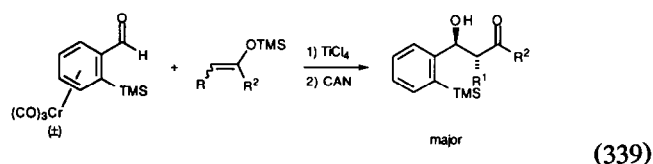
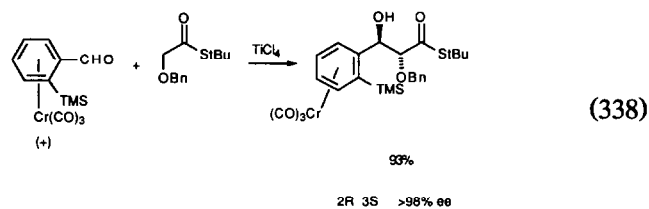
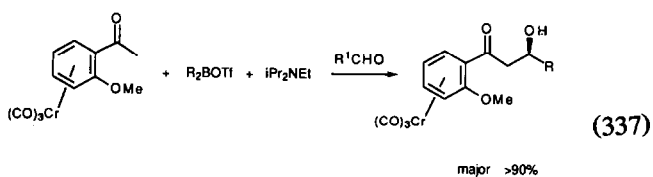


2.1.10. Alkylation of carbonyl compounds

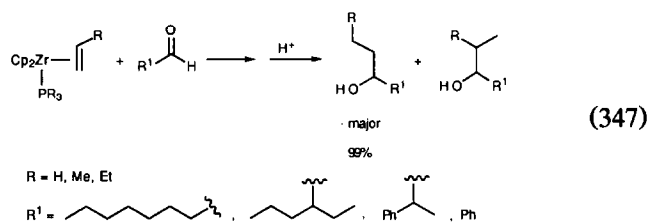
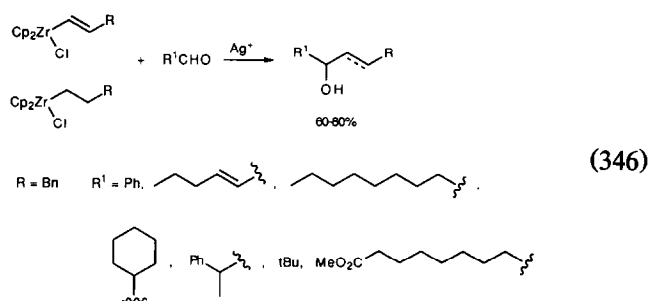
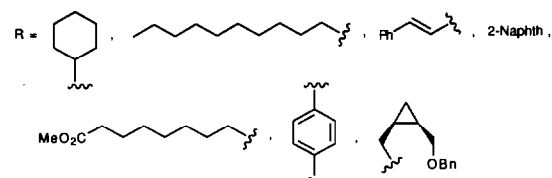
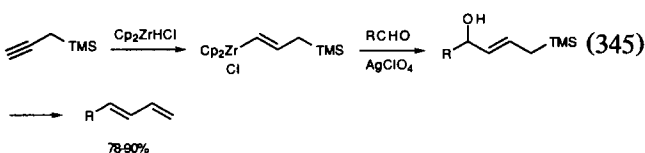
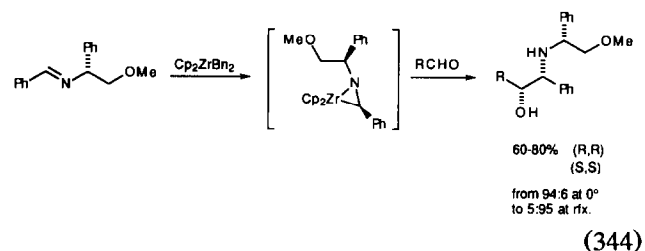
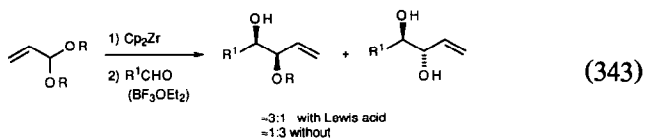
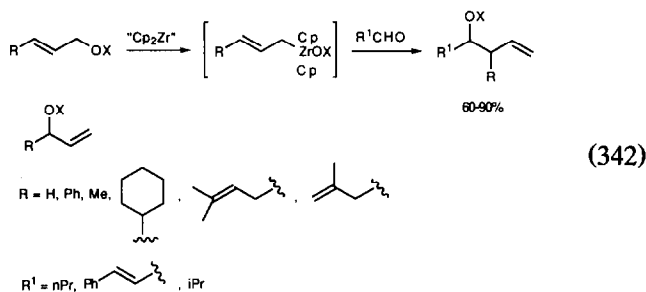
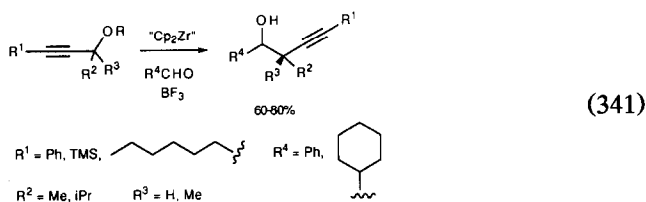
Tebbe's reagent effectively methylenated a complex lactone (eqn. (331) [367]). Several new carbonyl olefination reagents have been developed (eqn. (332) [368], eqn. (333) [369], and eqn. (334) [370]). Ruthenium salen complexes catalyzed the alkylation of ketones by silyl enol ethers (eqn. (335) [371]). Tungsten(II) complexes promoted the alkylation of ketones by alkenes (eqn. (336) [372]).



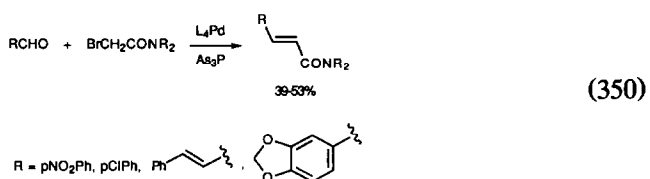
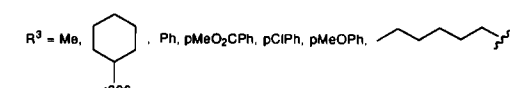
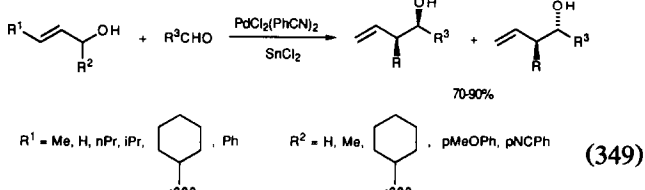
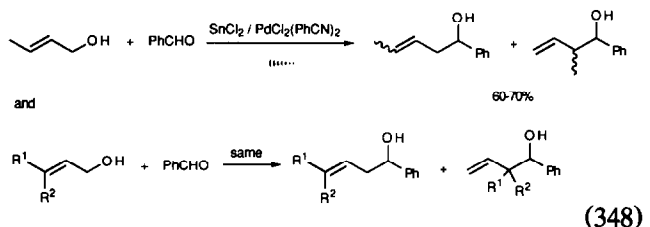
Chromium complexes of aryl ketones (eqn. (337) [373]) and aryl aldehydes (eqn. (338) [374] and eqn. (339) [375,376]) were alkylated with high stereoselectivity. Cobalt complexed propargyl aldehydes were alkylated by silyl ketene acetals (eqn. (340) [377]).

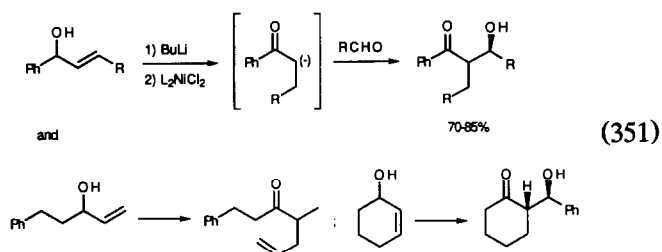


Aldehydes were alkylated by propargyl ethers (eqn. (341) [378]) and allyl ethers (eqn. (342) [379] and eqn. (343) [380]) in the presence of zirconocene. η^2 -zirconium complexes alkylated aldehydes (eqn. (344) [381]). Hydrozirconation of alkynes and alkenes gave zirconium alkyls which alkylated aldehydes (eqn. (345) [382], eqn. (346) [383], and eqn. (347) [384]).

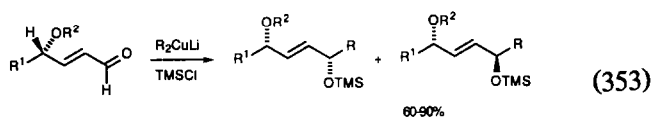
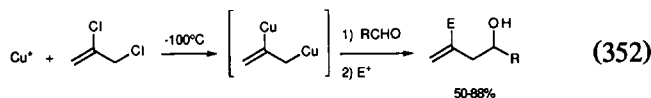


Palladium catalyzed the alkylation of aldehydes by allyl alcohols (eqn. (348) [385] and eqn. (349) [386]) and by α -bromoamides (eqn. (350) [387]). Nickel enolates alkylated aldehydes (eqn. (351) [388]).

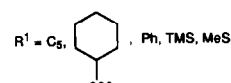
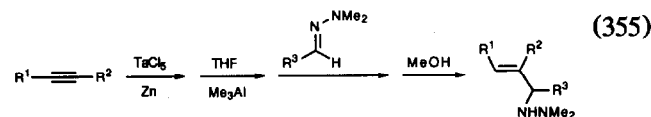
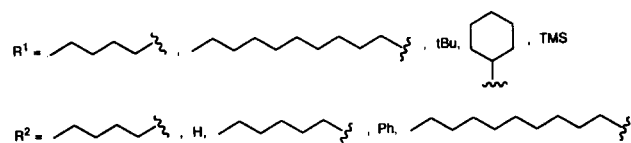
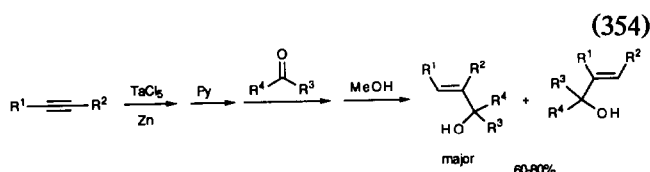




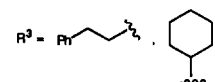
Aldehydes were alkylated by allyl cuprates (eqn. (352) [389]) and alkyl cuprates (eqn. (353) [390]). Reduced tantalum species alkylated aldehydes and ketones with alkynes (eqn. (354) [391,392]). Imines were alkylated in a similar manner (eqn. (355) [393]).



R = Ph, Bu, tBu

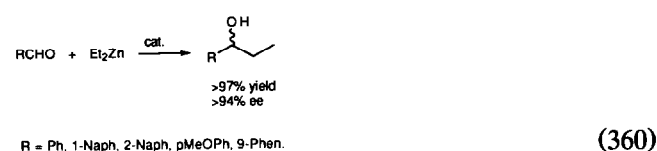
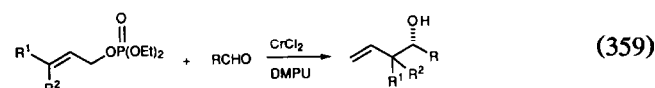
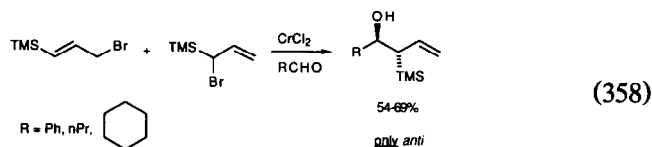
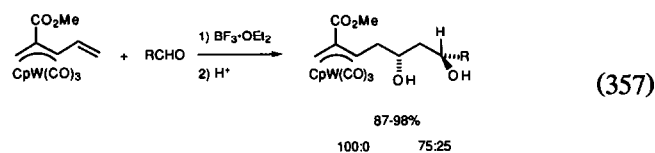
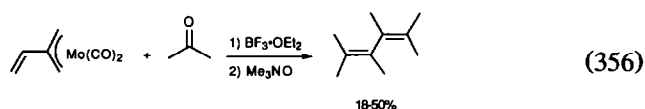


R² = C₅, C₆, C₁₀

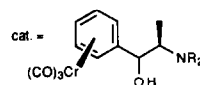


from 1:1 to >99%

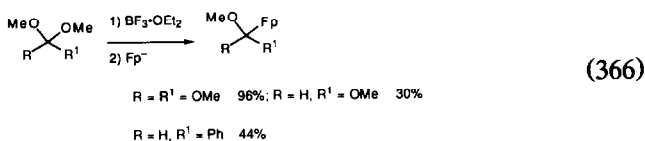
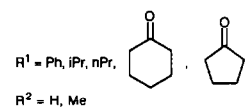
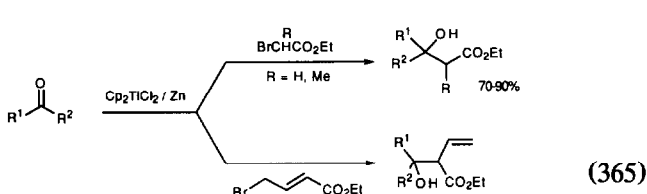
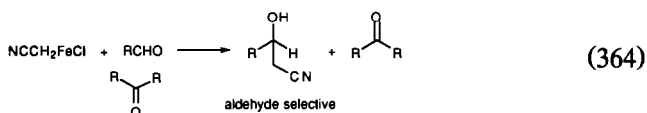
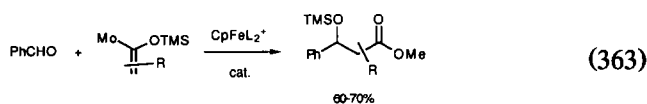
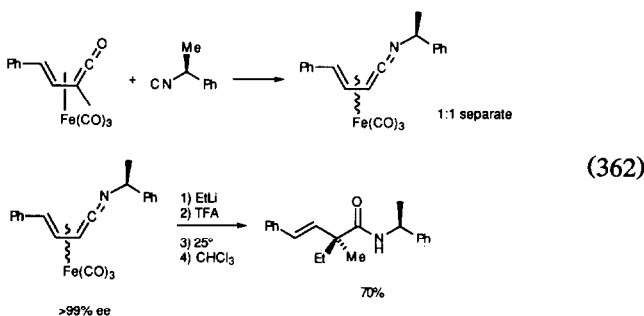
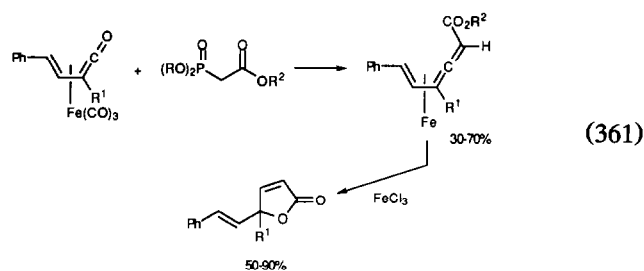
Allylmolybdenum complexes alkylated aldehydes (eqn. (356) [394], and eqn. (357) [395]). The addition of organochromium compounds to aldehydes – the Nozaki–Hiyama reaction – was the subject of a review (43 references) [396]. Allylic halides alkylated aldehydes when treated with chromium(II) salts (eqn. (358) [397] and eqn. (359) [398]). Optically active arene chromium complexes induced asymmetry in the alkylation of aldehydes by diethyl zinc (eqn. (360) [399]).



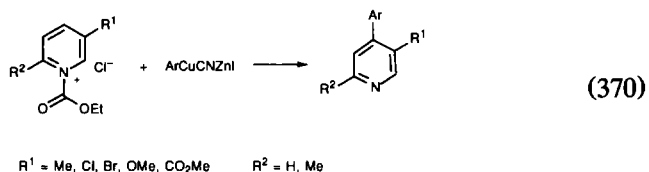
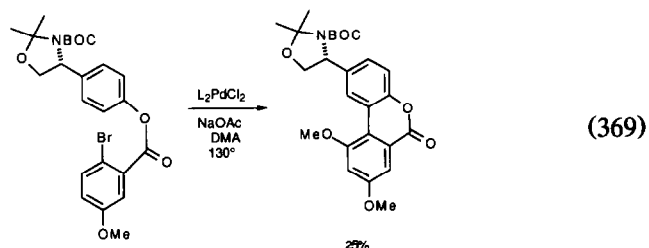
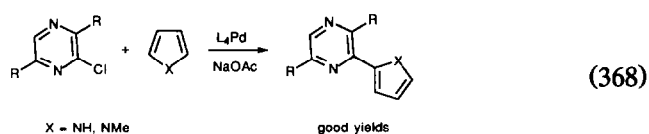
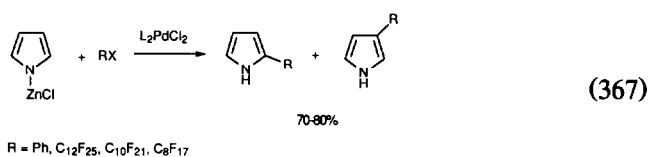
R = Ph, 1-Naph, 2-Naph, pMeOPh, 9-Phen.



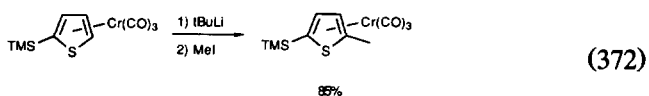
Iron complexes of vinyl ketenes were alkylated by stabilized ylides (eqn. (361) [400]) and by alkyllithium (eqn. (362) [401]). Cationic iron cyclopentadienyl complexes catalyzed the alkylation of aldehydes by ketene silyl acetals (eqn. (363) [402]). Iron(II) complexes of acetonitrile anion alkylated ketones [403] and aldehydes (eqn. (364) [404]). Titanocene alkylated ketones with α -bromoesters (eqn. (365) [405]). Iron carbene precursors were made by the reaction of ketals with anionic cyclopentadienyl iron complexes (eqn. (366) [406]).



reaction of Grignard reagents with aryl triflates (eqn. (371) [411]).

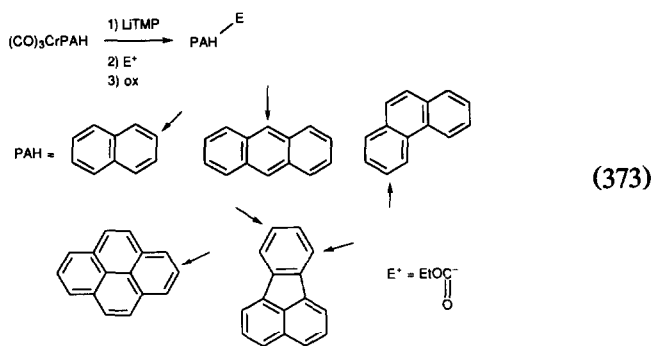


Chromium-complexed thiophene was lithiated, then α -alkylated (eqn. (372) [412]). Chromium-complexed polyaromatic hydrocarbons were similarly alkylated (eqn. (373) [413]). Chromium-complexed tetraline (eqn. (374) [414]), benzaldehyde SAMP derivatives (eqn. (375) [415]), oxazolines (eqn. (376) [416]) and acetophenone ketals (eqn. (377) [417]) were lithiated, then alkylated.

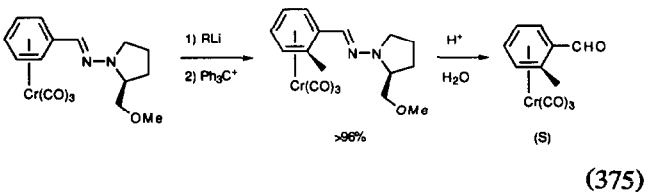
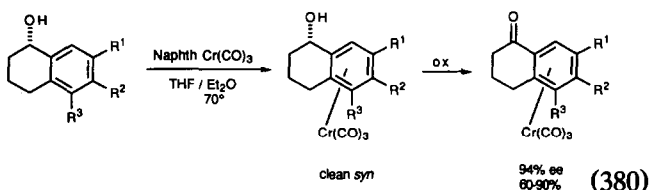
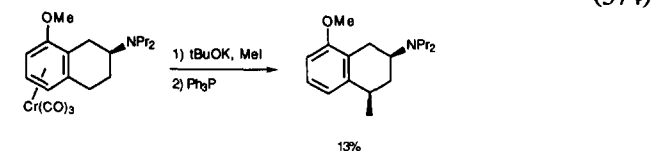
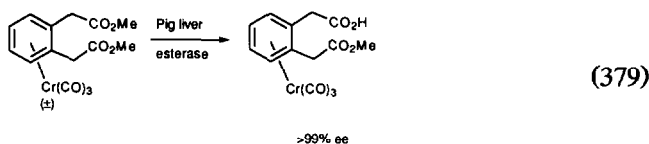
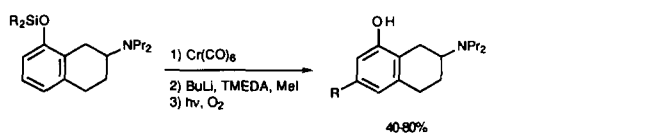
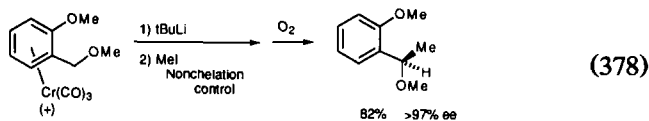


2.1.11. Alkylation of aromatic compounds

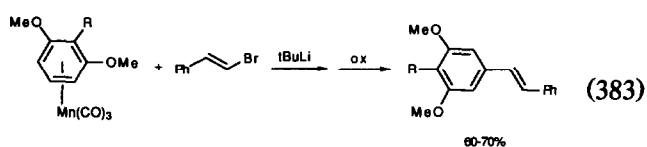
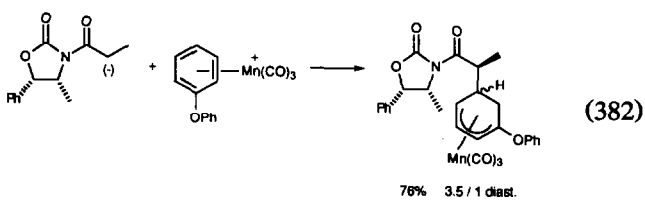
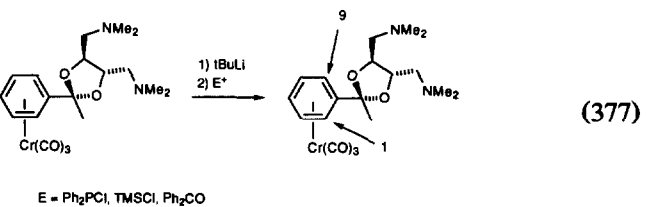
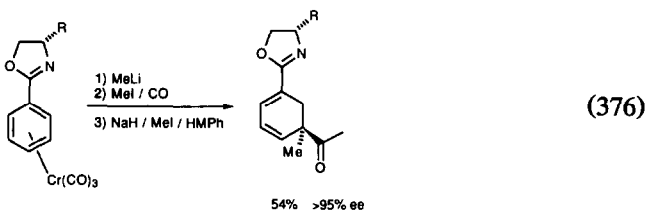
Palladium(II) complexes catalyzed the alkylation of pyrroles by organic halides (eqn. (367) [307] and eqn. (368) [408]), as well as intramolecular alkylation of arenes by aryl bromides (eqn. (369) [409]). Aryl cuprates added to the 4-position of N-acylpyridinium salts (eqn. (370) [410]). Nickel(II) acetylacetonate catalyzed the



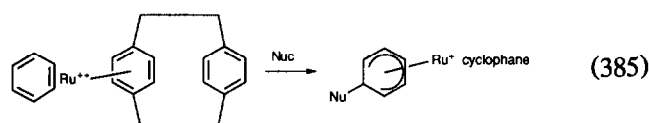
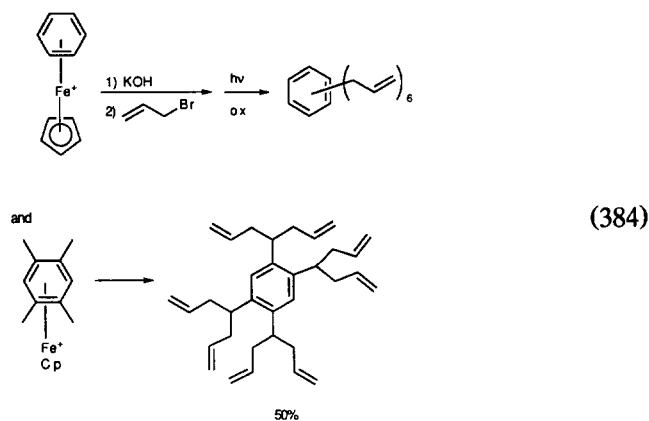
Stereoselective manipulation of acetals derived from *o*-substituted benzaldehyde chromium tricarbonyl complexes was the topic of a lecture [422].



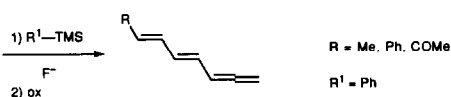
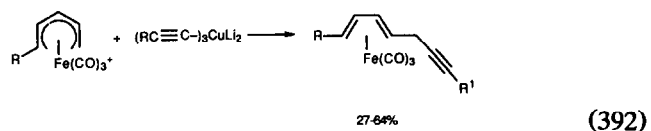
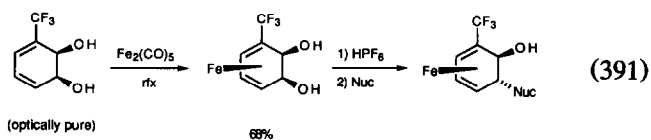
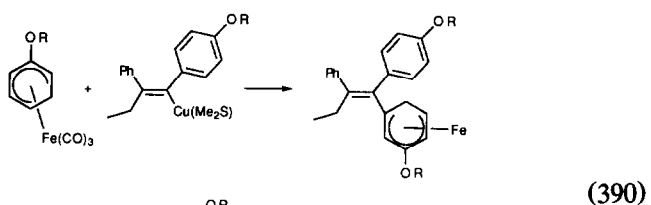
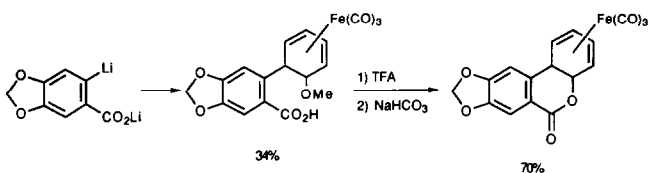
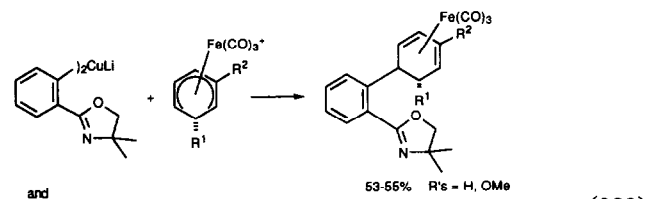
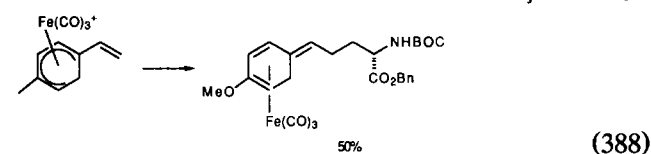
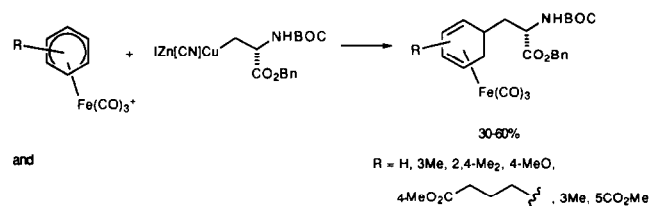
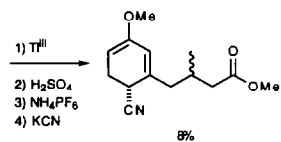
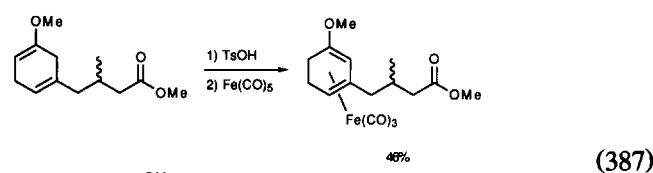
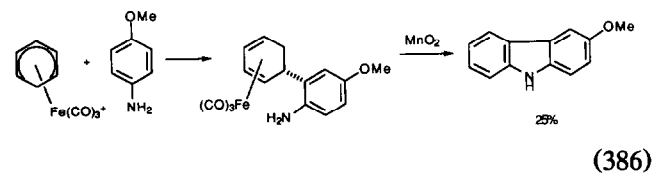
Arene complexes of manganese (eqn. (380) [423], eqn. (382) [424], eqn. (383) [425]), iron (eqn. (384) [426]) and ruthenium (eqn. (385) [427]) were alkylated.



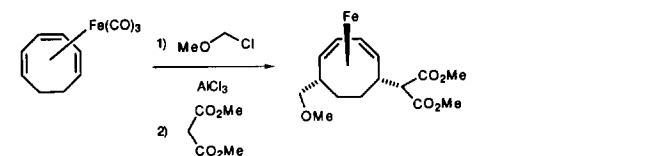
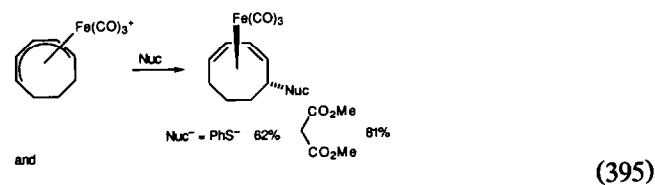
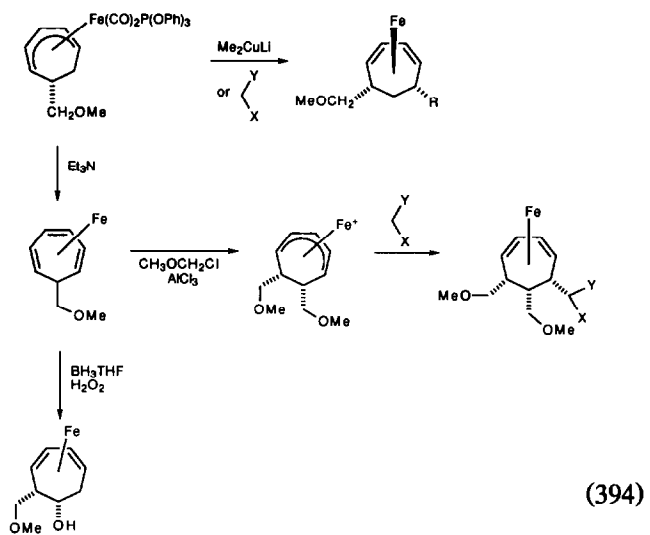
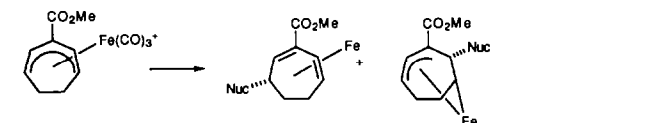
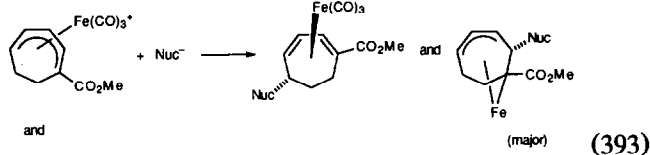
" α -Benchrotrenic carbanions. Formation, stabilization and synthetic applications" was the title of a review (69 references) [418]. Optically active arene-chromium tricarbonyl complexes were benzylically alkylated with high ee (eqn. (378) [419]). Optically active chromium arene complexes were prepared by the procedures shown in eqn. (379) [420] and eqn. (380) [421].



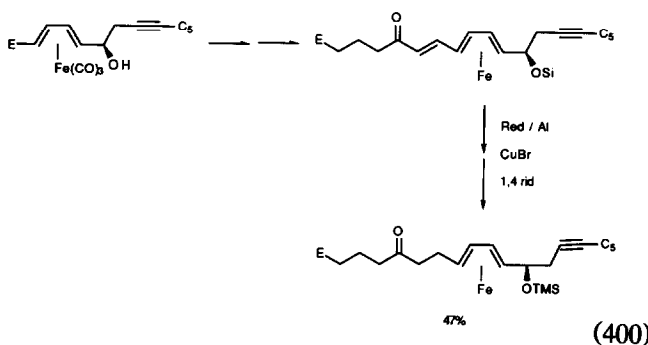
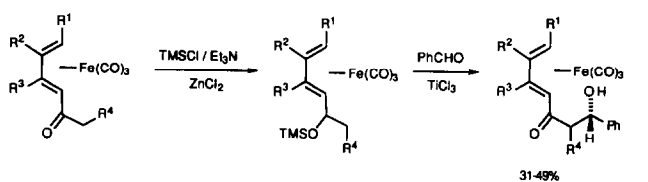
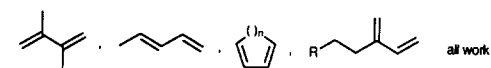
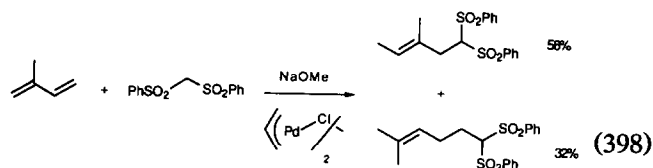
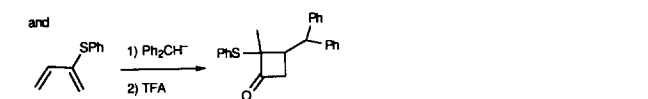
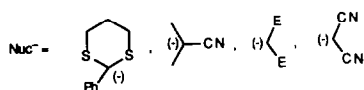
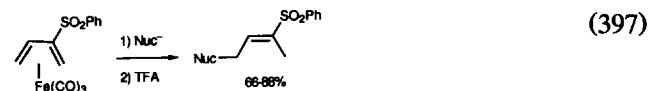
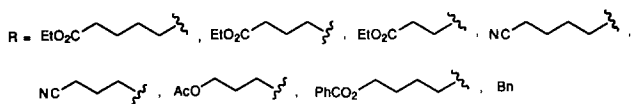
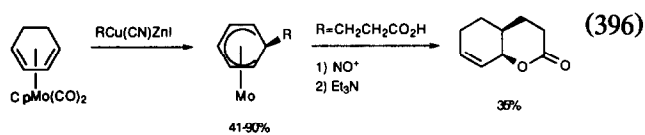
2.1.12. Alkylation of diene and dienyl complexes
 "Aryl substituted cyclohexadienyl iron complexes in alkaloid synthesis" was the title of a dissertation [428]. Indole alkaloids were synthesized in low yields by oxidative cleavage of an iron-diene complex (eqn. (386) [429]). The role of substituent patterns in selectivity of preparation of cationic iron cyclohexadienyl complexes by acid catalyzed demethoxylation of iron complexes alkoxy-cyclohexadienes was examined [430]. Iron dienyl complexes found numerous applications in organic synthesis (eqn. (387) [431], eqn. (388) [432], eqn. (389) [433], eqn. (390) [434], eqn. (391) [431], and eqn. (392) [436]).

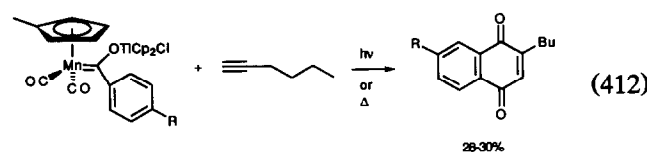
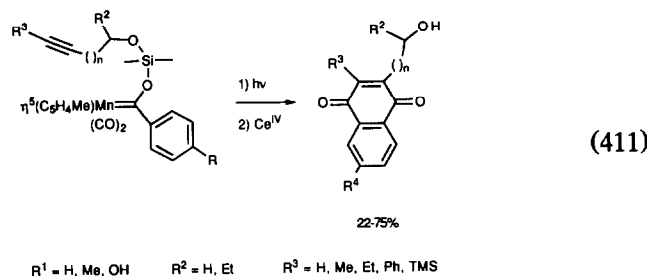
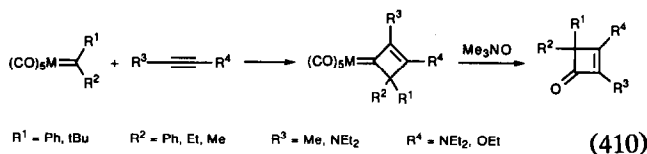


Cycloheptadienyl complexes (eqn. (393) [437], eqn. (394) [438] and cyclooctadienyl complexes eqn. (395) [439] were similarly useful.

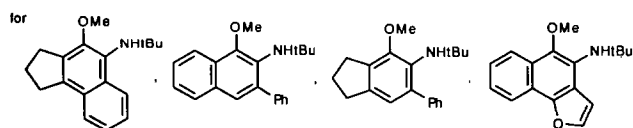
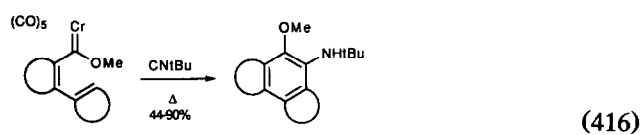
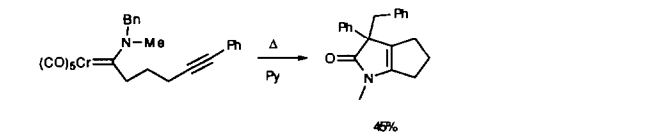
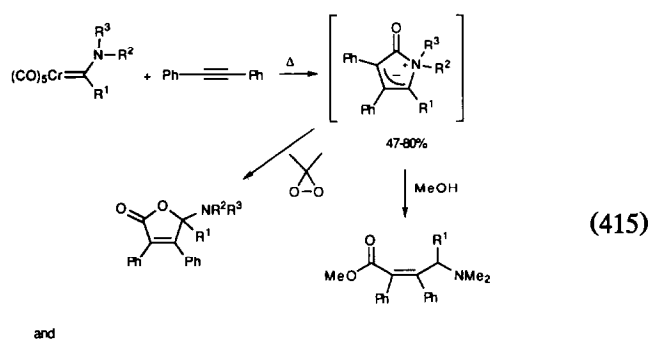
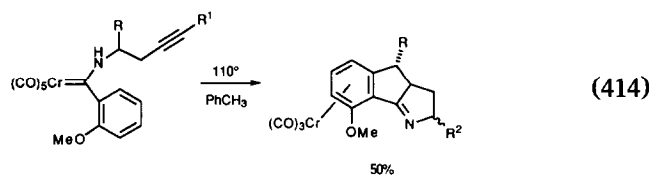
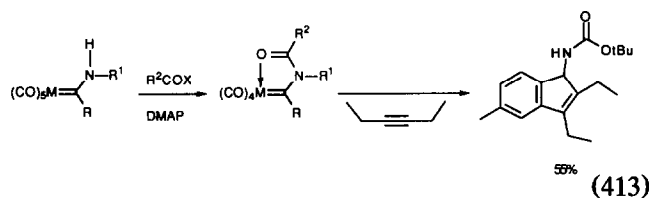


Diene complexes were also reactive toward nucleophiles (eqn. (396) [440], eqn. (397) [441], and eqn. (398) [442]). Dienes were complexed to iron to protect them from reagents used for carrying out reactions at remote sites (eqn. (399) [443], eqn. (400) [444], eqn. (401) [445]).

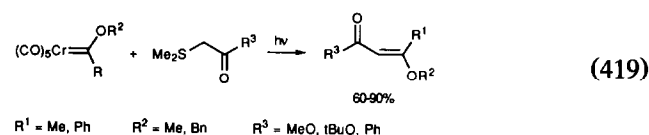
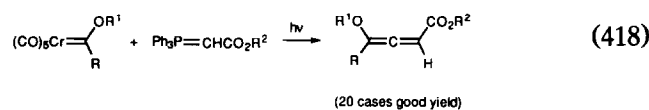
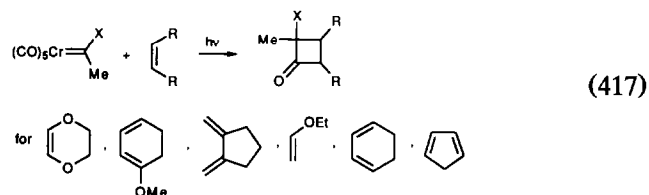




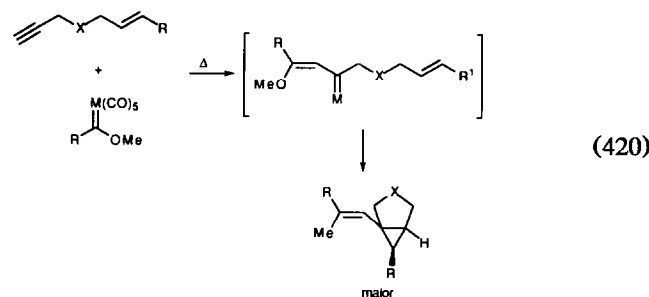
Aminocarbene complexes underwent reactions with alkynes without undergoing CO insertion (eqn. (413) [458], eqn. (414) [459], and eqn. (415) [460]). Polyunsaturated carbene complexes cyclized with isocyanides to give aminophenols (eqn. (416) [461]).

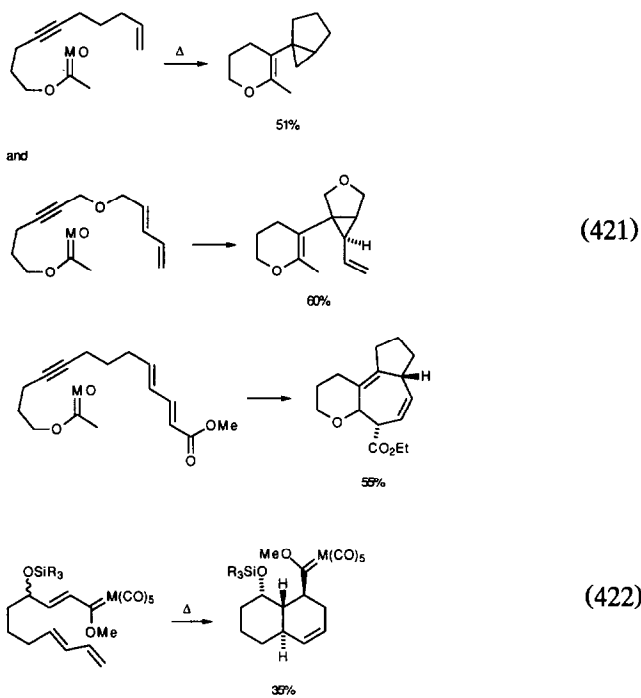


Chromium carbene complexes underwent photochemical cycloaddition with alkenes to give cyclobutanones (eqn. (417) [462]), with stabilized ylides to give push-pull allenes (eqn. (418) [463]) and with sulfur ylides to give conjugated enones (eqn. (419) [464]).

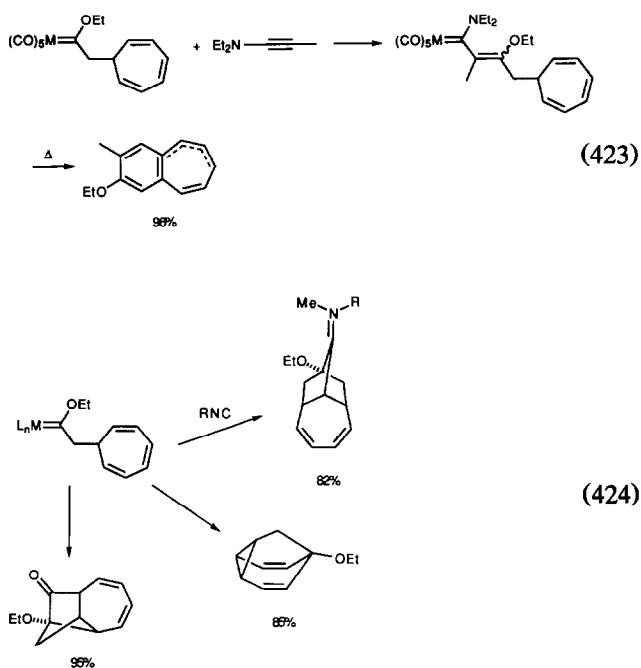


Carbene complexes underwent a series of "metathesis cascade" reactions with polyunsaturated substrates (eqn. (420) [465], eqn. (421) [466], and eqn. (422) [467]).



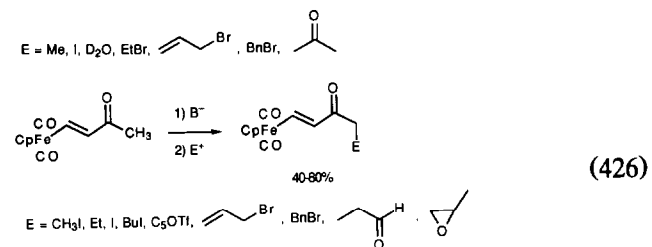
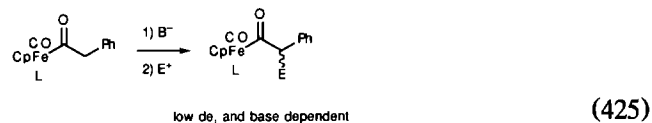


Other unusual reactions of carbene complexes are shown in eqn. (423) [468] and eqn. (424) [469].



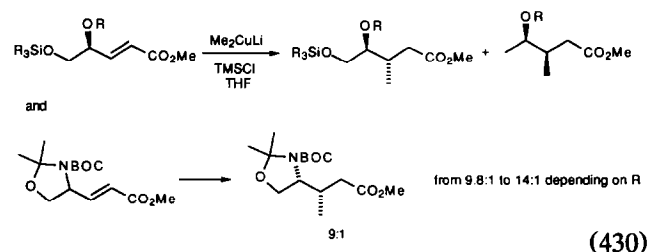
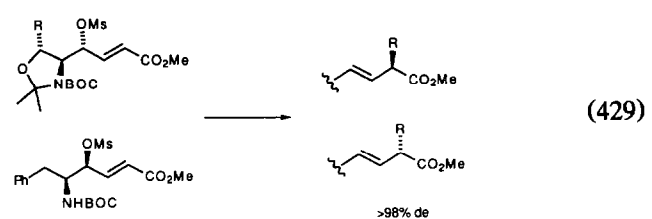
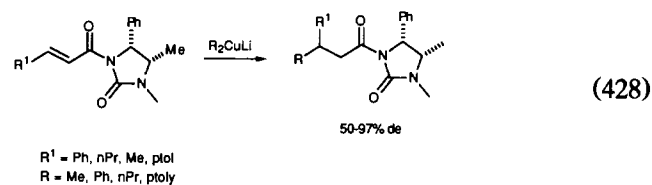
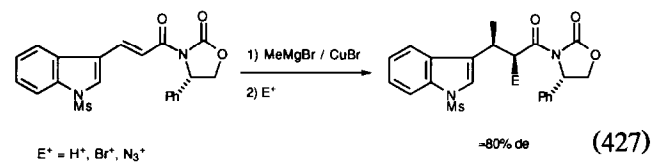
2.1.14. Alkylation of metal acyl enolates

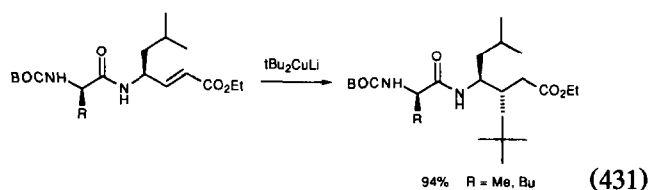
Only two examples were reported this year (eqn. (425) [470] and eqn. (426) [471]).



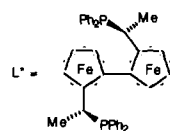
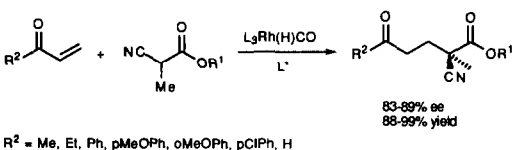
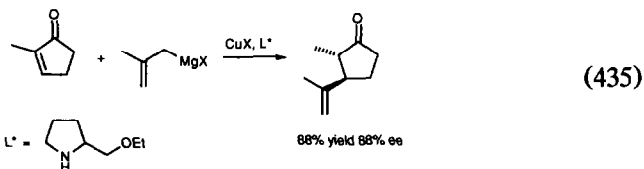
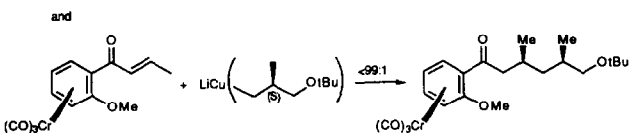
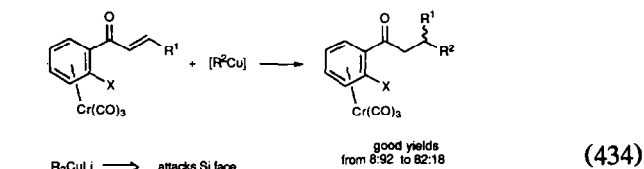
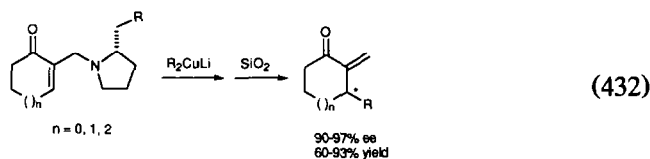
2.2. Conjugate addition

Organocuprates continued to be the reagents of choice for conjugate additions, and a review of this important class of reagents has appeared (1695 references) [472]. Asymmetric conjugate addition has been reviewed (124 references) [473] and has been achieved using chiral conjugated amides (eqn. (427) [474], eqn. (428) [475]), asymmetric γ -alkoxy substrates (eqn. (429) [476], eqn. (430) [477,478]), and asymmetric γ -alkyl substrates (eqn. (431) [479]).

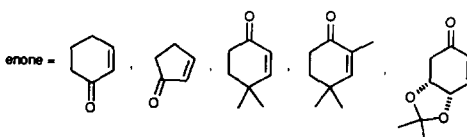
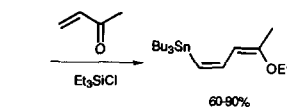
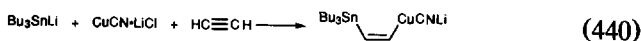
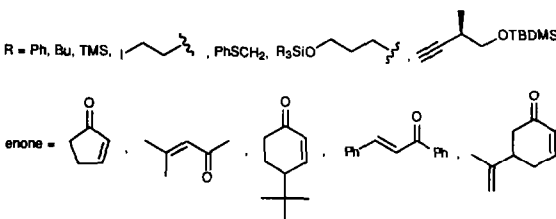
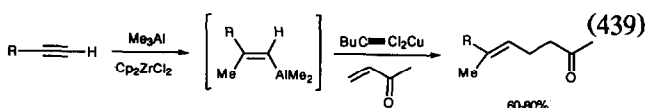
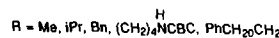
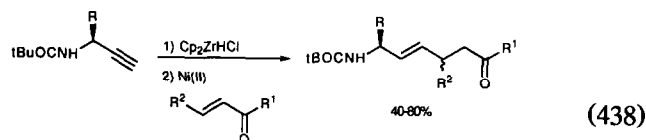
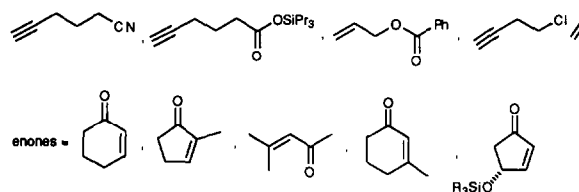
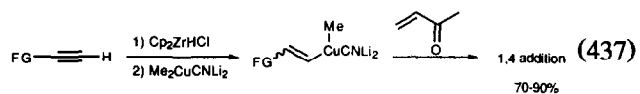


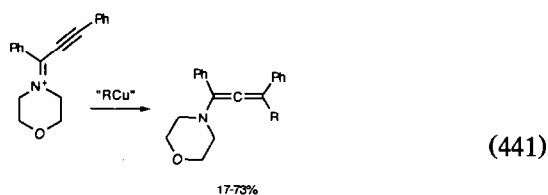


Asymmetric induction from more remote centers was also observed (eqn. (432) [480], eqn. (433) [481], and eqn. (434) [482]). Chiral ligands were also used to induce asymmetry (eqn. (435) [483] and eqn. (436) [484]).



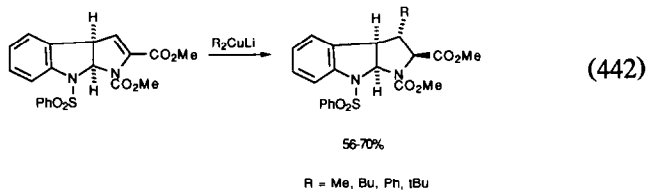
Vinyl cuprates, generated from hydrozirconation of alkynes (eqn. (437) [485], eqn. (438) [486], eqn. (439) [487]) and stannylcupration of alkynes (eqn. (440) [488]), underwent conjugate addition reactions. Cuprates added 1,4 to propargyl iminium ions (eqn. (441) [489]).



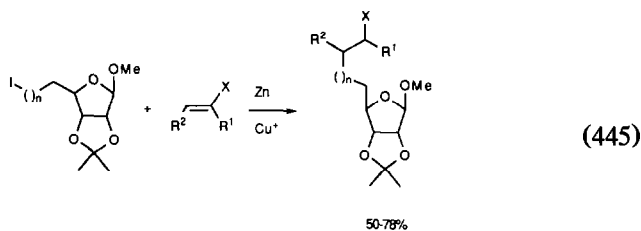
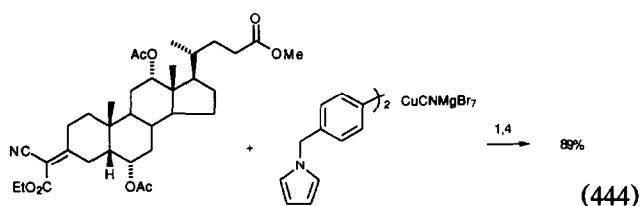
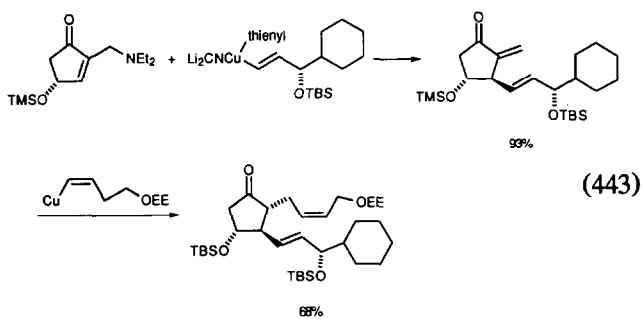


R = nBu, secBu, tBu, $\text{CH}_2=\text{CH}-$, Ph, $\text{PhC}\equiv\text{C}-$, Ph_3Sn , Ph_3Si , Ph_2tBuSi

Cuprates added in a 1,4 manner to a number of complex systems (eqn. (442) [490], eqn. (443) [491], eqn. (444) [492], and eqn. (445) [493]).

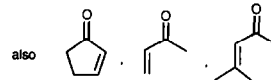
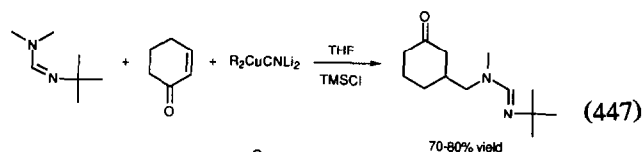
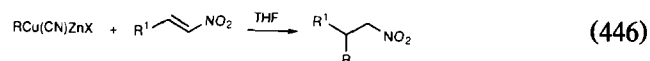


R = Me, Bu, Ph, tBu

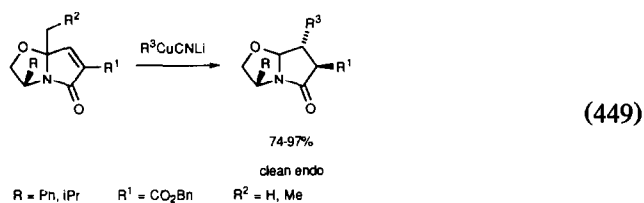


X = CN, CONH₂, CO₂Me R¹ = H, Me, NHAc R² = H, Me

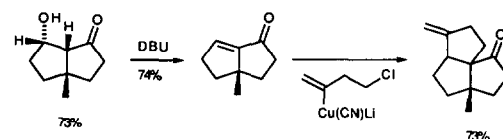
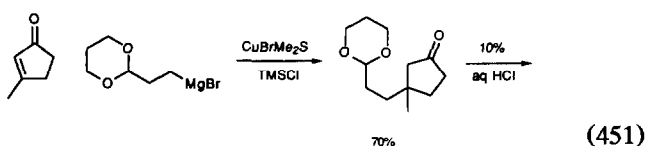
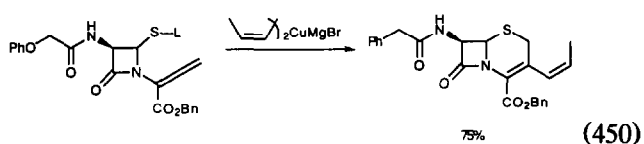
cis and *trans* enones underwent conjugate addition to give the same product (eqn. (448) [496]).



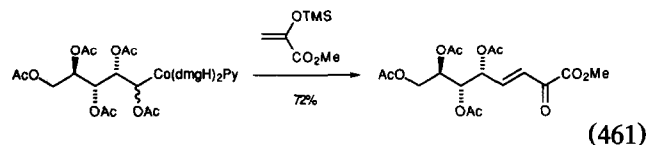
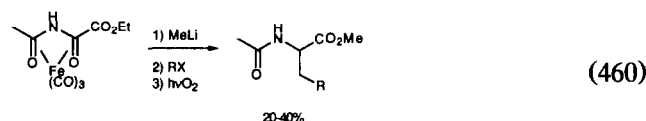
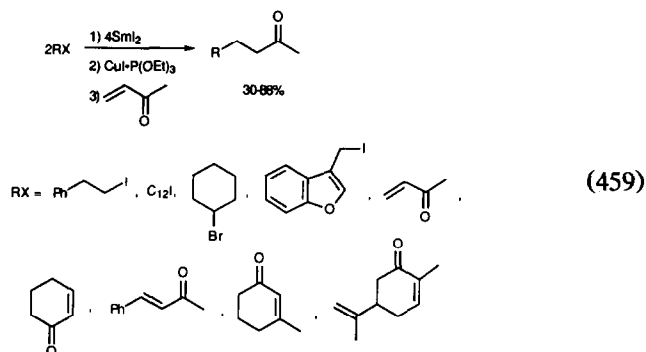
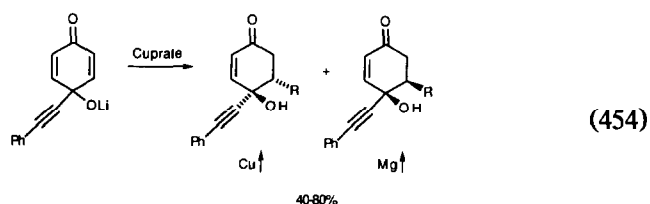
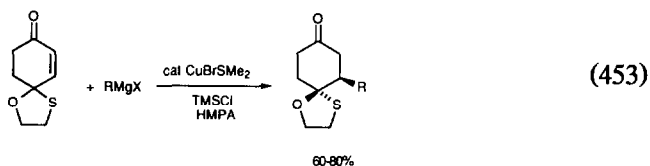
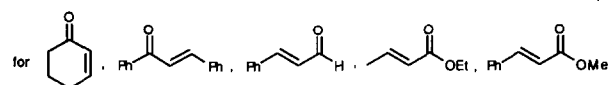
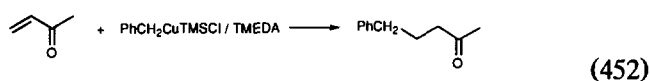
Other useful conjugate additions are reported in eqn. (449) [497], eqn. (450) [498], eqn. (451) [499], eqn. (452) [500], eqn. (453) [501], and eqn. (454) [502].



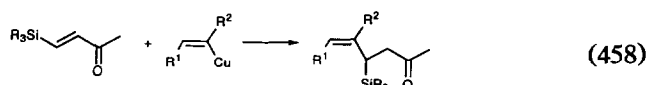
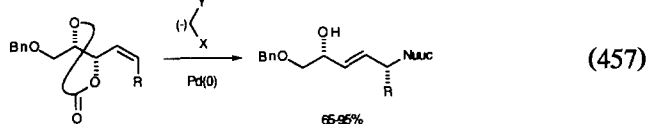
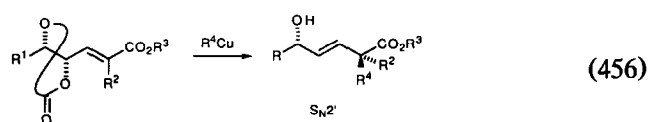
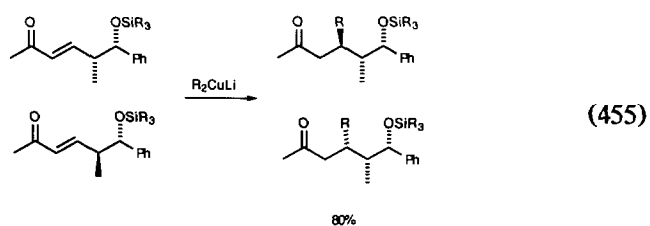
R = Ph, iPr R¹ = CO₂Bn R² = H, Me



Cuprates alkylated the β -position of nitroalkenes (eqn. (446) [494]). Cuprates catalyzed the conjugate addition of amidines to enones (eqn. (447) [495]). Both



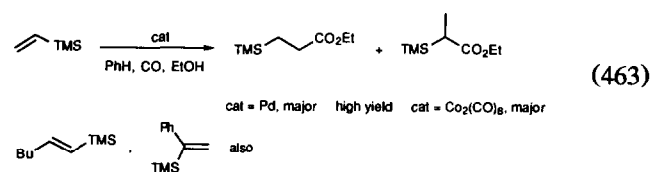
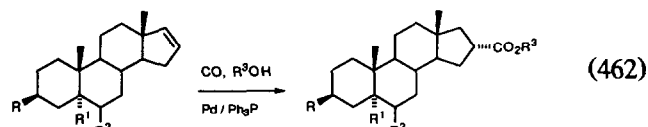
Cuprates added mainly *anti* to γ -alkyl enones (eqn. (455) [503], eqn. (456) [504] and eqn. (457) [505]). Cuprates added to β -silylenones (eqn. (458) [506]). Organic halides added 1,4 to enones in the presence of samarium iodide and copper iodide (eqn. (459) [507]). Iron complexes of α -acetamidoacrylates were alkylated in a 1,4-manner (eqn. (460) [508]). Cobalt alkyls β -alkylated enones (eqn. (461) [509]).

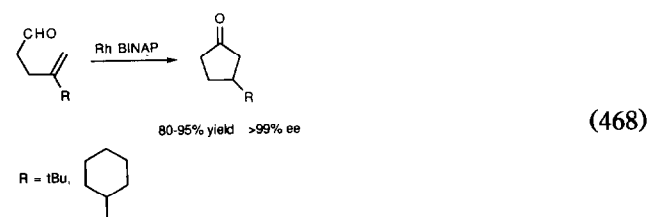
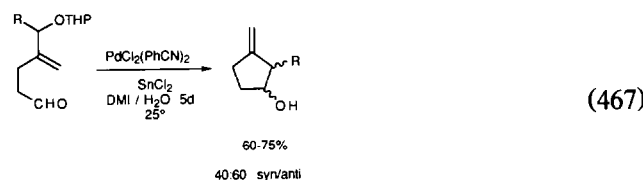
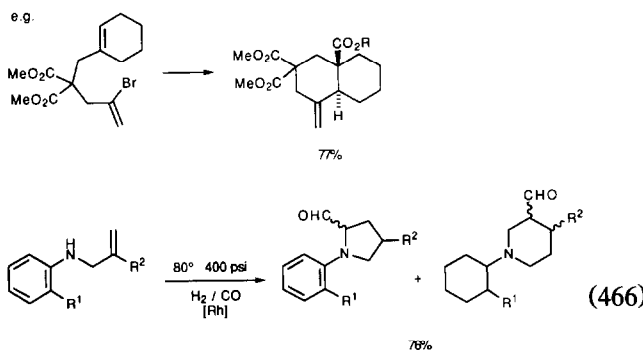
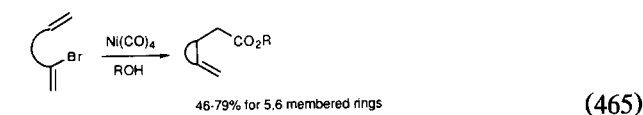
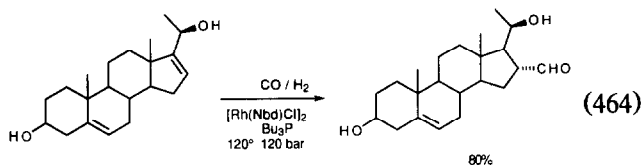


2.3. Acylation reactions (excluding hydroformylation)

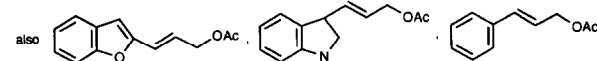
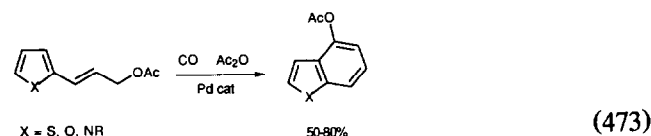
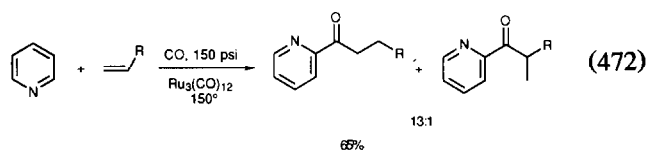
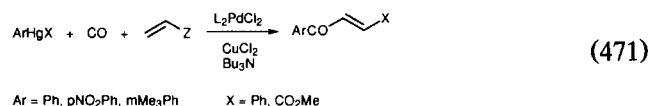
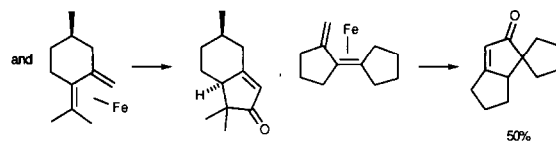
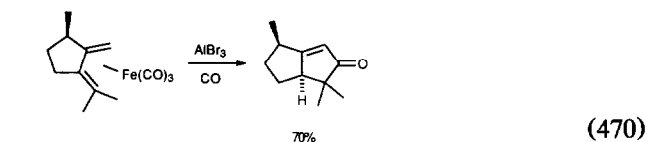
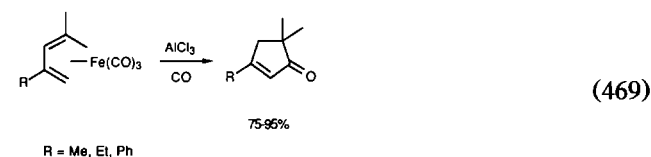
2.3.1. Carbonylation of alkenes and arenes

Androstene was hydroalkoxycarbonylated in the presence of palladium phosphine catalysts (eqn. (462) [510]). Vinyl silanes were carbonylated to α or β -silyl esters, depending on the catalyst (eqn. (463) [511]). Allylic alcohols were stereospecifically carbonylated using rhodium(I) catalysts (eqn. (464) [512]). Nickel carbonyl cyclocarbonylated ω -olefinic vinyl halides (eqn. (465) [513]). Allyl amines were cycloaminoacylated using rhodium catalyst (eqn. (466) [514]) (for a review with 36 references see [515]). Palladium (eqn. (467) [516]) and rhodium (eqn. (468) [517]) catalyzed the intramolecular acylation of alkenes by aldehydes.



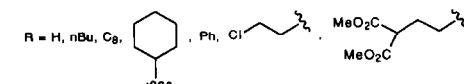
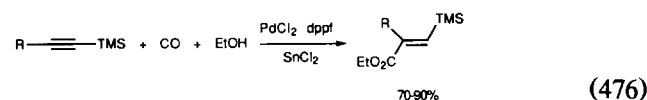
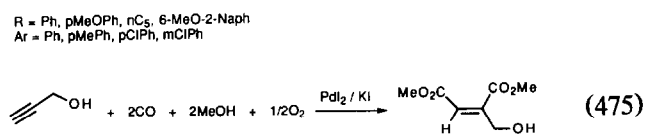
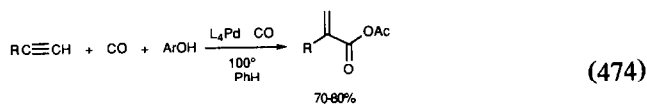


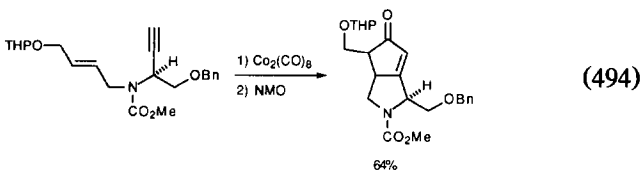
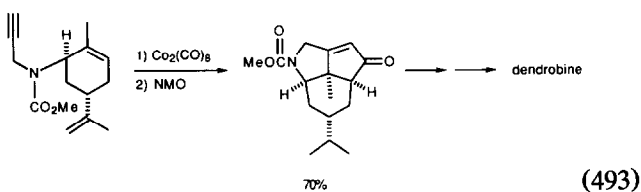
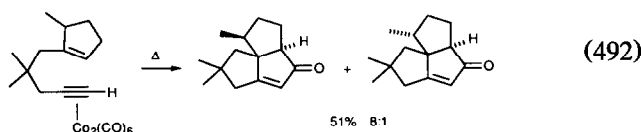
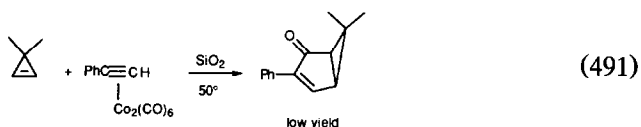
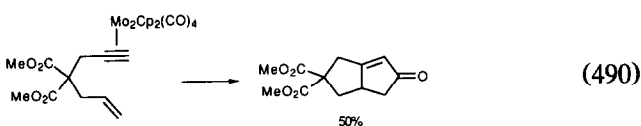
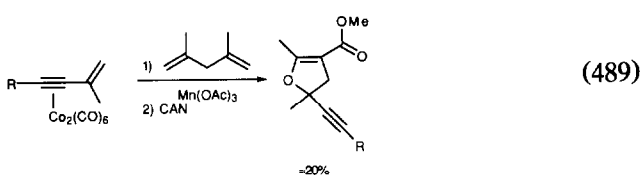
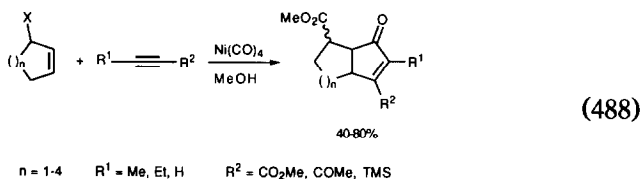
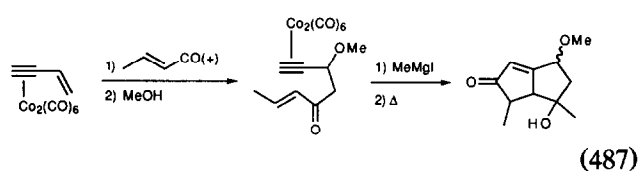
Treatment of iron diene complexes with aluminum halides produced cyclopentenones (eqn. (469) [518] and eqn. (470) [519]). Chromium complexes of polycyclic aromatic hydrocarbons were carboethoxylated with ethyl chloroformate [520]. Aryl mercuric halides were carbonylatively vinyllated using palladium catalysts (eqn. (471) [521]). Pyridine (eqn. (472) [522]) and other arenes (eqn. (473) [523]) were also carbonylated using transition metal catalysts.



2.3.2. Carbonylation of alkynes (including the Pauson-Khand reaction)

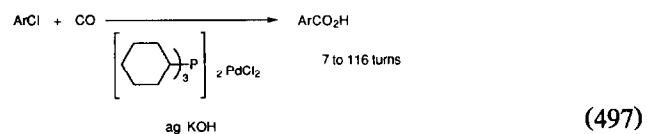
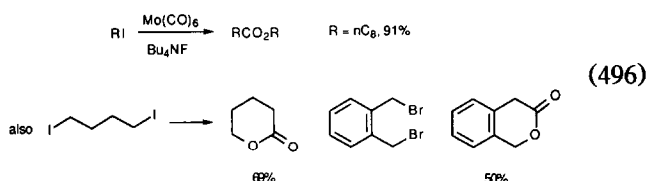
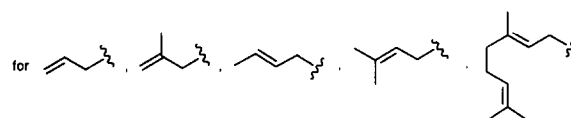
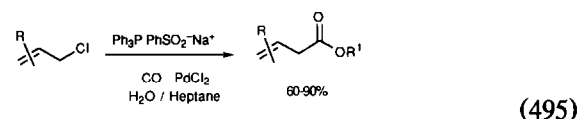
Palladium (eqn. (474) [524], eqn. (475) [525], and eqn. (476) [526]), rhodium (eqn. (477) [527]) and nickel complexes (eqn. (478) [528]) all catalyzed the carbonylation of alkynes.



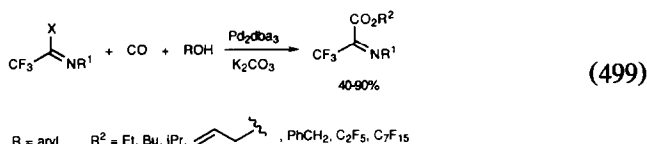
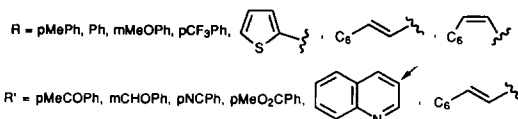


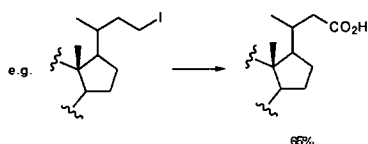
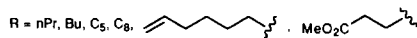
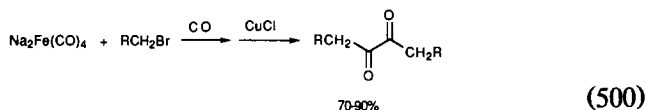
2.3.3. Carbonylation of halides and triflates

Palladium complexes catalyzed the carbonylation of allylic chlorides (eqn. (495) [548]). Polyvinylpyrrolidone-supported palladium catalysts were more efficient for this process [549]. Aliphatic iodides were carbonylated by molybdenum hexacarbonyl (eqn. (496) [550]). Aryl chlorides (eqn. (497) [551]) and iodides [552] were carbonylated using palladium catalysts. Dicobalt octacarbonyl carbonylated 1,4-bis(chloromethyl) benzene and allyl chloride [553]. Palladium(0) catalyzed the carbonylation of iodides and fluorosilanes to ketones (eqn. (498) [554]), and the carbonylation of iminoyl chlorides (eqn. (499) [555]). Collmans reagent coupled halides to give diketones (eqn. (500) [556]). Ruthenium complexes oxidized iodides to acids (eqn. (501) [557]).

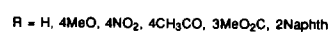
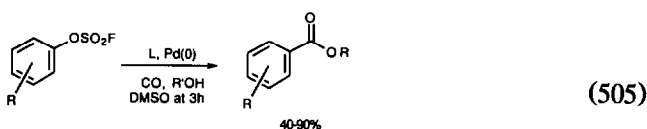
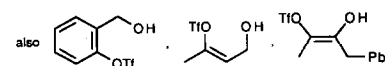
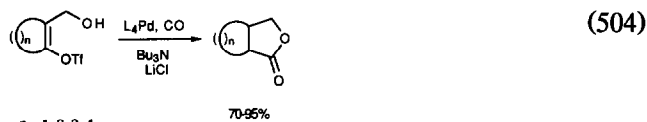
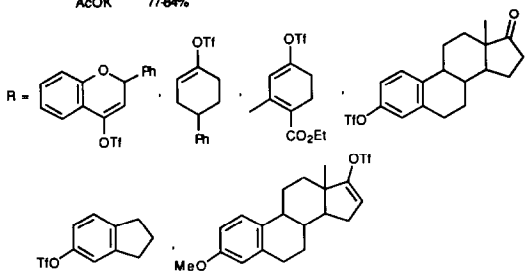
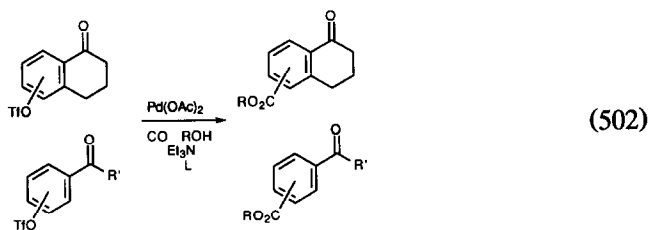


Ar = Ph, 1-Naphth, pMePh, mMePh, oMePh, pMeOPh, mPhCOPh, nCF₃Ph // pNO₂Ph fails



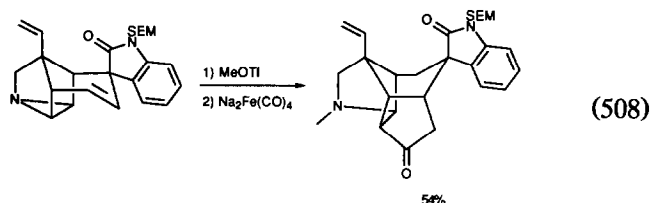
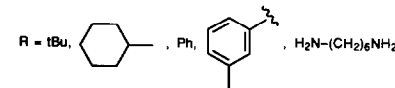
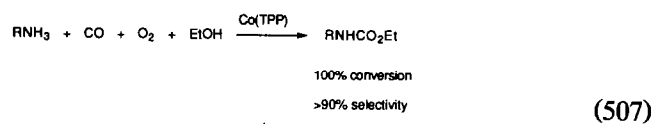
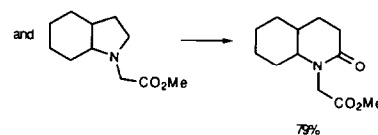
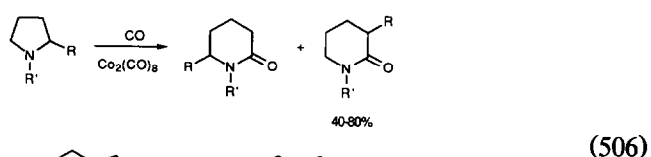


Palladium(0) complexes catalyzed the carbonylation of aryl (eqn. (502) [558]) and vinyl (eqn. (503) [599]), eqn. (504) [560], triflates and fluorosulfonates (eqn. (505) [561]).



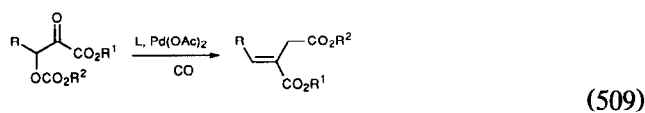
2.3.4. Carbonylation of nitrogen compounds

Catalytic carbonylation of nitrogen-containing organic compounds has been reviewed (41 references) [562]. Palladium catalyzed the synthesis of diphenyl urea from nitrobenzene, aniline and carbon monoxide [563] the carbonylation of N-chloroamines to carbamoyl chlorides [504] and the acylation of anilines by aryl halides [565]. Cobalt carbonyl catalyzed the unusual conversion of pyrrolidines to piperidones (eqn. (506) [566]). Cobalt porphyrins catalyzed the N-carbonylation of amines (eqn. (507) [567]). An unusual carbonylation is seen in eqn. (508) [568].

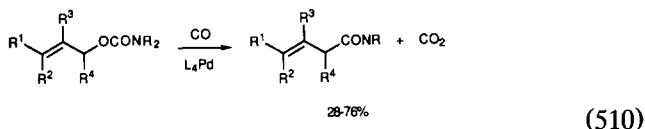


2.3.5. Carbonylation of oxygen compounds

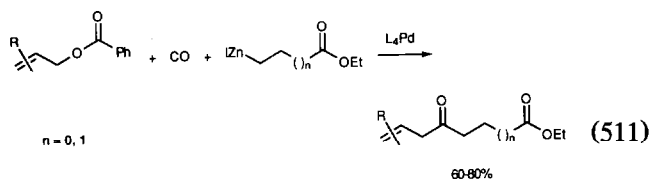
Palladium catalyzed the carbonylation of allylic alcohols in the presence of lithium chloride and titanium isopropoxide [569], the carbonylation of allyl carbonates (eqn. (509) [570]), allyl carbamates (eqn. (510) [571]) and allyl carboxylates (eqn. (511) [572] and eqn. (512) [573]). Benzyl alcohols were carbonylated by rhodium(I) catalysts [574]. "Transition metal complex catalyzed new carbonylation of oxiranes, acetals, and allylic acetates using N-silyl amines" was the title of a review (12 references) [575]. An example is seen in eqn. (513) [576].



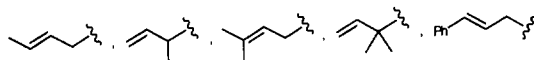
R = Me, Et, iPr, tBu
 R¹ = Me, tBu, Et, iPr, CONMe₂
 R² = Me, Et



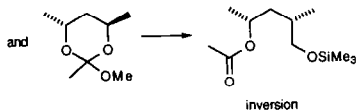
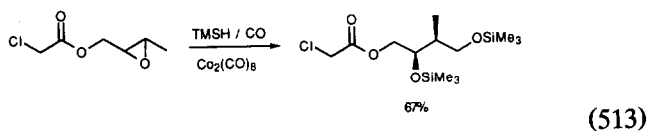
R¹ = H, ,
 R² = H, Me, tBu
 R³ = H, Me, CO₂Me
 R⁴ = Et, tBu



n = 0, 1

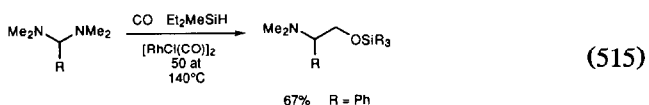
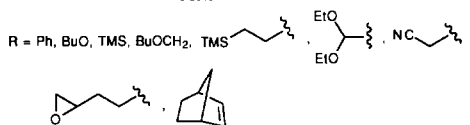


X = OAc, Cl, OCO₂Me R = Ph, nPr

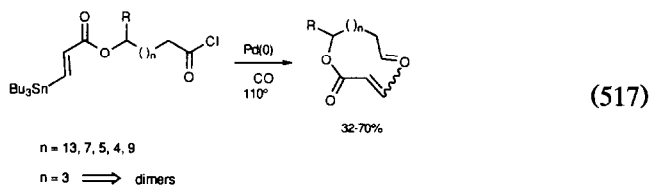
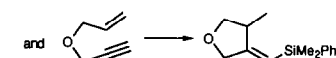
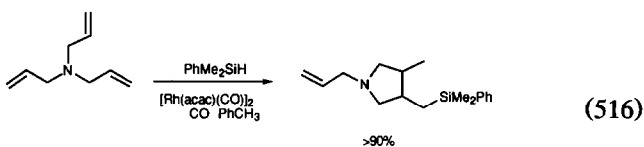


2.3.6. Miscellaneous carbonylations

The synthesis of carbonyl compounds utilizing organomanganese pentacarbonyl complexes was the topic of a dissertation [577]. A review dealing with the synthesis of C-2 compounds directly from synthesis gas *via* organometallic catalysts has appeared (118 references) [578]. Alkenes (eqn. (514) [579]) and amines (eqn. (515) [580] and eqn. (516) [581]) underwent catalyzed silylcarbonylation. ω -Stannyl acid chlorides were cyclocarbonylated by palladium catalysts (eqn. (517) [582]).



67% R = Ph

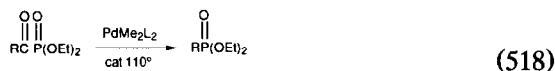


n = 13, 7, 5, 4, 9

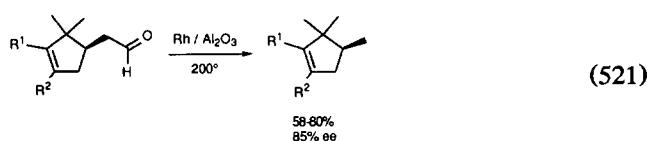
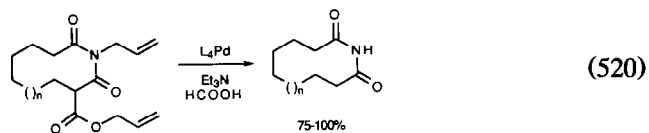
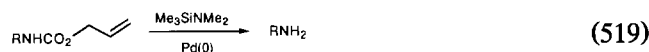
n = 3 \Rightarrow dimers

2.3.7. Decarbonylation reactions

Acylophosphates (eqn. (518) [583]), allyl carbamates (eqn. (519) [584]), and allyl carbonates (eqn. (520) [585]) were deacylated by palladium(0) catalysts. Rhodium on alumina deformed aldehydes (eqn. (521) [586]).



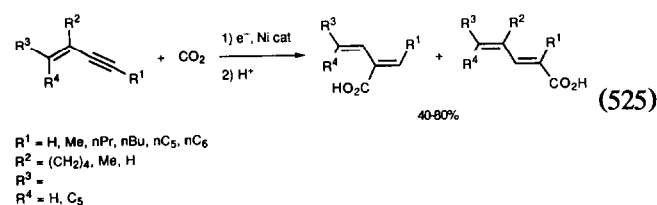
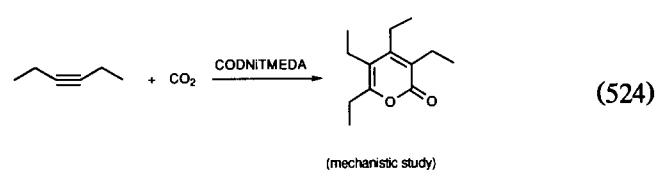
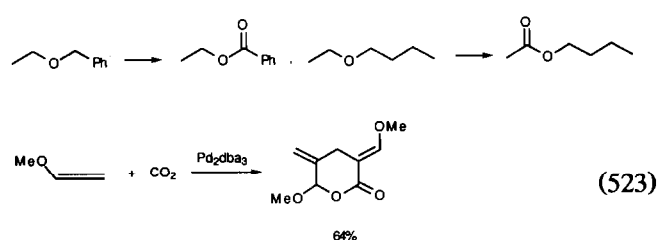
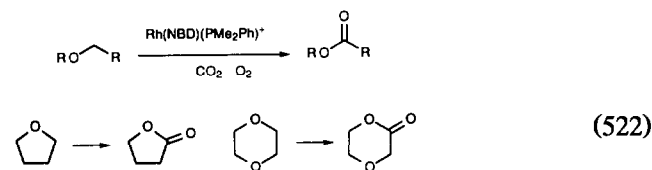
R = Ph, 100%; ptoyl, 100%; pMeOPh, 72%; pClPh, 35%; Me, 55%; Et, 9%



58-80%
85% ee

2.3.8. Reactions of carbon dioxide

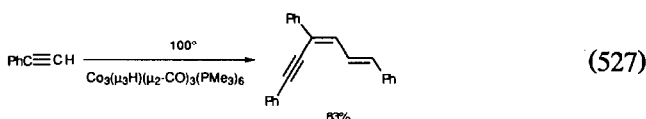
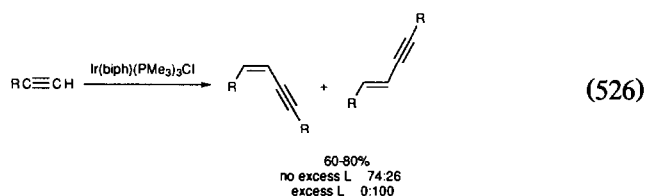
A review dealing with carbon dioxide in organic synthesis and polymer synthesis by means of transition metal complex catalysts has appeared (20 references) [587]. η^6 -Arene chromium tricarbonyl complexes were lithiated, then carboxylated with carbon dioxide [588]. Unsaturated carbamates, carbonates, and ureas were synthesized by ruthenium(II) catalyzed reactions of propargyl alcohols [589]. Ethers were converted to esters by CO_2 in the presence of a rhodium(I) catalyst (eqn. (522) [590]). Methoxy allene was cocyclotrimerized with CO_2 in the presence of palladium catalysts (eqn. (523) [591]). A review dealing with the synthesis of unsaturated lactones using carbon monoxide with transition metals (22 references) has appeared [592]. An example is seen in eqn. (524) [593]. Enynes were carboxylated by CO_2 in the presence of nickel catalysts (eqn. (525) [594]).



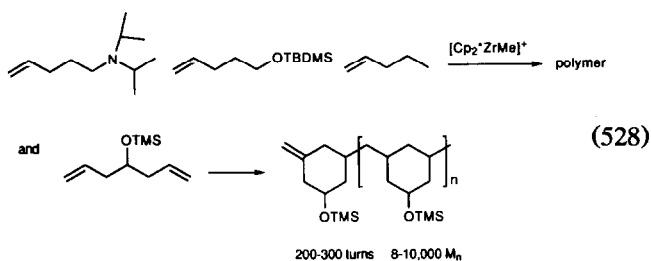
2.4. Oligomerization (including cyclotrimerization of alkynes and metathesis polymerization)

A review dealing with ethylene dimerization and oligomerization to α olefins (194 references) has appeared [595] as has one dealing with dimerization of ethene to 2-butene and subsequent metathesis [596].

Nickel(0) complexes in the presence of phosphines dimerized acrylonitrile to 2,4-dicyanobutene [597,598]. Vinyl ketones were head-to-tail dimerized using modified rhodium catalysts [599]. Conjugated dienes were linearly dimerized by nickel(0) aminophosphonite systems [600]. Iridium complexes dimerized terminal alkynes (eqn. (526) [601]) while cobalt complexes trimerized phenylacetylene (eqn. (527) [602]).

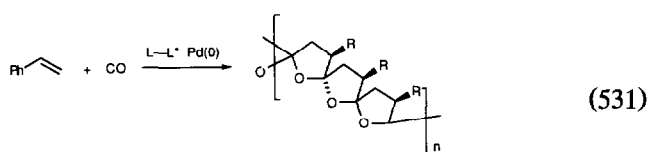
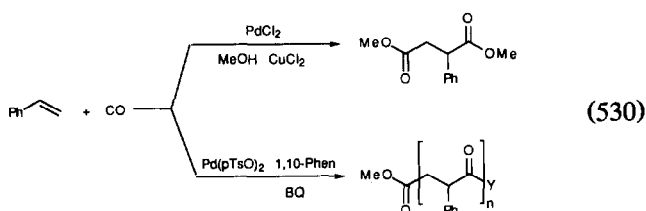
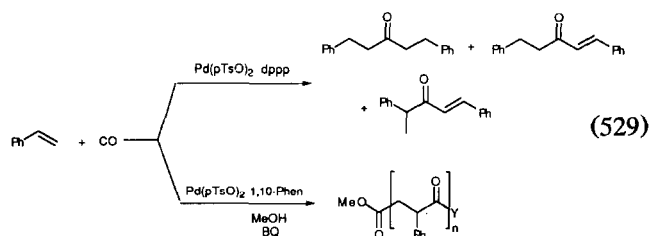


Developing new, specific catalysts for Ziegler-Natta polymerization continued to be a very active area. The catalyst $[(\text{indenyl})_2\text{TiMe}]^+$ catalyzed ethylene polymerization and its activity decreased with decreasing solvent polarity [603]. The related catalysts $[\text{Cp}'_2\text{M-Me}]^+$ when $\text{M} = \text{Ti} \ll \text{HF} < \text{Zr}$ and $\text{Cp}' = \text{C}_5\text{H}_4\text{TMS}$ also polymerized ethylene [604]. Silicon bridged indenylzirconium and silylated cyclopentadienylzirconium complexes catalyzed isotactic polymerization of propylene, butene, pentene and hexene [605,606]. The cationic complex $[\text{Cp}'_2\text{ZrH}_2]^+\text{B}(\text{C}_6\text{H}_5)_3^-$ was an active olefin polymerization catalyst [607]. Stereoselective propene polymerization was achieved using cholestanylindene-derived non-bridged group 4 bent metallocene catalysts [608]. *trans*-1,2-Bis-indenylcyclohexane zirconium dichloride catalyzed propene polymerization with high stereoselectivity even at 50°C [609]. Functionalized alkenes could also be Ziegler-Natta polymerized (eqn. (528) [610]).

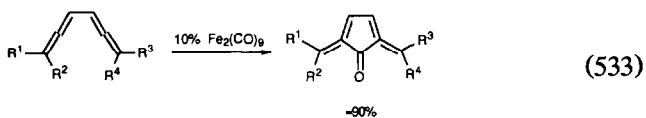
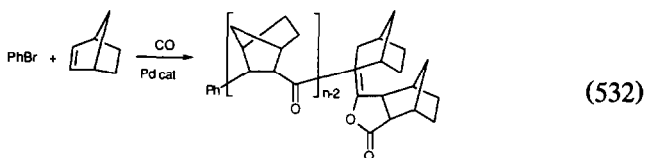


The mechanism of rhodium(I) catalyzed alternating cooligomerization of carbon monoxide with alkenes has been studied [611], and ways to control the regioselectivity

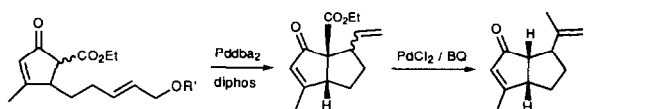
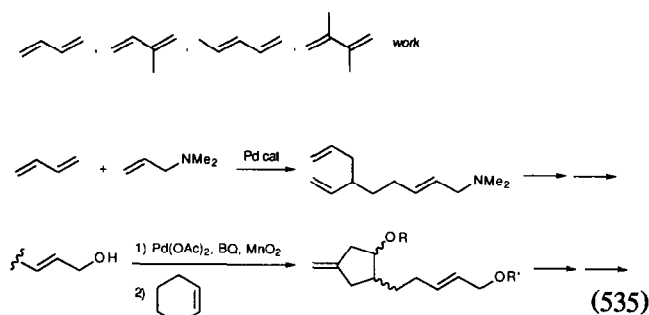
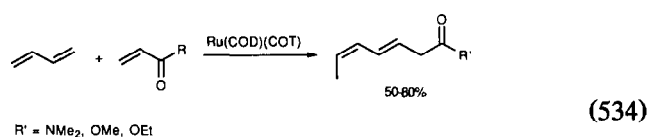
tivity of palladium catalyzed copolymerization of propene and carbon monoxide have been developed [612]. The complex $[(\text{bipy})\text{Pd}(\text{CH}_3)(\text{MeCN})]^+$ is a living catalyst for the copolymerization of carbon monoxide with 4-*t*-butylstyrene [613]. Unusual palladium catalyzed cooligomerizations of alkenes with carbon monoxide are seen in eqn. (529) [614], eqn. (530) [615], eqn. (531) [616], and eqn. (532) [617]. Iron carbonyl cyclocarbonylated bis allenes (eqn. (533) [618]).



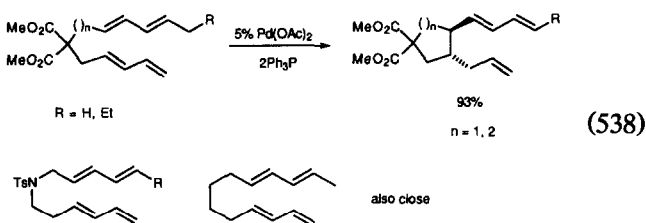
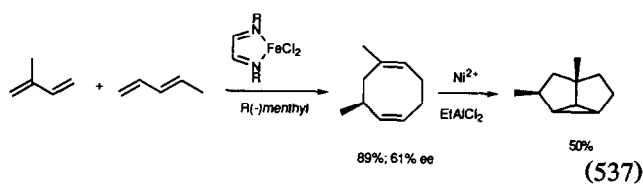
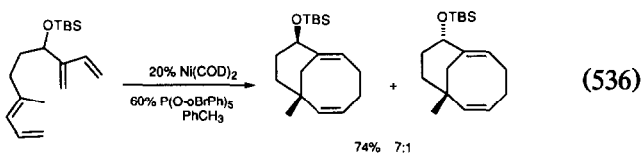
$\text{LL}^* = (\text{S})\text{-}(6,6'\text{-dimethylbiphenyl-2,2'}\text{-diyl})\text{bis}(\text{dicyclohexylphosphine})$

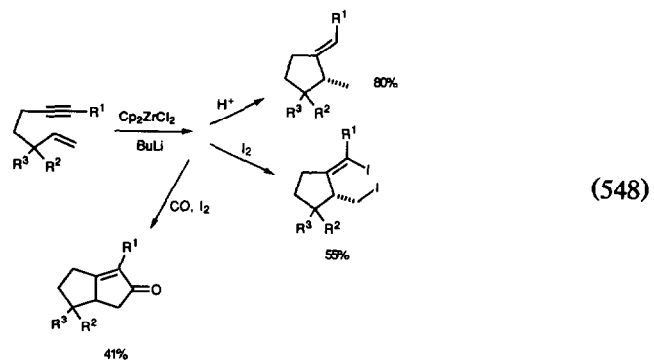
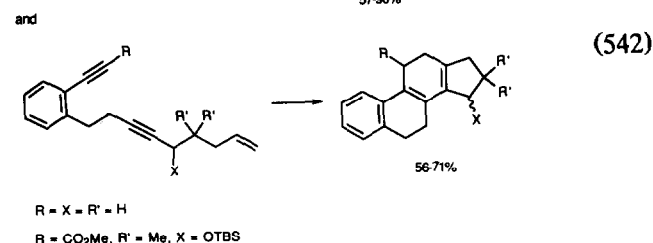
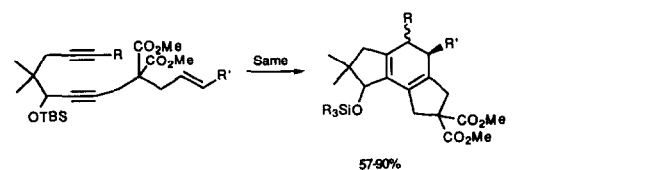
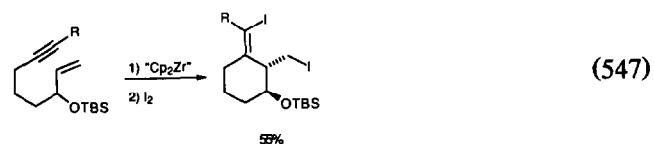
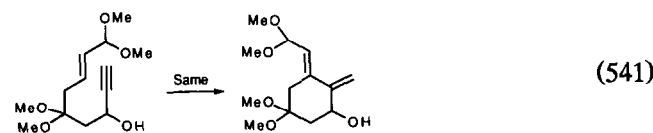
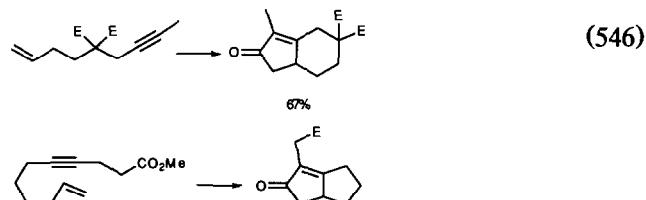
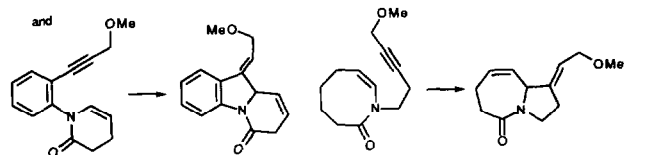
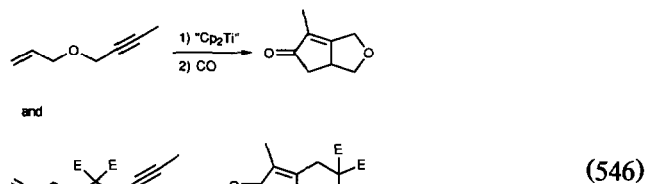
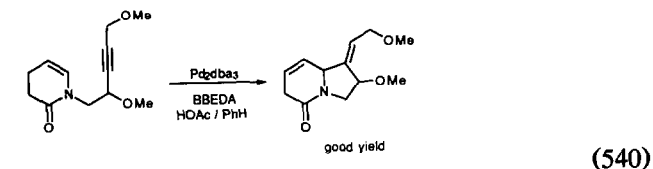
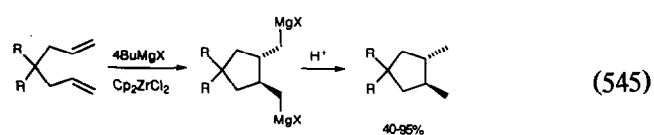
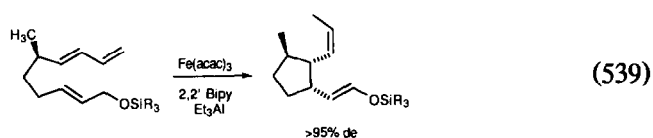


Palladium complexes catalyzed the telomerization of isoprene with primary amines [619] and *N*-substituted tosamides [620]. Ruthenium codimerized butadiene with conjugated enones (eqn. (534) [621]). Palladium was used extensively in the synthesis shown in eqn. (535) [622].

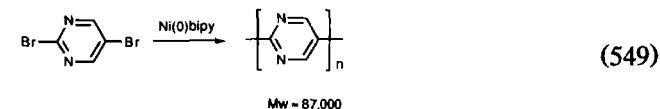
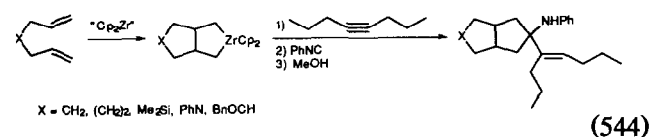
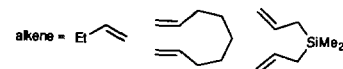
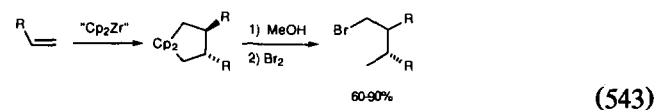


Butadiene was cyclotrimerized by $\text{TiCl}_4/\text{Et}_x\text{AlCl}_{3-x}$ systems [623]. Nickel(0) complexes cyclodimerized butadienes (eqn. (536) [624]) as did iron azadiene complexes (eqn. (537) [625]). Palladium acetate cyclodimerized tetraenes (eqn. (538) [626]) while iron complexes cyclodimerized alkenes with dienes (eqn. (539) [627]). Palladium complexes in acetic acid cyclodimerized enynes (eqn. (540) [628], eqn. (541) [629], and eqn. (542) [630]).



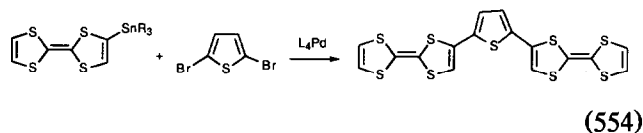
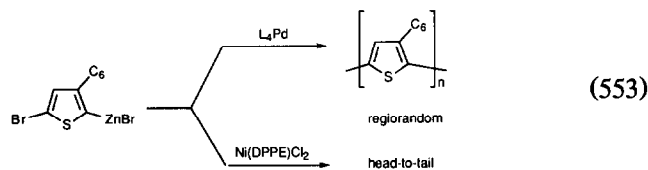
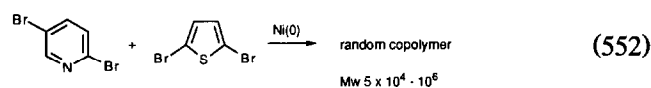
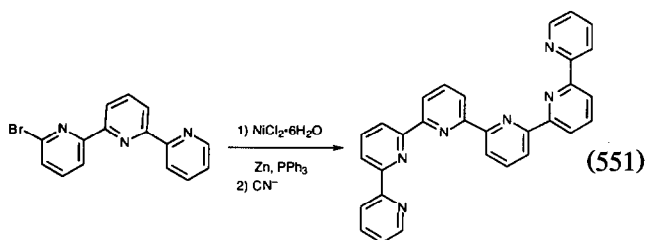
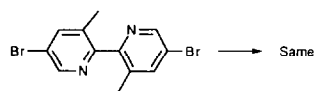
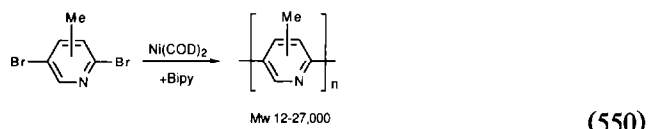


“Zirconocene” cyclodimerized alkenes (eqn. (543) [631], eqn. (544) [632] and eqn. (545) [633]) while both “titanocene” (eqn. (546) [634]) and “zirconocene” (eqn. (547) [635] and eqn. (548) [636]) cocyclized alkenes and alkynes.

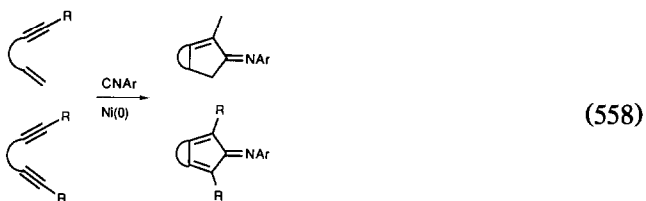
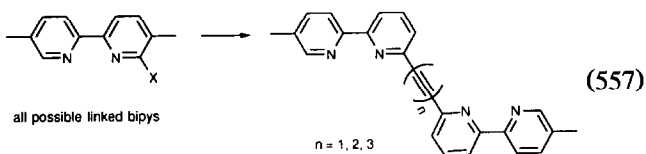
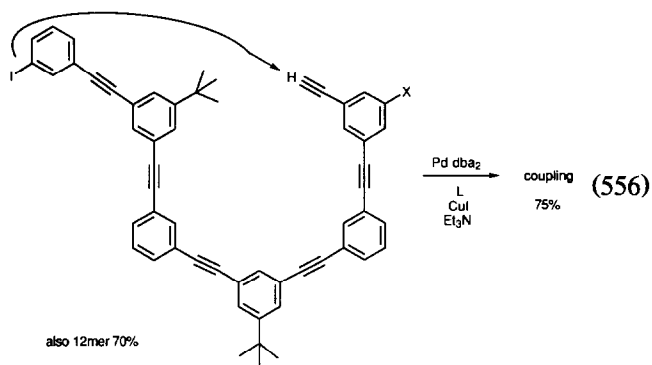
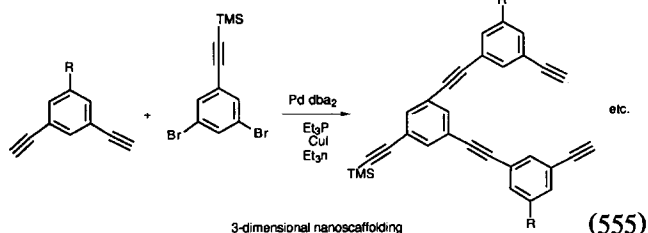


Mw = 87,000

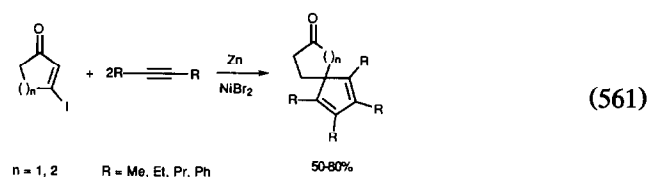
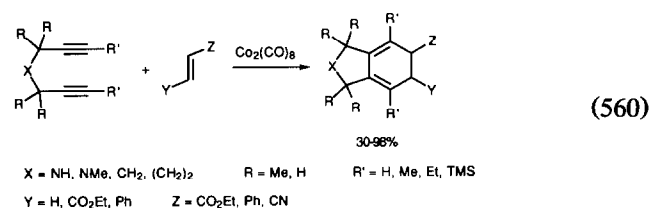
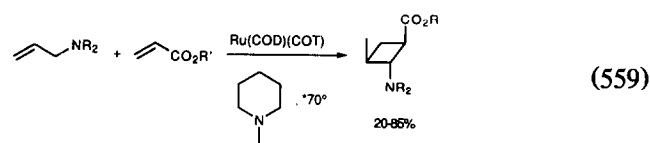
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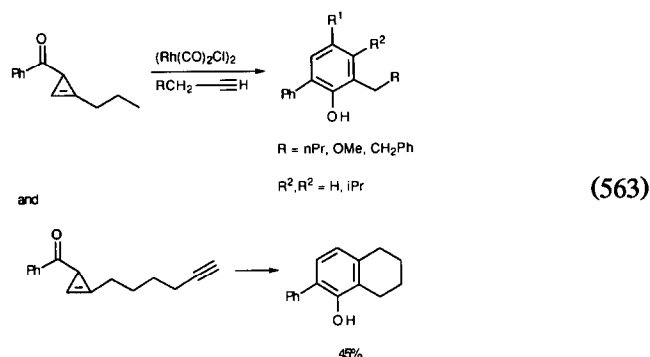
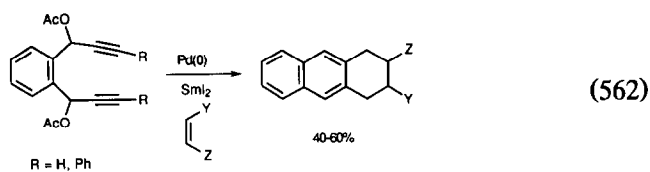


Palladium catalyzed the coupling of arenes with alkynes (eqn. (555) [643]), (eqn. (556) [644]), and (eqn. (557) [645]). Nickel(0) complexes catalyzed the cyclootrimerization of enynes and isocyanides (eqn. (558) [646]).

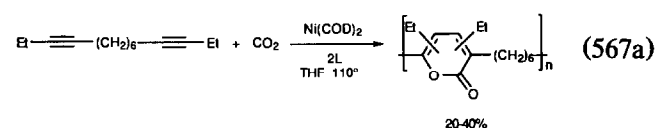
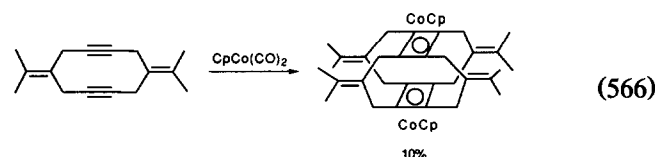
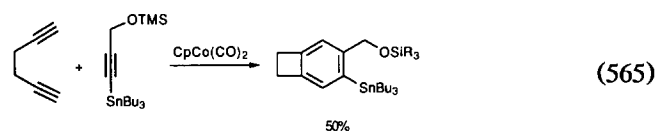
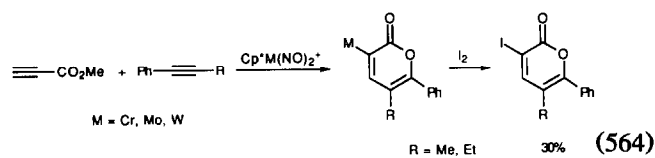


Ruthenium catalyzed the codimerization of acrylates with allyl amines (eqn. (559) [647]). Cobalt catalyzed the codimerization of diynes with alkenes (eqn. (560) [648]), as did nickel (eqn. (561) [649]). Palladium cyclodimerized alkynes (eqn. (562) [650]). Rhodium complexes cooligomerized cyclopropenes with alkynes (eqn. (563) [651]).

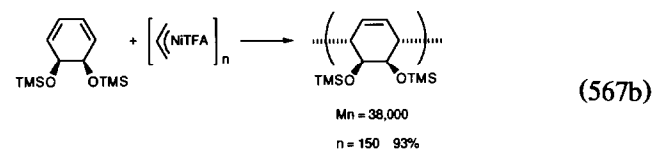




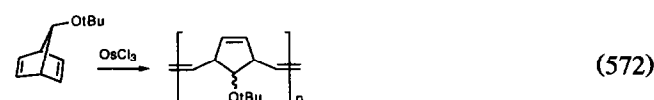
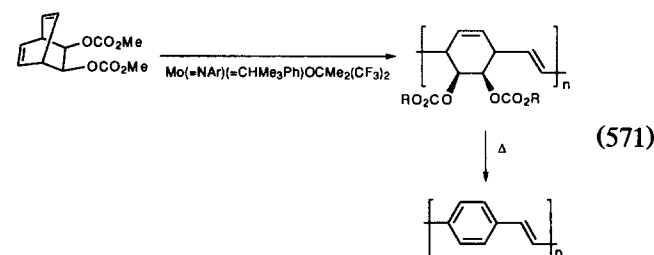
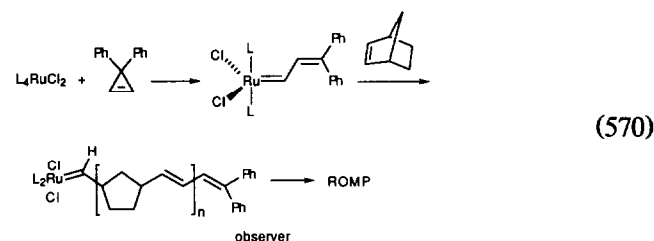
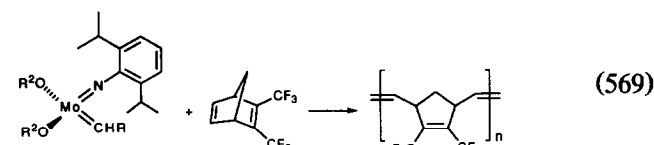
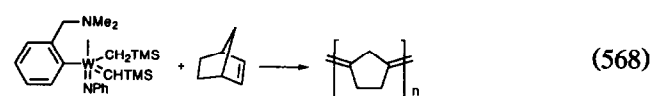
Group 6 complexes cocyclooligomerized alkyne (eqn. (564) [652]) as did cobalt (eqn. (565) [653]). Cycloaddition reactions of alkyne in organic synthesis was the topic of a review (12 references) [654], as was "Cycloalkadiynes – from bent triple bonds to strained Cage compounds" (109 references) [655]. Cobalt cyclooligomerized diynes (eqn. (566) [656]). Nickel(0) complexes cocyclooligomerized carbon dioxide and diynes (eqn. (567a) [657]). "3 + 2 Cycloaddition reactions of transition metal 2-alkynyl and η^1 -allyl complexes and their utilization in five membered ring compound synthesis" was the title of a review (63 references) [658].



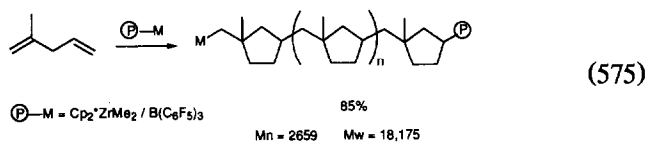
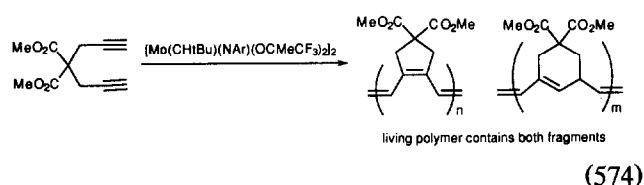
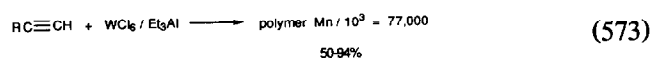
Cationic nickel allyl complexes catalyzed the clean 1,4-*cis* polymerization of butadiene [659] while cationic ruthenium diene complexes catalyzed trimerization of butadienes to C₁₂ linear oligomers [660]. π -Allylnickel trifluoroacetate polymerized *cis*-dihydroxycyclohexadiene (eqn. (567b) [661]), while reduced nickel species catalyzed the cooligomerization of sorbic acid esters with butadiene [662]. Palladium complexes catalyzed the cyclooligomerization of quadacyclane with norbornene [663].



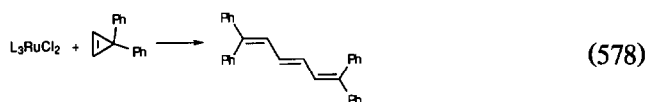
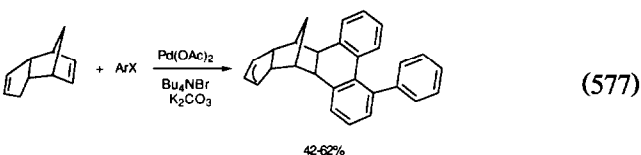
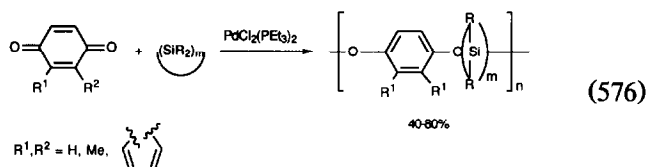
A number of new ROMP catalyst systems have been developed, and many will tolerate functionality (eqn. (568) [664], eqn. (569) [665], eqn. (570) [666], eqn. (571) [667], and eqn. (572) [668]).



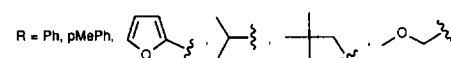
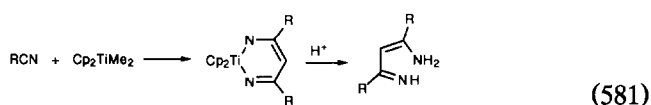
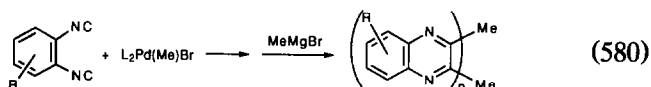
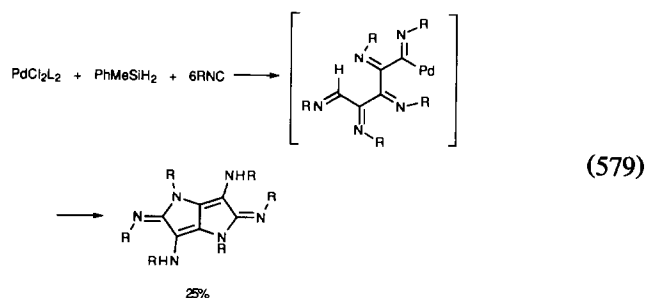
Alkynes were polymerized by tungsten (eqn. (573) [669]) and molybdenum catalysts (eqn. (574) [670]). Cationic zirconium complexes cyclooligomerized 1,4-dienes (eqn. (575) [671]).



Group 6 metal carbonyls polymerized terminal alkynes but metathesized internal ones [672]. Nickel(0) complexes catalyzed the oligomerization of methylene cyclopropane [673]. Substituent effects in nickel catalyzed olefin oligomerization have been studied [674]. Palladium catalysts cooligomerized quinones with polysilanes (eqn. (576) [675]) and polyarylated norbornenes (eqn. (577) [676]). Ruthenium complexes dimerized diphenylcyclopropene (eqn. (578) [677]).



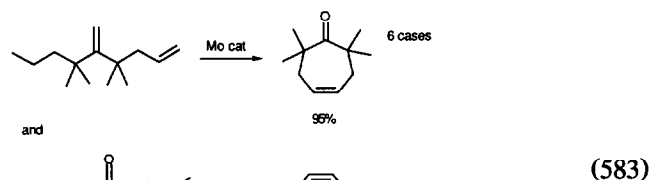
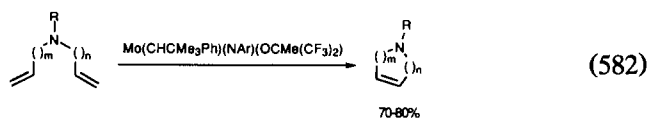
Stereoselection in nickel-catalyzed polymerization of chiral isocyanides was studied [678]. Palladium catalysts cooligomerized isonitriles and silanes (eqn. (579) [679]) and converted bis isonitriles into helical oligomers (eqn. (580) [680]). Dimethyl titanocene dimerized nitriles (eqn. (581) [681]).



2.5. Rearrangements

2.5.1. Metathesis

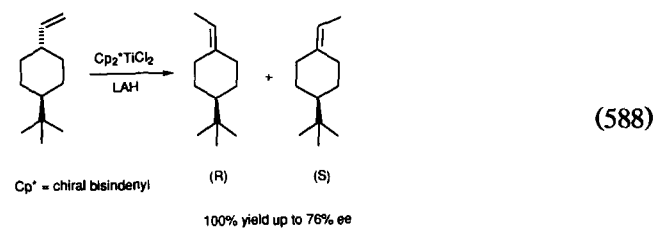
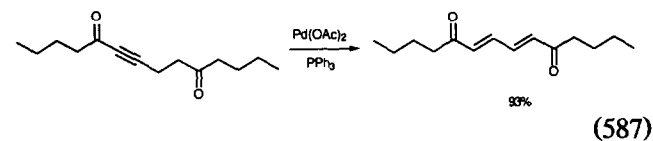
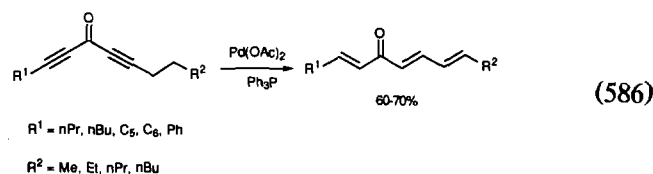
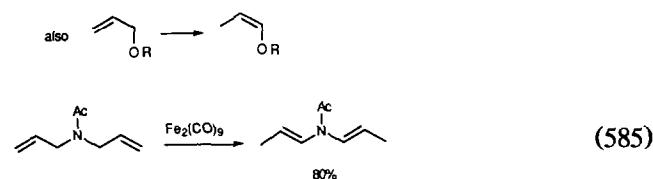
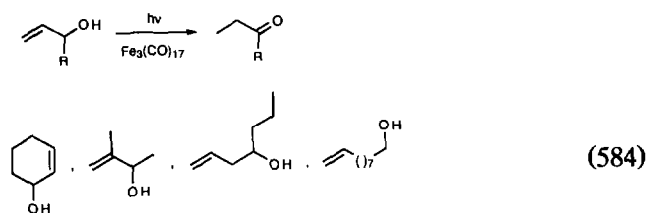
A symposium article on the olefin metathesis reaction has been published [682]. Metathesis of 2-pentene by tungsten carbonyl has been studied [683]. Dinitrosomolybdenum catalysts metathesized functionalized alkenes [684]. Metathesis was used to synthesize heterocycles (eqn. (582) [685]) and carbocycles (eqn. (583) [686]).



2.5.2. Olefin isomerization

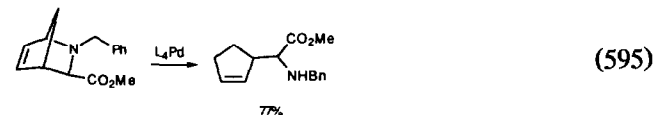
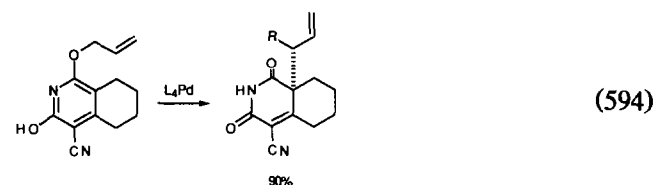
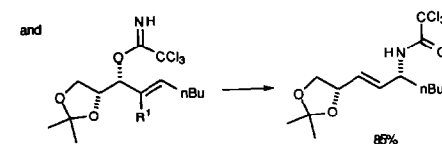
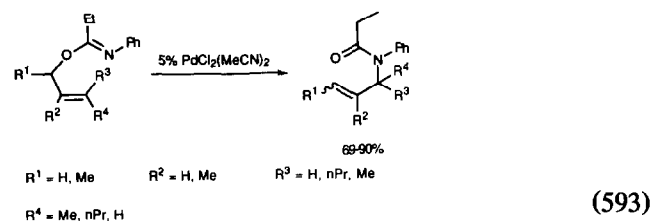
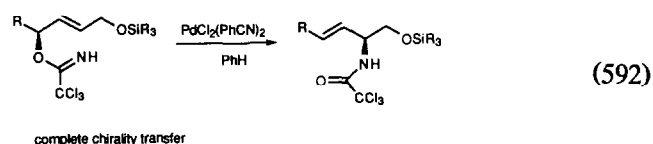
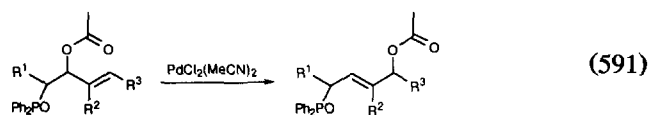
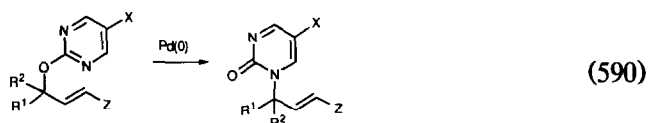
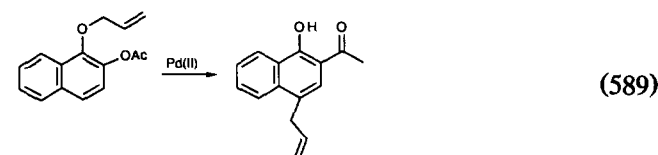
Styrene/divinylbenzene copolymers containing RuCl_2L_3 isomerized 1-hexene [687], as did hexa-aquoruthenium (2+) [688]. Iron carbonyls were used to isomerize alkenes (eqn. (584) [689] and eqn. (585) [690]). Palladium acetate isomerized acylenic ketones (eqn. (586) [691] and eqn. (587) [692]). Chiral ti-

tanocenes isomerized alkenes with reasonable asymmetric induction (eqn. (588) [693]).

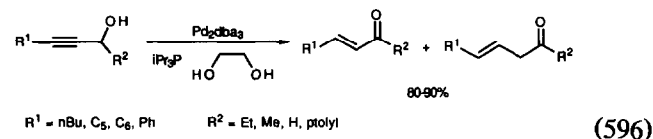


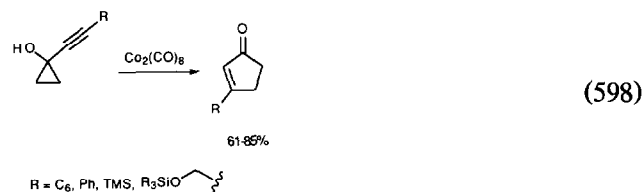
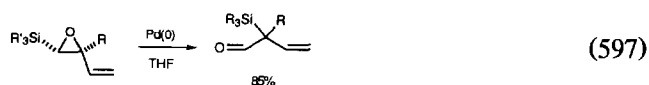
2.5.3. Rearrangement of allylic and propargylic compounds

Palladium(II) complexes catalyzed a large number of allylic transposition rearrangements (eqn. (589) [694], eqn. (590) [695], eqn. (591) [696], eqn. (592) [697], eqn. (593) [698], eqn. (594) [699], and (eqn. (595) [700]).



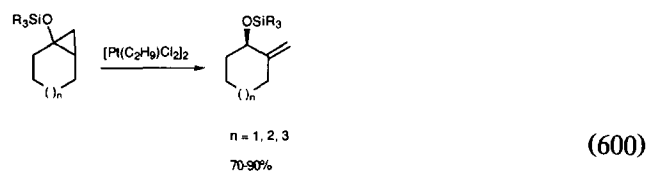
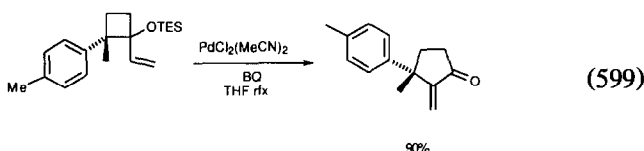
Palladium(0) complexes rearranged propargyl alcohols to conjugated enones (eqn. (596) [701]) and allyl epoxides to unconjugated enones (eqn. (597) [702]). Cobalt carbonyl ring expanded cyclopropyl-propargyl alcohols (eqn. (598) [703]).



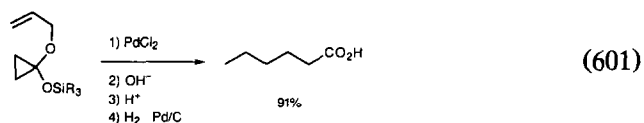
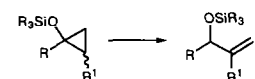


2.5.4. Skeletal rearrangements

Palladium and platinum catalyzed a number of skeletal rearrangements in small ring compounds (eqn. (599) [704], eqn. (600) [705], and eqn. (601) [706]).

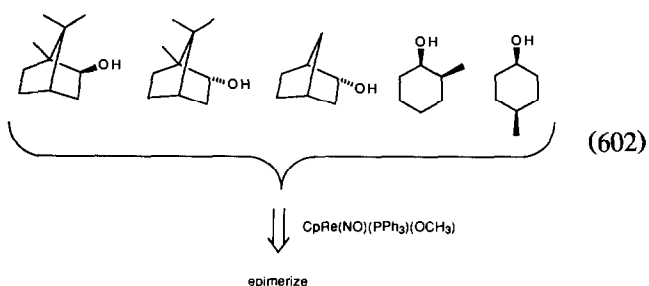


and



2.5.5. Miscellaneous rearrangements

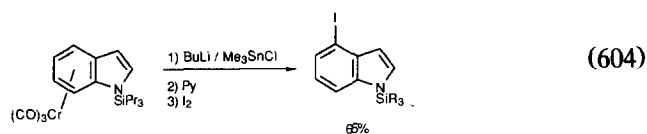
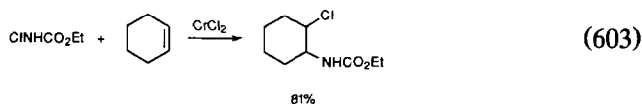
Rhenium complexes epimerized secondary alcohols (eqn. (602) [707]). Rhodium(II) complexes catalyzed the vinylogous Wolff rearrangement [708].



3. Functional group preparations

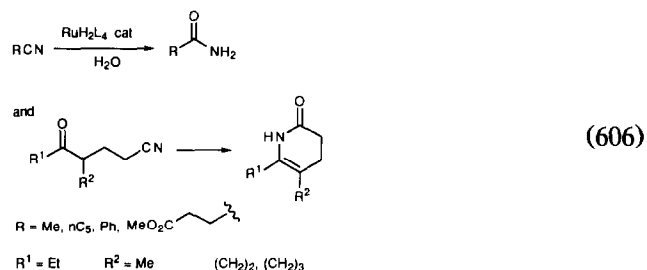
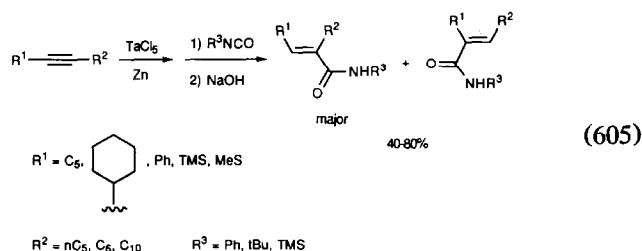
3.1. Halides

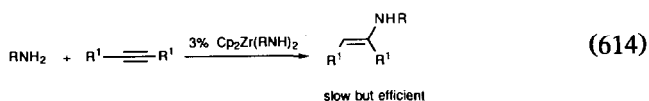
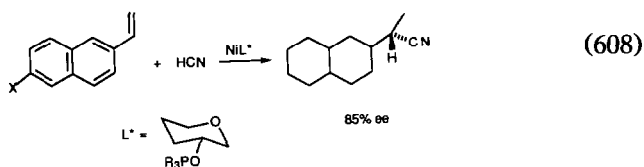
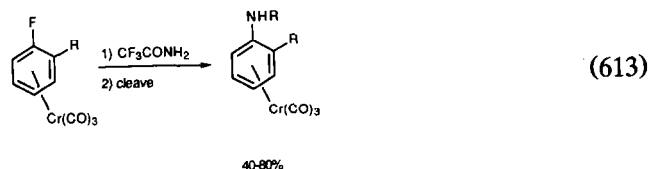
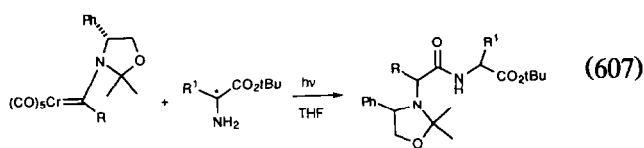
Chromium(II) halides promoted the addition of N-chlorocarbamates to alkenes (eqn. (603) [709]). Wilkinson's complex (Rh(PPh₃)₃Cl) catalyzed the chlorination of adamantane to 1-chloroadamantane in 80% yield, by CH₂Cl₂ [710]. Chromium complexed indole was iodinated (eqn. (604) [711]).



3.2. Amides, nitriles

Reduced tantalum species combined alkynes with isocyanates to produce unsaturated amides (eqn. (605) [712]). Nitriles were converted to amides by ruthenium hydrides (eqn. (606) [713]). E-Alkene isosteric dipeptides were synthesized using copper chemistry (review, 19 references) [714]. Dipeptides were synthesized by photolysis of chromium aminocarbene complexes in the presence of α -amino acid esters (eqn. (607) [715]). Chiral nickel complexes catalyzed the asymmetric hydrocyanation of styrenes (eqn. (608) [716]).

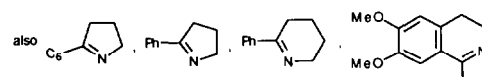
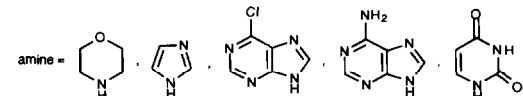
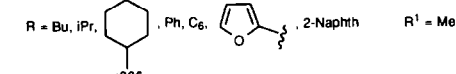
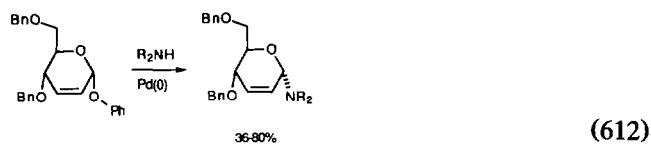
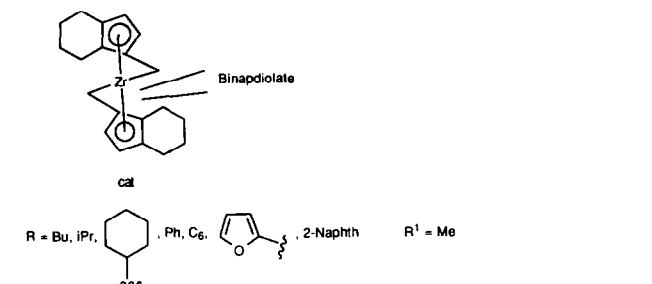
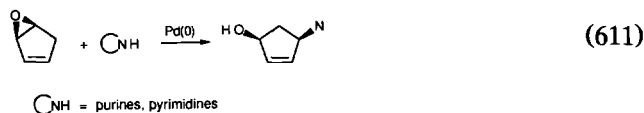
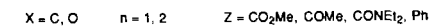
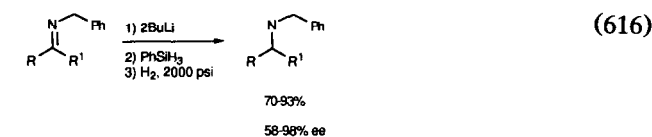
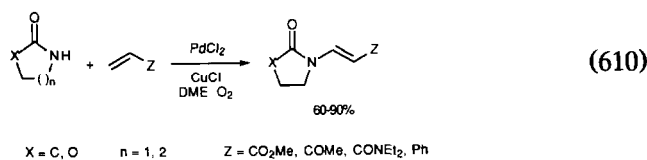
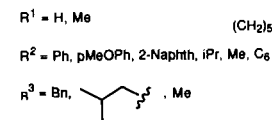
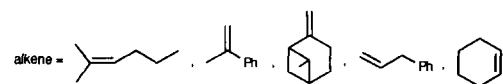
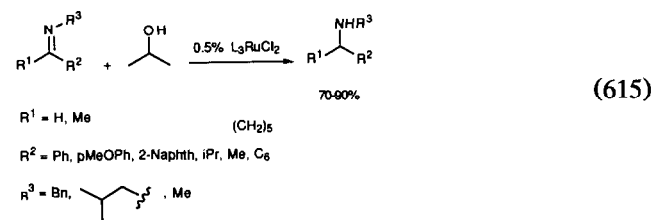
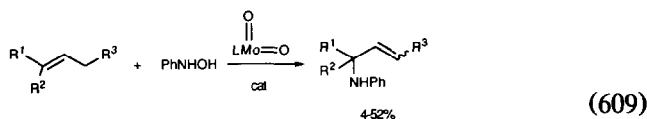


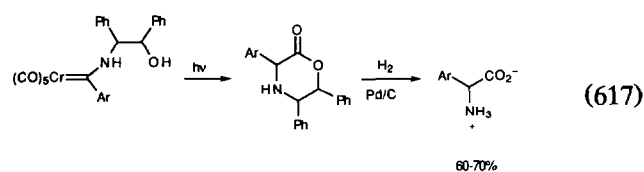


3.3. Amines, alcohols

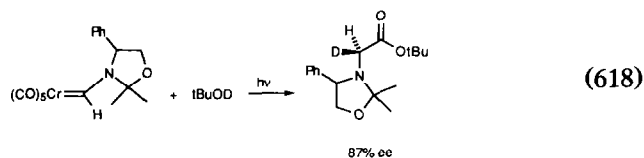
Stereoselective syntheses of acyclic chiral amines by α -amidoalkylation was the topic of a dissertation [717]. Molybdenum oxo species catalyzed the allyl amination of alkenes by hydroxylamines (eqn. (609) [718]). Palladium catalyzed the amination of alkenes (eqn. (610) [719]), allyl epoxides (eqn. (611) [720]), and allyl acetals (eqn. (612) [721]). Fluorobenzene complexes of chromium were aminated by trifluoroacetic acid (eqn. (613) [722]). Zirconium amine complexes aminated alkynes (eqn. (614) [723]).

Ruthenium carbonyl complexes reduced nitroarenes to amines [724]. Ruthenium complexes catalyzed the reduction of imines by *i*-propanol (eqn. (615) [725]). Chiral zirconium complexes catalyzed the asymmetric reduction of imines (eqn. (616) [726]). Aryl glycines were synthesized using chromium carbene photochemistry (eqn. (617) [727]), as was deuteroglycine (eqn. (618) [728]).

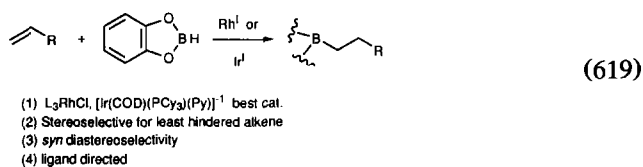




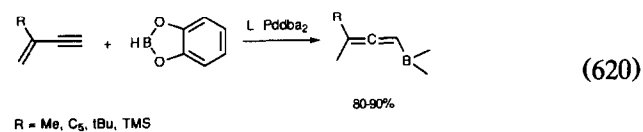
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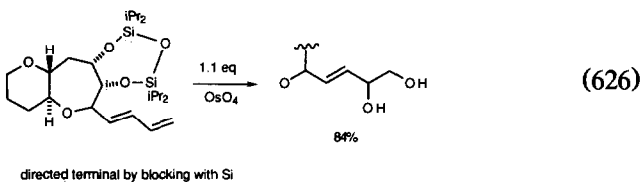
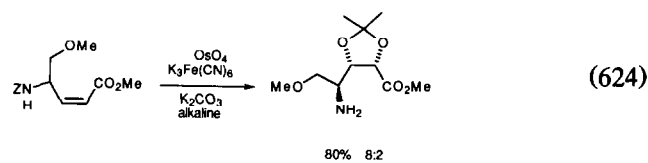
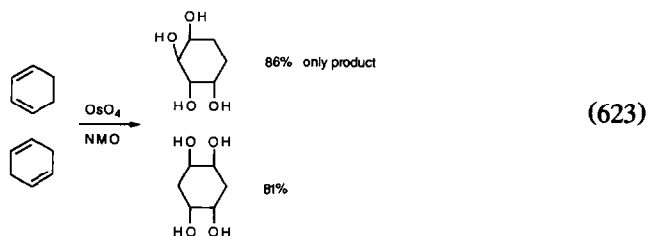
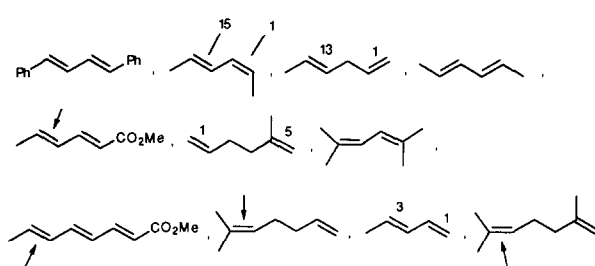
Stereocontrol in catalyzed and uncatalyzed hydroborations has been reviewed (43 references) [729] and was the subject of two mechanistic studies [730,731]. Some key features of the process are given in eqn. (619) [732]. Styrenes underwent catalyzed hydroborations to place the boron α to the aryl group, exclusively [733,734]. Palladium catalyzed the hydroboration of enynes to give allene boranes (eqn. (620) [735]).



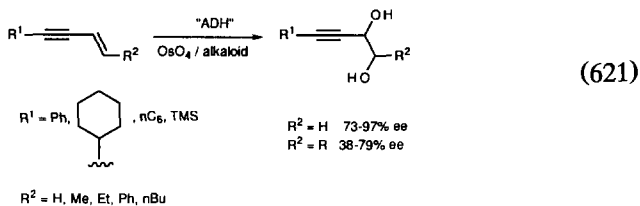
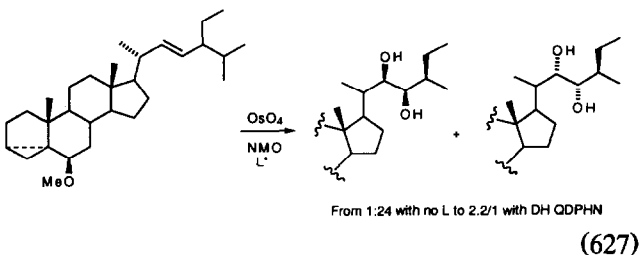
- (1) L₃RhCl, [Ir(COD)(PCy₃)(Py)]⁺ best cat.
- (2) Stereoselective for least hindered alkene
- (3) *syn* diastereoselectivity
- (4) ligand directed



R = Me, C₆, tBu, TMS



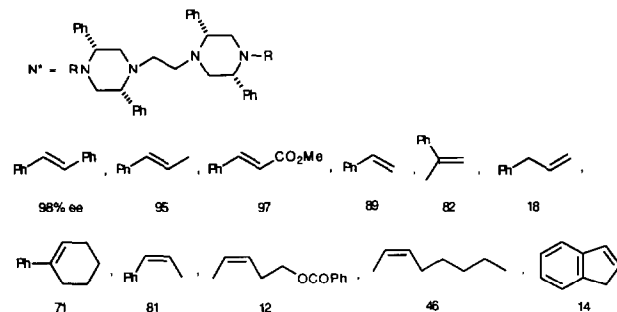
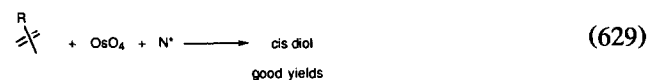
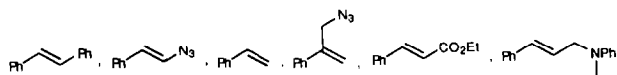
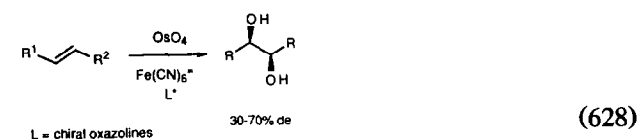
directed terminal by blocking with Si



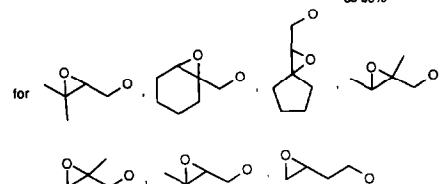
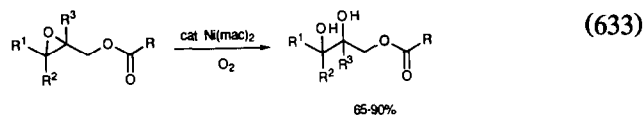
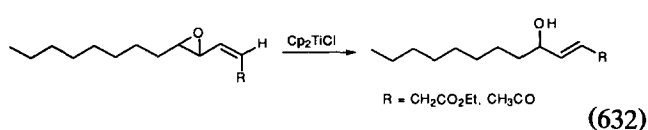
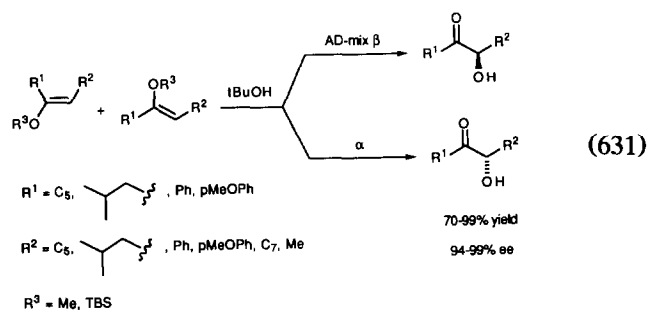
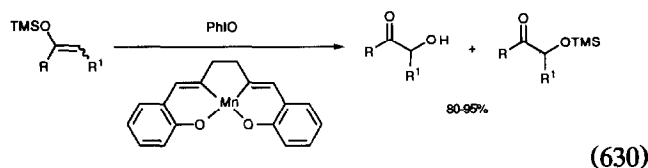
R¹ = Ph, , nC₆, TMS

R² = H 73-97% ee
R² = R 38-79% ee

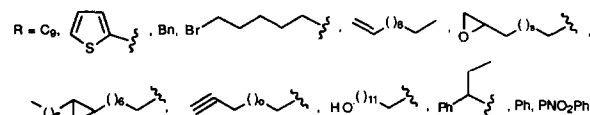
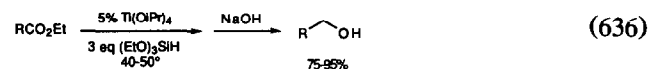
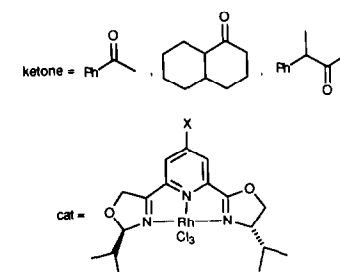
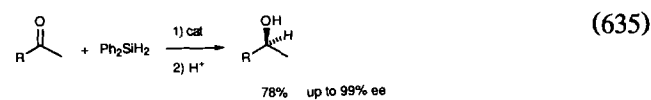
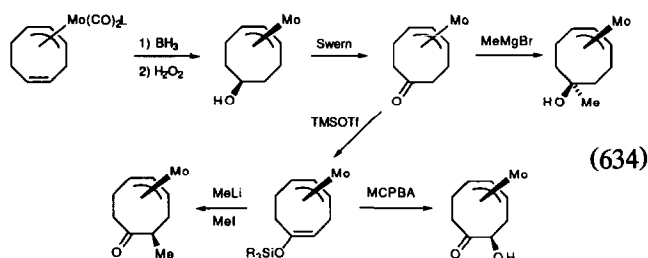
R² = H, Me, Et, Ph, nBu

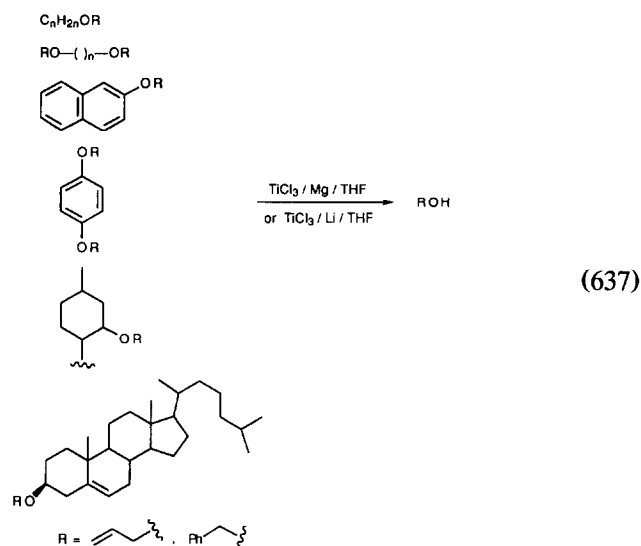


Enol ethers were converted to α -hydroxyketones by iodossyl benzene (eqn. (630) [748] and osmium (eqn. (631) [749]). Reduced titanocenes reduced allyl epoxides to allyl alcohols (eqn. (632) [750]), while nickel complexes catalyzed the air oxidation of epoxides to diols (eqn. (633) [751]). Alkylated naphthols were oxidized to α -hydroxyketones by t-butyl hydroperoxide in the presence of zirconium(II) [752].



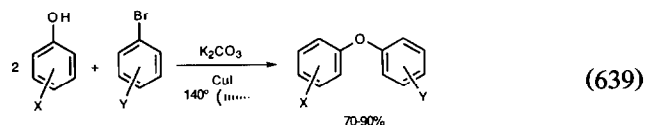
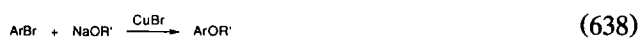
π -Allylmolybdenum compounds were hydroborated and converted to a variety of oxygenated compounds (eqn. (634) [753]). Ketones were asymmetrically reduced by silanes in the presence of rhodium(I) catalysts (eqn. (635) [754]). Esters were reduced to alcohols by silanes in the presence of titanium salts (eqn. (636) [745]). Allyl and benzyl ether protecting groups were removed by low valent titanium species (eqn. (637) [756]).



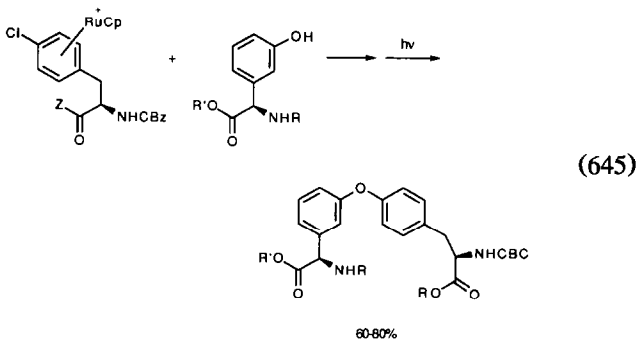
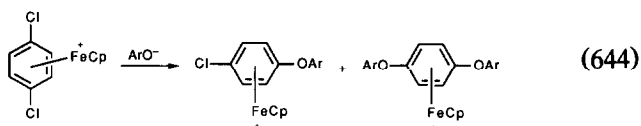
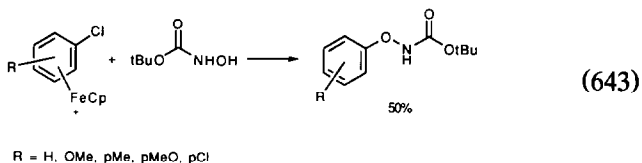
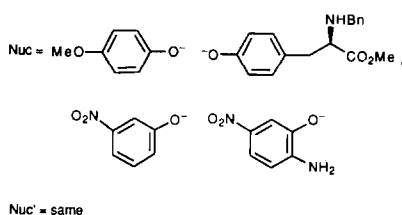
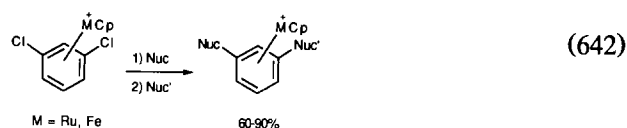
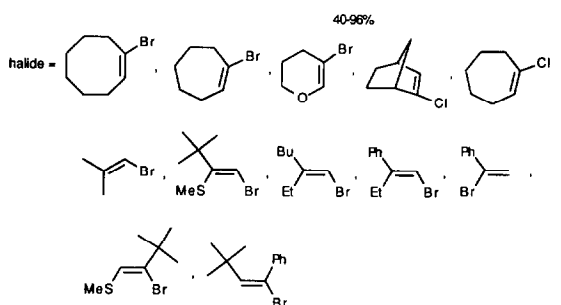
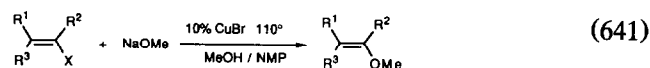
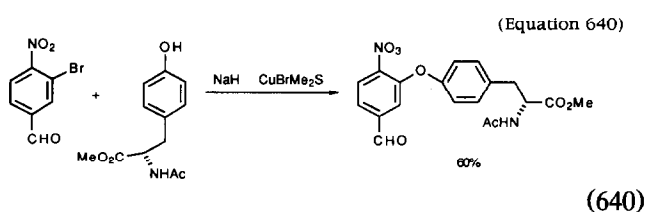


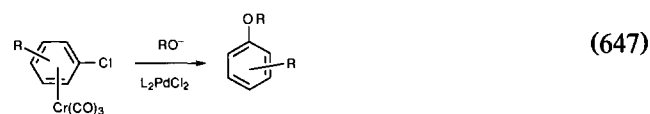
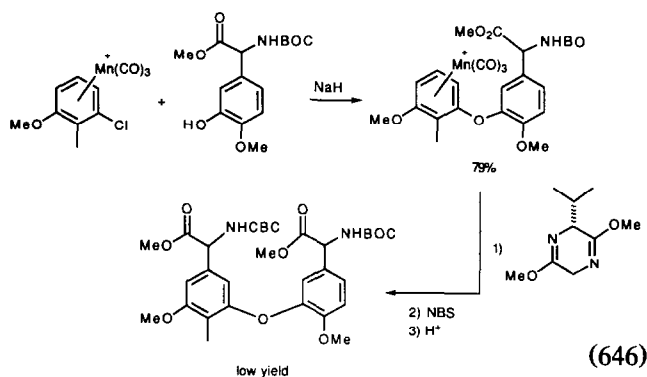
3.4. Ethers, esters, acids

Aryl ethers were prepared by the copper catalyzed reactions of aryl halides with alkoxides (eqn. (638) [757], eqn. (639) [758] and eqn. (640) [759]), as were enol ethers (eqn. (641) [760]). Aryl ethers were also made by reaction of alkoxides with iron (eqn. (642) [761], eqn. (643) [762], eqn. (644) [763]), ruthenium (eqn. (645) [764]), manganese (eqn. (646) [765]), and chromium (eqn. (647) [766]) complexes of chloroarenes.

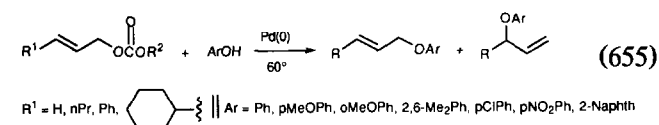
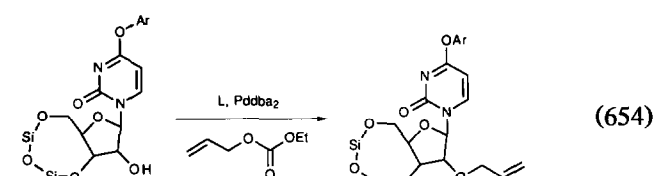
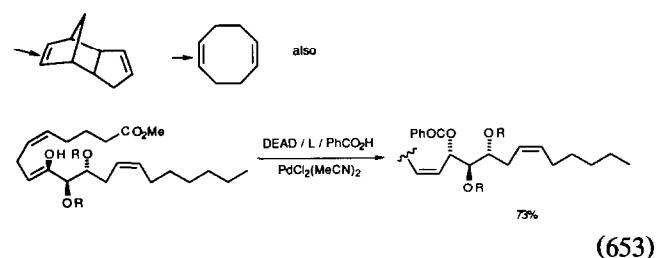
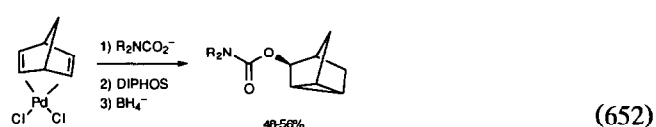
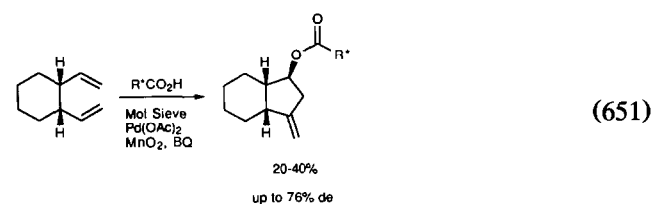
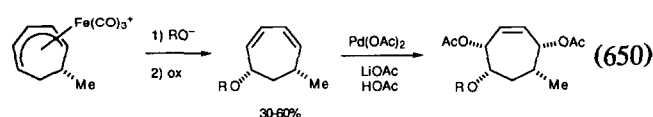
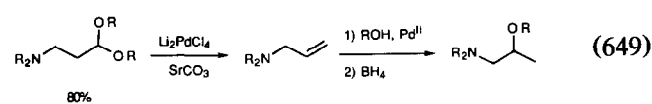
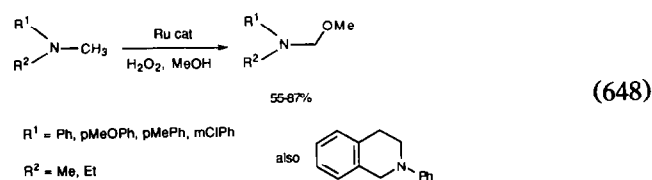


$X = \text{H}, 2\text{Me}, 3\text{Me}, 4\text{Me}, 2\text{MeO}, 3,5\text{-Me}_2$
 $Y = 2\text{MeO}, 4\text{MeO}, 2\text{Me}, 3\text{Me}, 4\text{Me}, \text{H}, 1\text{Naphth}$

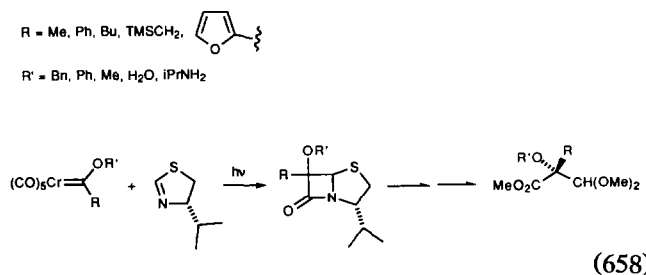
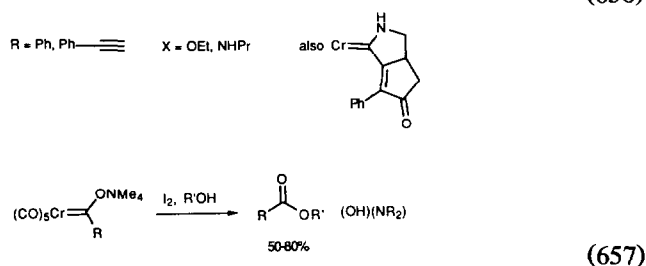
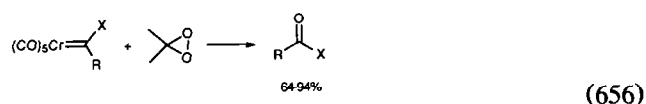


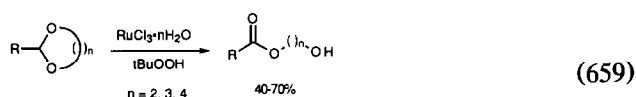


N-Methylamines were oxidized to methoxymethyl amines by ruthenium catalysts and hydrogen peroxide (eqn. (648) [767]). Palladium catalyzed the alkoxylation (eqn. (649) [768]) and the acetoxylation (eqn. (650) [769], eqn. (651) [770], eqn. (652) [771], eqn. (653) [772]) of alkenes, the allylation of protected ribonucleosides (eqn. (654) [773]), and the conversion of allyl carbonates to allyl aryl ethers (eqn. (655) [774]).

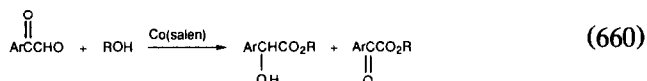


Oxidation of chromium carbene complexes gave esters (eqn. (656) [775] and eqn. (657) [776]). Chiral esters were made from chromium carbene derived β -lactams (eqn. (658) [777]). Ruthenium oxidized ketals to hydroxy esters (eqn. (659) [778]). Cobalt salen complexes converted α -ketoaldehydes to α -hydroxyesters (eqn. (660) [779]).





R = Ph, C₆, pCIPh, pMeOPh, pMePh, Ph, PhCH₂, EtO₂CCH₂

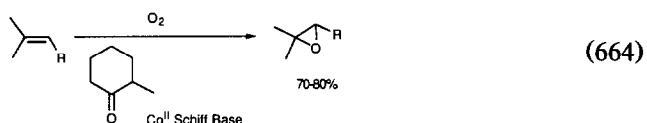
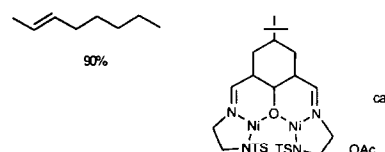
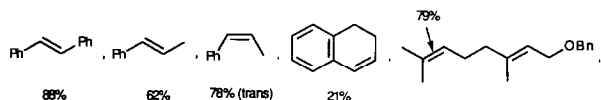
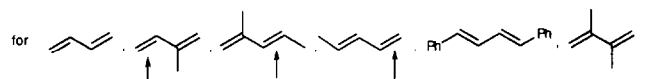
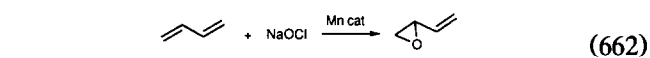
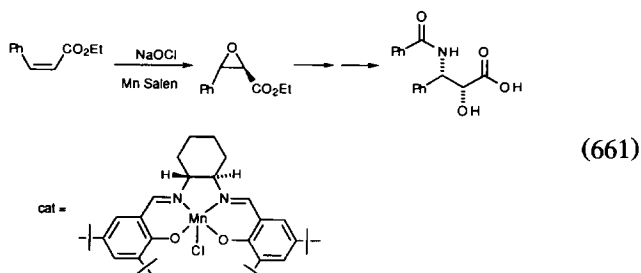


3.5. Heterocycles

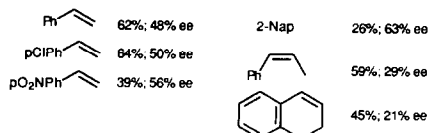
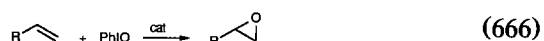
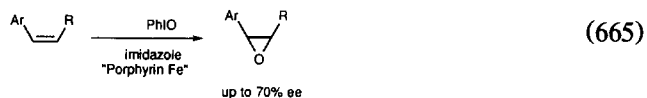
The following reviews on heterocyclic syntheses have appeared: "Synthesis of heterocyclic systems by activation of isocyanide, carbonyl, trifluoromethyl and nitrile ligands in platinum(II) complexes" (more than 62 references) [780]; "Asymmetric synthesis of nitrogen-containing biologically active compounds utilizing intramolecular aminocyclization to olefin of secondary allylic alcohols" (more than 35 references) [781]; "Synthesis of oxygen-containing heterocycles using palladium(II) catalysts" (more than 50 references) [782]; "Titanium metallacycles as intermediates in the synthesis of acyclic and heterocyclic compounds" (37 references) [783]; "Iron-mediated synthesis of heterocyclic ring systems and applications in alkaloid chemistry" (87 references) [784].

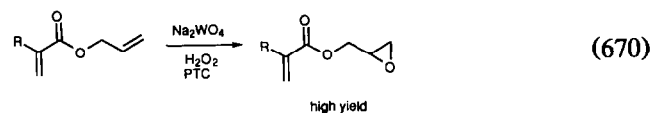
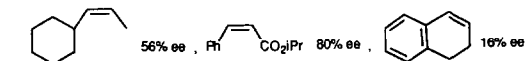
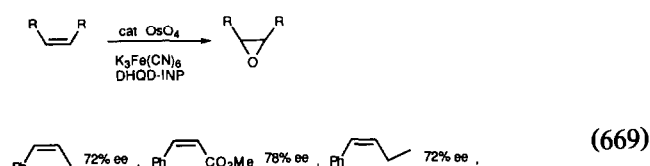
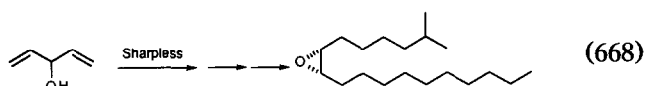
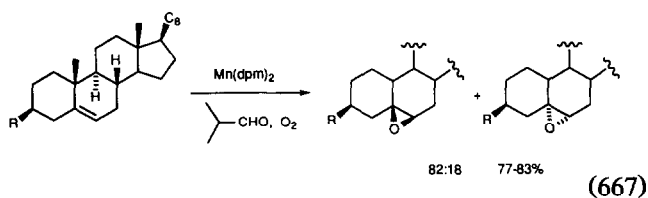
Epoxidation has been extensively studied. Hydroxy chalcones were epoxidized by t-butylhydroperoxide in the presence of V₂O₅ [785]. C-2 symmetrical binaphthyl bridged indenyl titaniumocene complexes catalyzed the asymmetric epoxidation of unfunctionalized alkenes, but with low ($\approx 20\%$) ee [786]. A conference report on shape selectivity in the epoxidation of olefins over supported titanium catalysts has appeared [787]. Titanium tetraphenyl porphyrin catalyst epoxidized alkenes [788], as did ruthenium(III)/EDTA complexes [789], and dimeric molybdenum(VI) catalyses [790].

Asymmetric epoxidation using salen metal complexes was the topic of a review (9 references) [791]. Examples are seen in eqn. (661) [792], eqn. (662) [793], and eqn. (663) [794]. Cobalt Schiff's base complexes also catalyzed the epoxidation of olefins (eqn. (664) [795]).

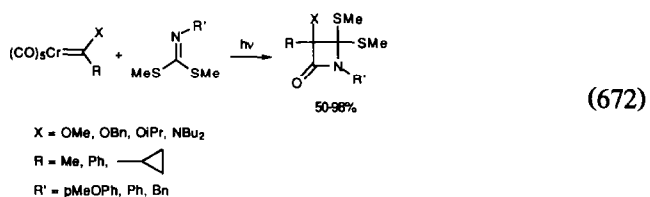
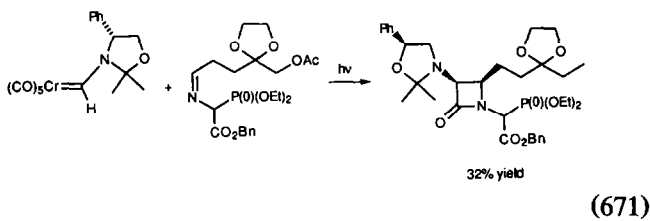


Iron and manganese "twin coronet" porphyrin complexes (eqn. (665) [796]) and iron binaph capped porphyrins (eqn. (666) [797]) catalyzed the asymmetric epoxidation of alkenes. Steroidal olefins were epoxidized by manganese compounds and air in the presence of isobutyraldehyde (eqn. (667) [798]). A full paper on the nature of the Katsuki-Sharpless Asymmetric Epoxidation catalyst has appeared [799]. Examples of its use are seen in eqn. (668) [800] and eqn. (669) [801]. Tungstate catalyzed the epoxidation of alkenes by peroxides (eqn. (670) [802]).

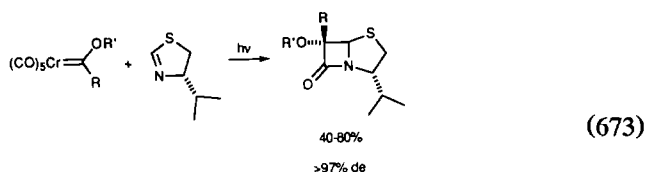




Chiral β -lactones were made from optically active iron acylenolates [803]. Chromium carbene mediated β -lactam syntheses was the topic of a dissertation [804]. Photolysis of chromium carbene complexes in the presence of imines gave β -lactams (eqn. (671) [805], eqn. (672) [806], and eqn. (673) [807]). Rhodium-catalyzed diazodecomposition also produced β -lactams (eqn. (674) [808]).

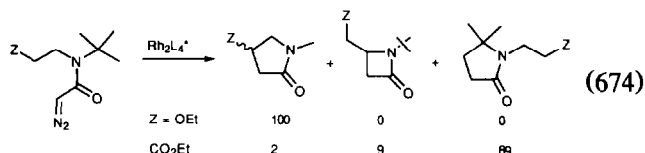


X = OMe, OBn, OiPr, NBU₂
 R = Me, Ph,
 R' = pMeOPh, Ph, Bn

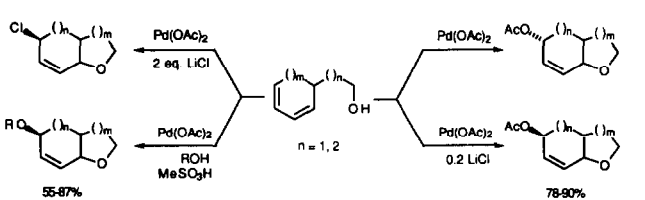
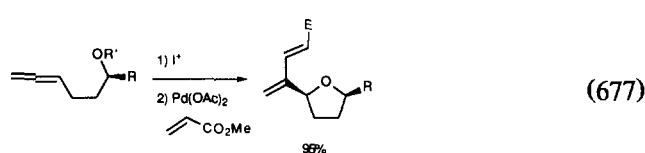
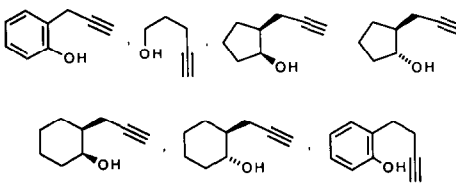
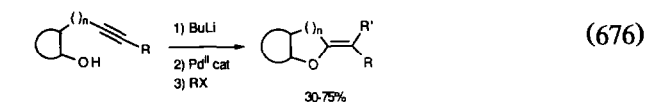
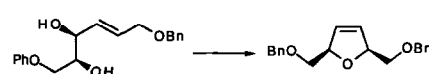
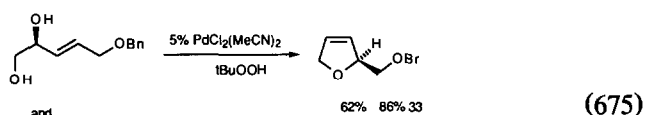


R = Me, Bu, , pMePh, pMeOPh, (CH₂)₃,

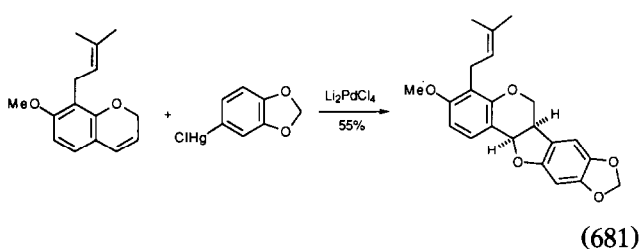
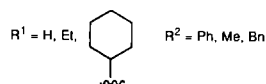
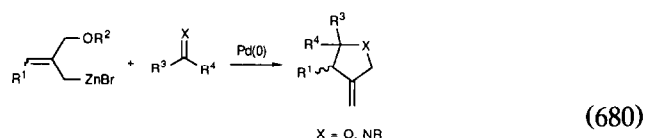
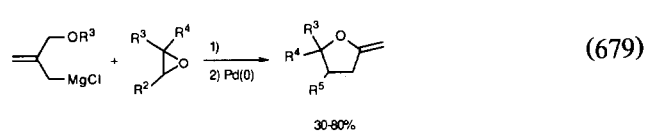
R' = Bn



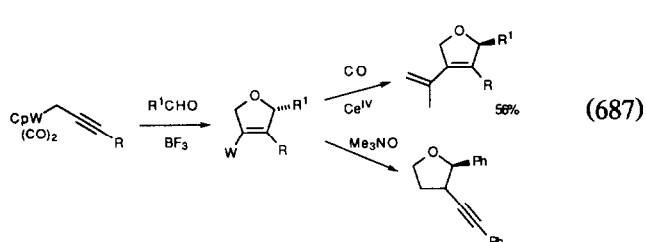
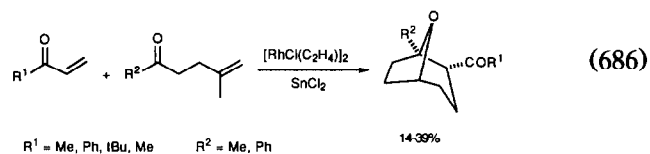
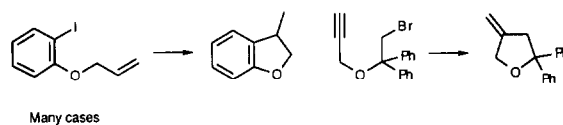
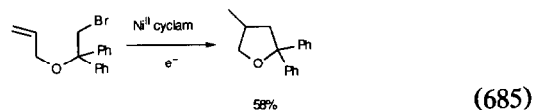
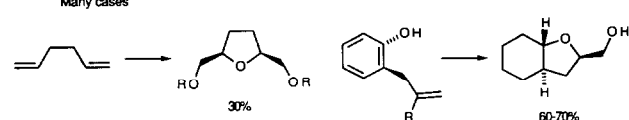
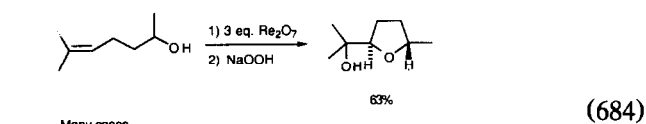
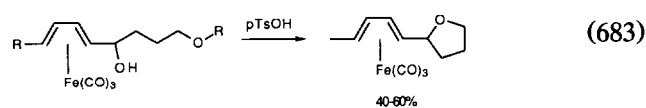
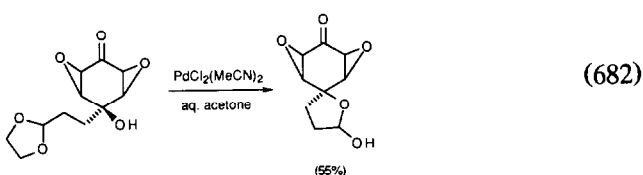
Five-membered oxygen heterocycles were synthesized by a variety of palladium catalyzed cyclization reactions (eqn. (675) [809], eqn. (676) [810], eqn. (677) [811], eqn. (678) [812], eqn. (679) [813], eqn. (680) [814], and eqn. (681) [815]).



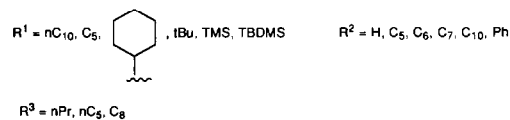
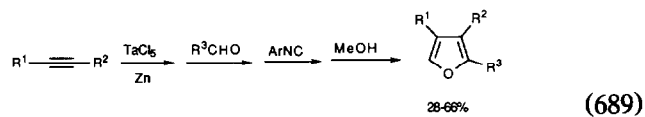
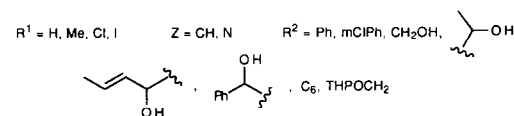
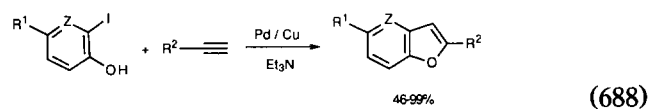
(678)

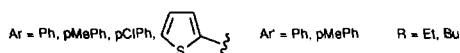
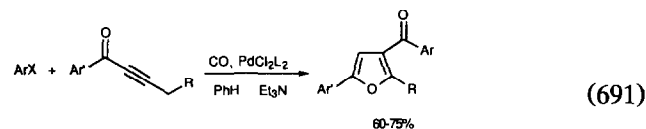
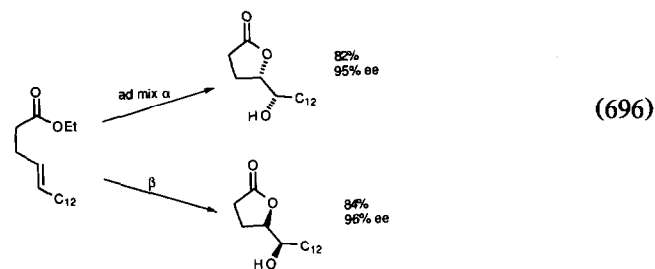
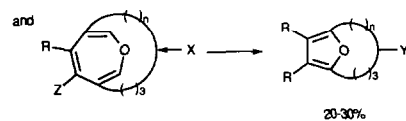
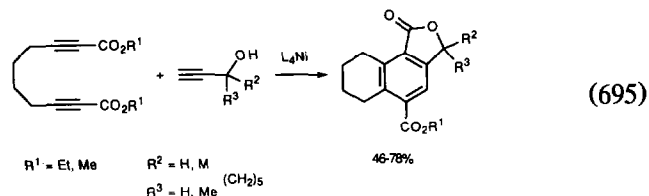
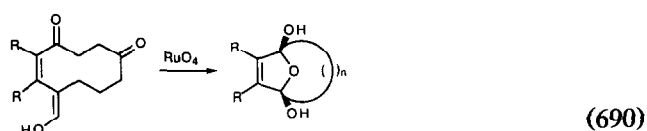


Five membered oxygen heterocycles were made by hydrolytic (eqn. (682) [816], eqn. (683) [817]), oxidative (eqn. (684) [818], eqn. (819) [820]), and reductive (eqn. (685) [821]) procedures. Rhodium(I) catalyzed the cyclization of olefinic ketones (eqn. (686) [822]). Propargyl tungsten complexes reacted with aldehydes to give five membered oxygen heterocycles (eqn. (687) [823]).

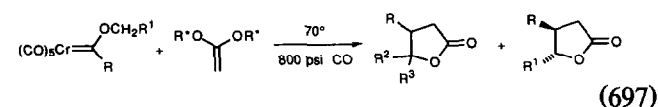


Palladium catalyzed the reaction between *o*-iodophenol and alkynes to give benzofurans (eqn. (688) [824,825]). Tantalum combined alkynes with isonitriles to produce furans (eqn. (689) [826]). Ruthenate oxidatively produced furans (eqn. (690) [827]). Palladium catalyzed the reaction between acetylenic ketones and aryl halides to produce furans (eqn. (691) [828]).

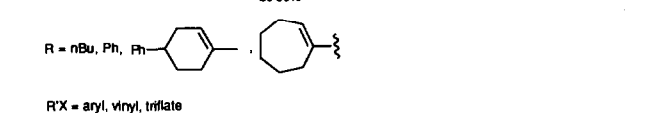
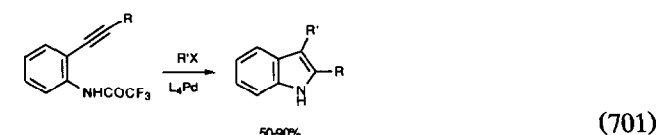
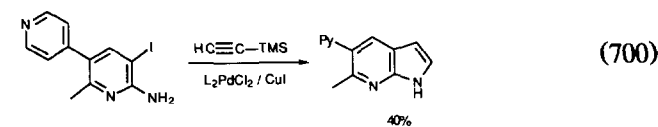
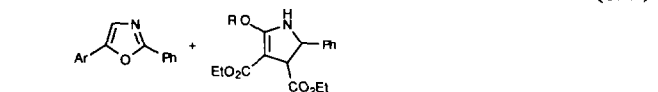
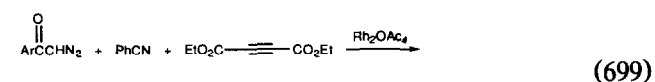
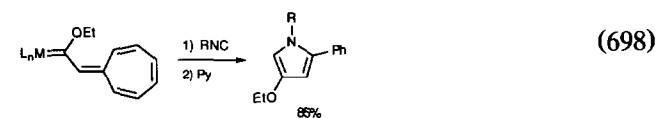
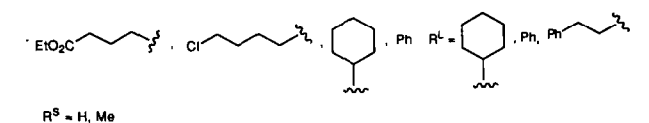
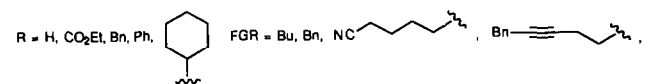
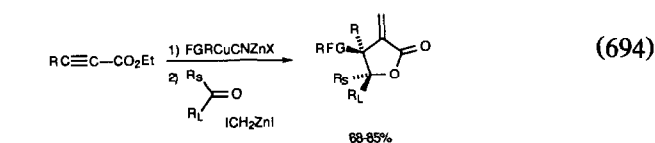
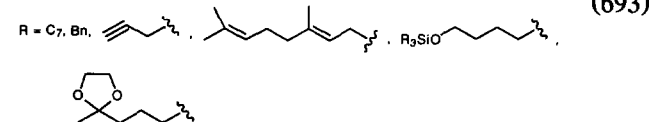
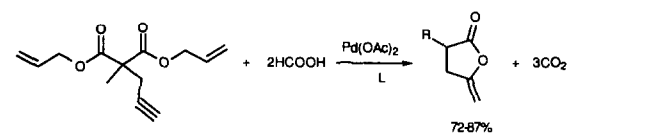
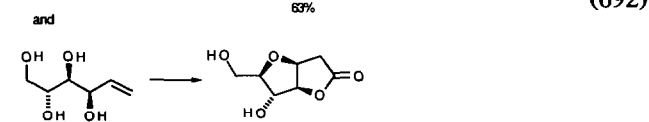
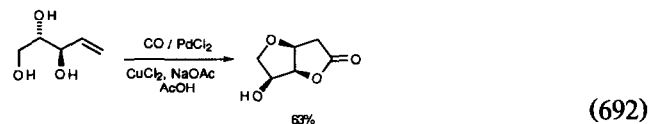


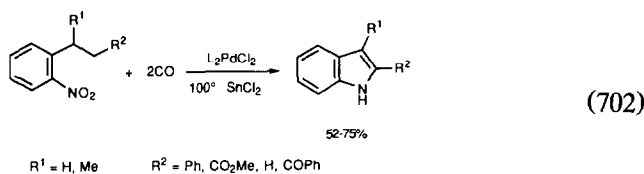


Furanones were synthesized by palladium catalyzed cyclizations (eqn. (692) [829,830], eqn. (693) [831]), by copper (eqn. (694) [832]), nickel (eqn. (695) [833]), and oxidative chemistry (eqn. (696) [834]). Chromium carbene complexes reacted with ketene acetals to give furanones (eqn. (697) [835]).

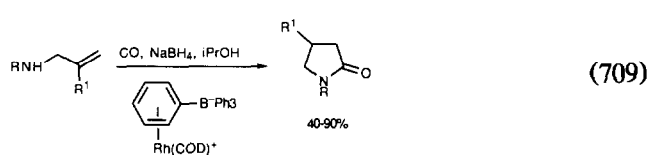
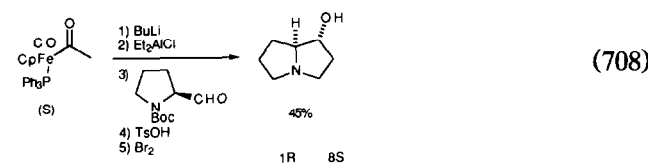
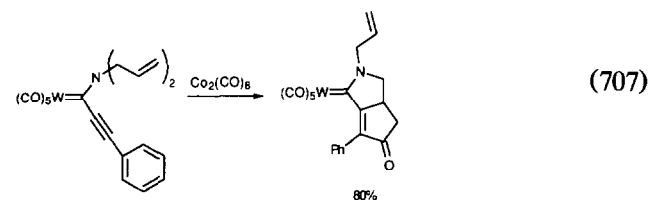
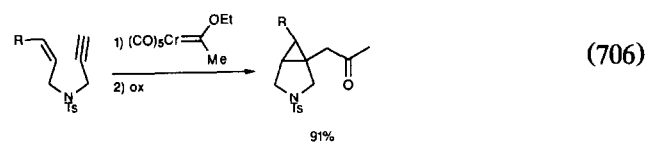
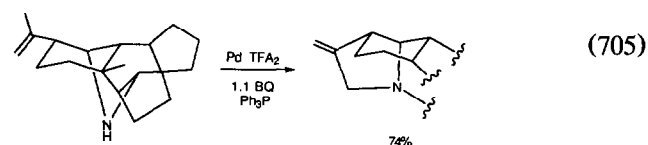
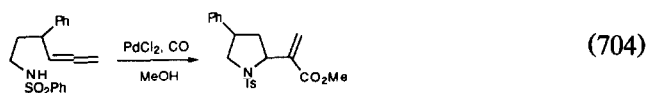
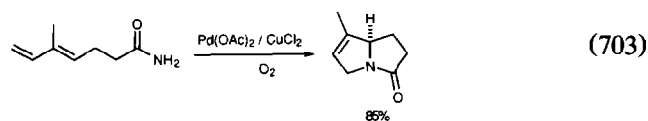


Pyrroles were made *via* chromium (eqn. (698) [836]) and rhodium (eqn. (699) [837]) carbene chemistry. Indoles were synthesized from alkynes and *o*-iodoanilines *via* palladium chemistry (eqn. (700) [838], eqn. (701) [839]), and by reductive carbonylation of nitrostyrenes (eqn. (702) [840]).

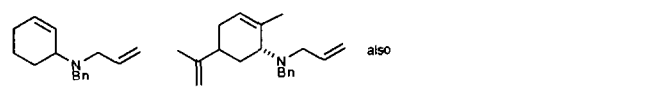
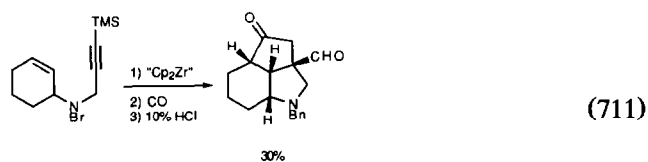
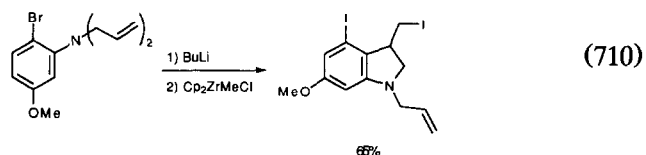




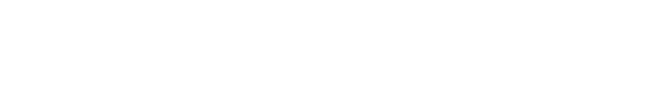
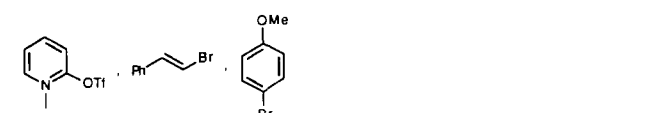
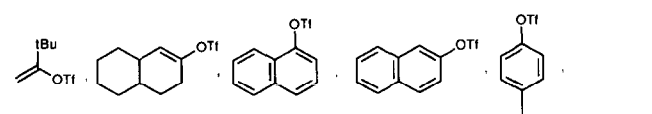
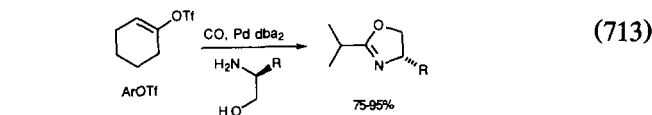
Five membered nitrogen heterocycles were made by palladium catalyzed olefin amination (eqn. (703) [841], eqn. (704) [842], eqn. (705) [843]), from chromium carbene chemistry (eqn. (706) [844], eqn. (707) [845]), and by iron acyl anion chemistry (eqn. (708) [846]). Rhodium catalyzed the cyclization of amino olefins (eqn. (709) [847]).

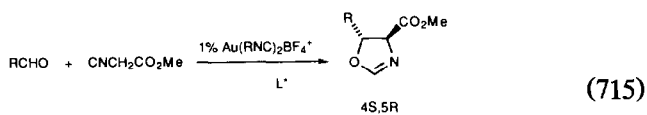
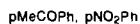
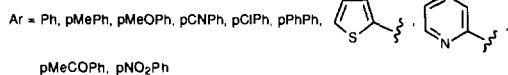
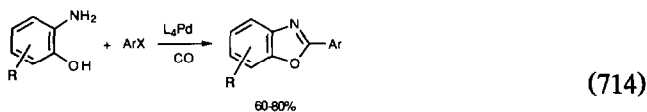


Five membered nitrogen heterocycles were also made by zirconium (eqn. (710) [848], eqn. (711) [849]) and titanium insertion chemistry (eqn. (712) [850]).



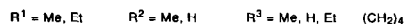
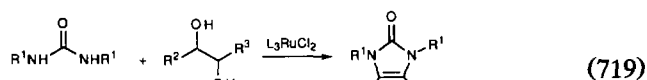
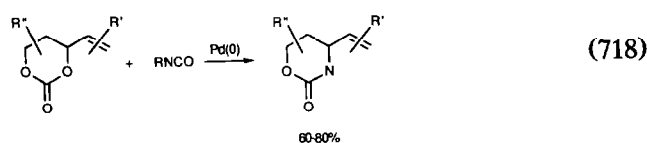
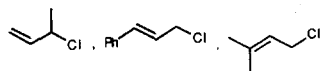
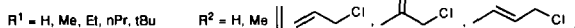
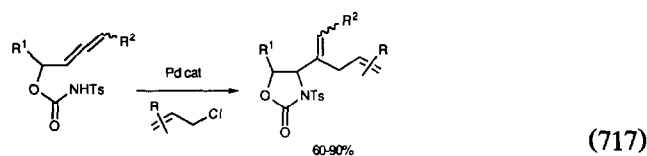
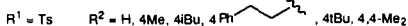
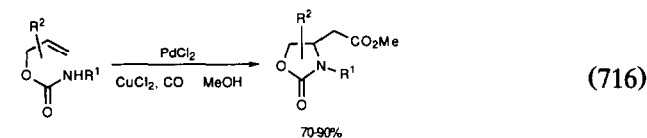
Oxazolines were synthesized by the palladium catalyzed aminocarbonylation of triflates (eqn. (713) [851]) and aryl halides (eqn. (714) [852]) and the gold-catalyzed condensation of aldehydes with α -isocyanoesters (eqn. (715) [853]). Oxazolidinones were prepared by palladium catalyzed olefin amidation (eqn. (716) [854], eqn. (717) [855]), palladium catalyzed isocyanate exchange into cyclic carbonates (eqn. (718) [856]), and the ruthenium catalyzed reaction of ureas with diols (eqn. (719) [852]).



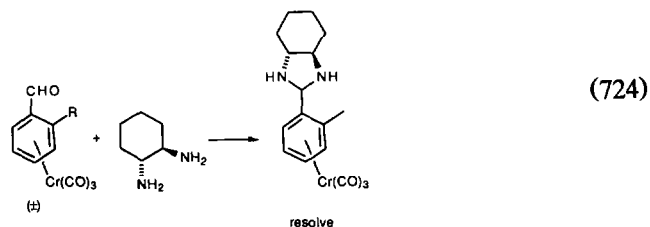
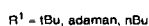
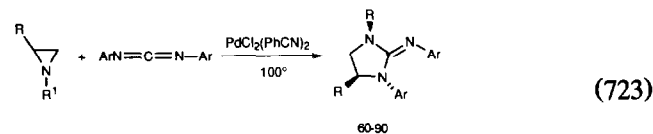
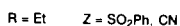
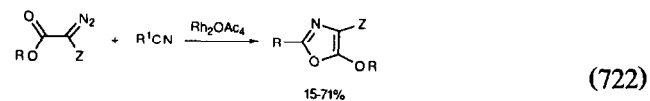
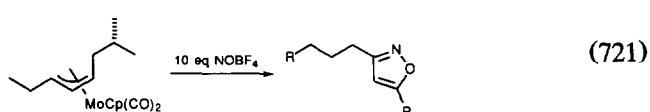
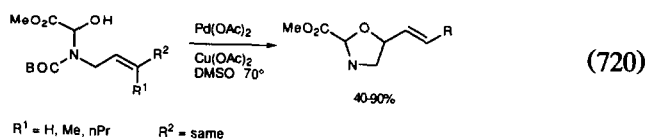


many cases

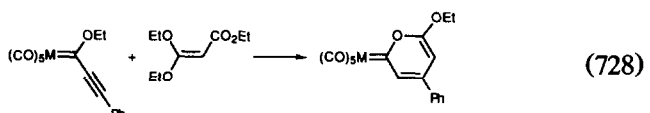
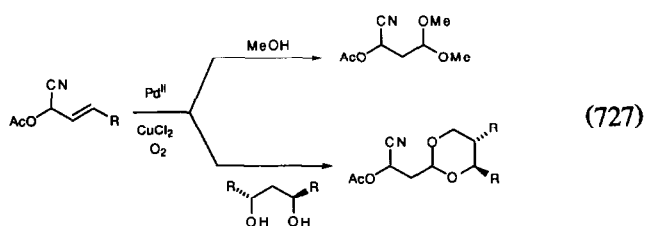
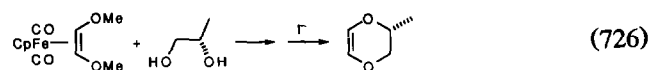
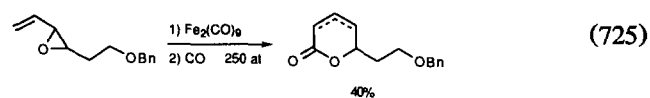
high yield up to 97% ee

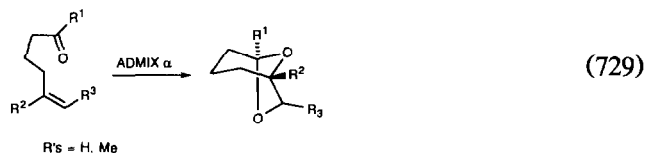


Other N, O-heterocycles were synthesized as in eqn. (720) [858], eqn. (721) [859], eqn. (722) [860], eqn. (723) [861], and eqn. (724) [862].

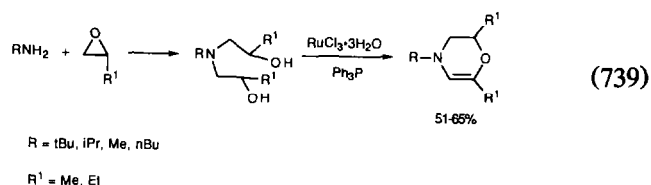
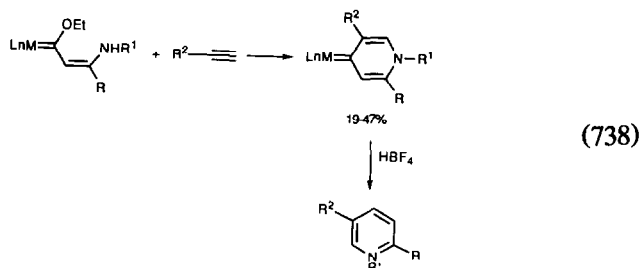
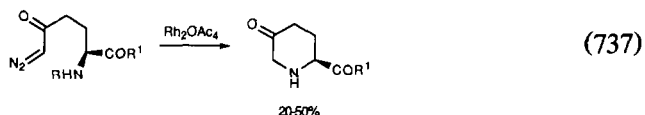
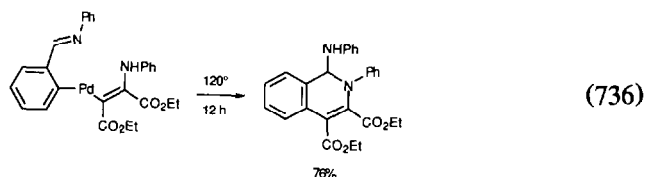
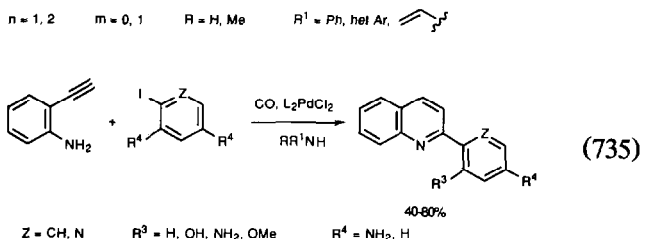
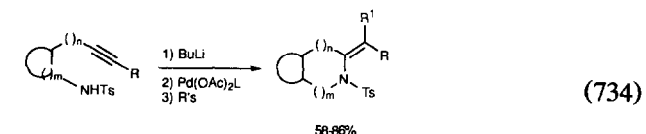
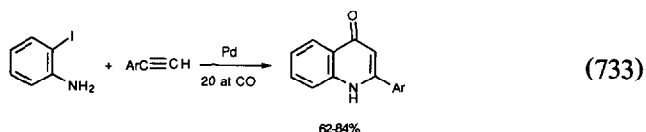
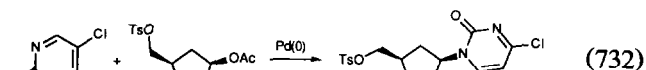
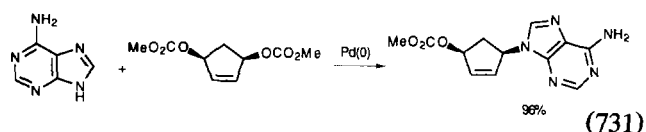
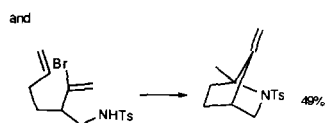
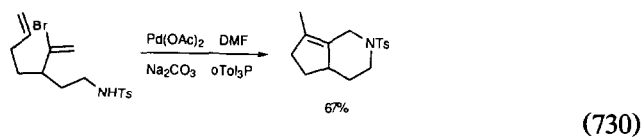


Six membered oxygen heterocycles were synthesized from allyl epoxides *via* iron chemistry (eqn. (725) [863]), iron alkene chemistry (eqn. (726) [864]), palladium catalyzed alkoxylation of olefins (eqn. (727) [865]), chromium carbene chemistry (eqn. (728) [866]) and osmium oxidation chemistry (eqn. (729) [867]).

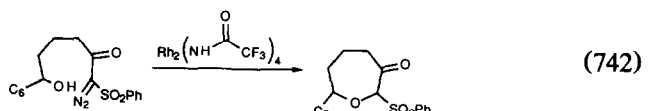
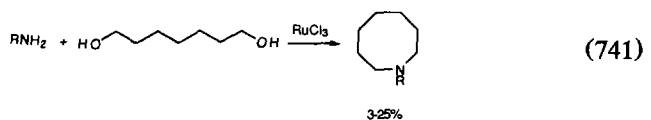
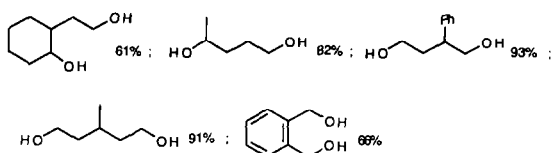
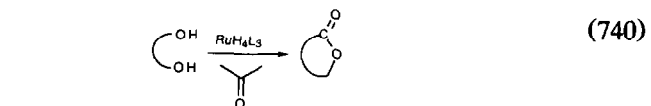


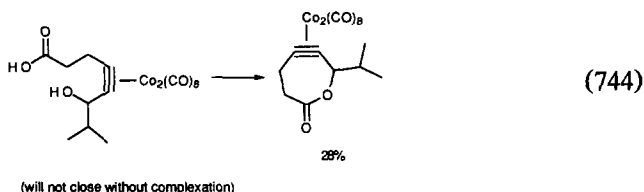
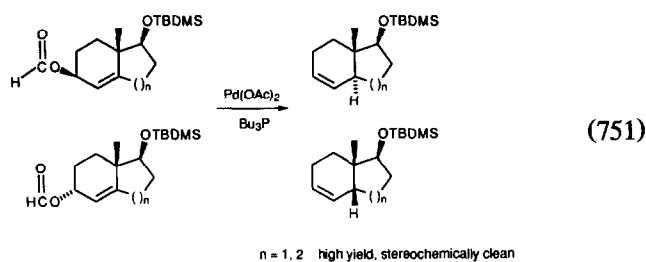
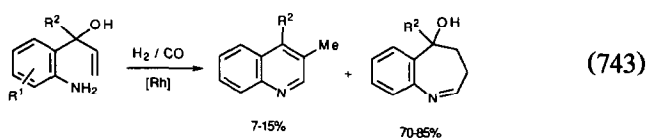


Six membered nitrogen heterocycles were prepared by palladium catalyzed olefin amination (eqn. (730) [868]), allylic amination (eqn. (731) [869], eqn. (732) [870]), palladium catalyzed alkyne amination (eqn. (733) [871], eqn. (734) [872], eqn. (735) [873], eqn. (736) [874]), rhodium catalyzed N-H insertion (eqn. (737) [875]), chromium carbene chemistry (eqn. (738) [876]), and ruthenium catalyzed oxidation chemistry (eqn. (739) [877]).

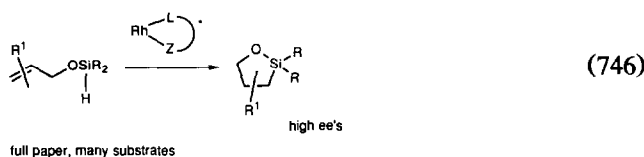
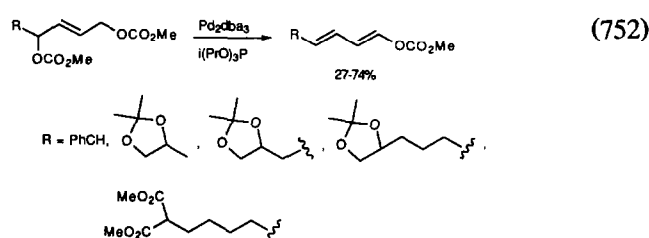
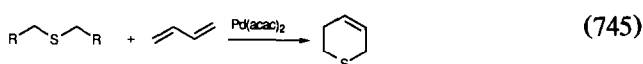


Other miscellaneous approaches to heterocycles are seen in eqn. (740) [878], eqn. (741) [879], eqn. (742) [880], eqn. (743) [881], eqn. (744) [882], eqn. (745) [883], and eqn. (746) [884].



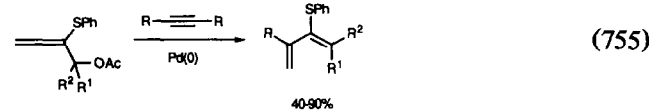
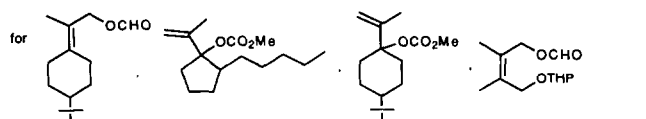
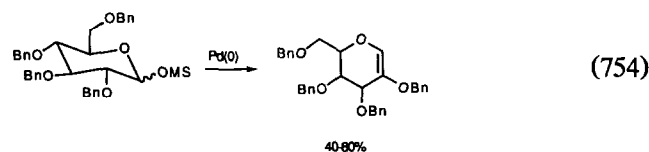
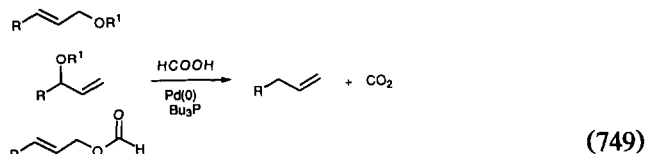
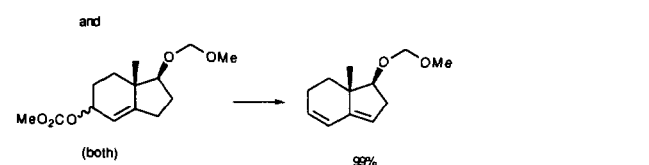
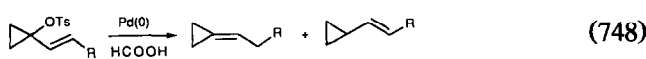
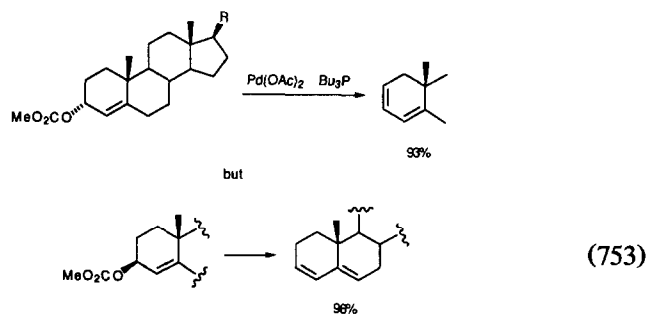
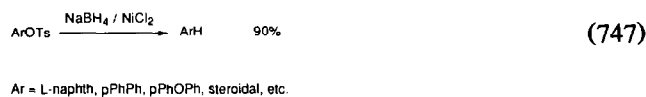


Palladium catalyzed the elimination of allyl esters to give dienes (eqn. (752) [891], eqn. (753) [892], eqn. (754) [893] and eqn. (755) [894]). Molybdenum isonitriles catalyzed a similar process (eqn. (756) [895]).

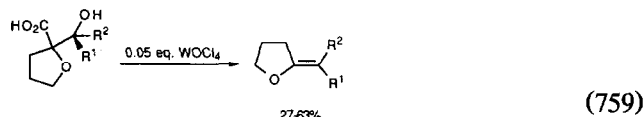
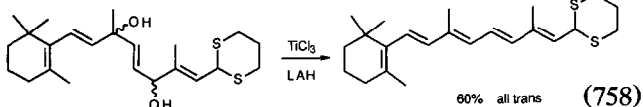
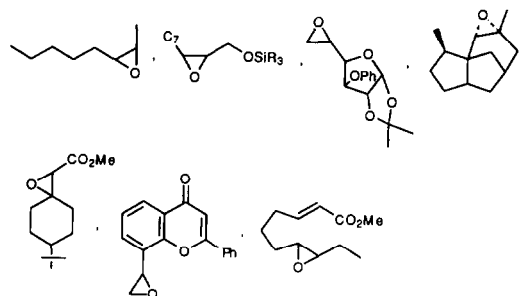
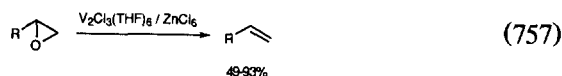


3.6. Alkanes, alkenes

Palladium (or nickel) catalyzed the reduction of aryl sulfonates (review, 11 references) [885], (eqn. (747) [886]), allyl sulfonates (eqn. (748) [887]), allyl ethers (eqn. (749) [888]), and allyl esters (eqn. (750) [889] and eqn. (751) [890]).



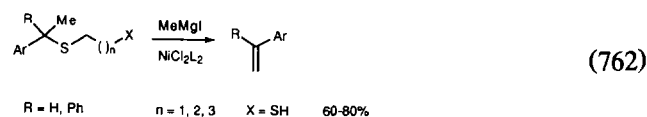
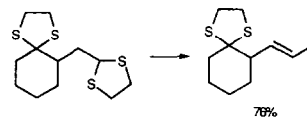
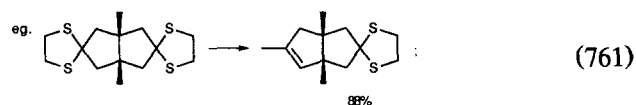
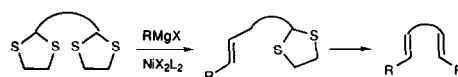
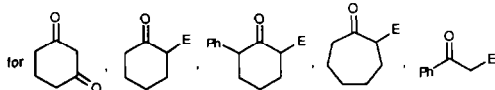
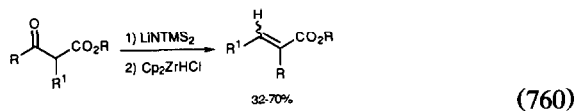
Reduced vanadium species deoxygenated epoxides (eqn. (757) [896]) while reduced titanium species deoxygenated diols (eqn. (758) [897]). Tungsten (eqn. (759) [898]) and zirconium species (eqn. (760) [899]) deoxygenated organic compounds.



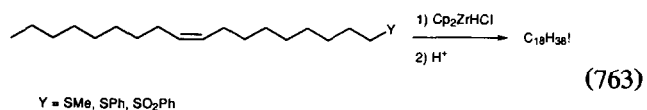
$R^1 = \text{H, Ph, pMeOPh, pNO}_2\text{Ph, Et}$

$R^2 = \text{H, Me, Et}$

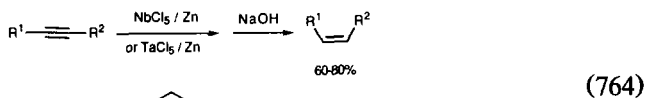
$(\text{CH}_2)_4$



$R = \text{H, Ph}$ $n = 1, 2, 3$ $X = \text{SH}$ 60-80%



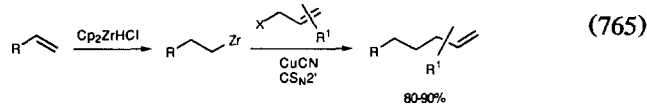
$Y = \text{SMe, SPh, SO}_2\text{Ph}$



$R^1 = \text{C}_{10}, \text{Ph, C}_5, \text{C}_6, \text{IBu, TMS}$

$R^2 = \text{H, C}_5, \text{C}_6, \text{C}_7, \text{C}_{10}$

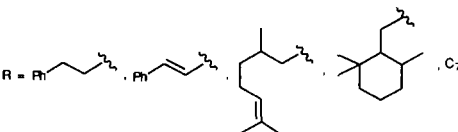
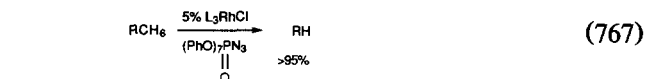
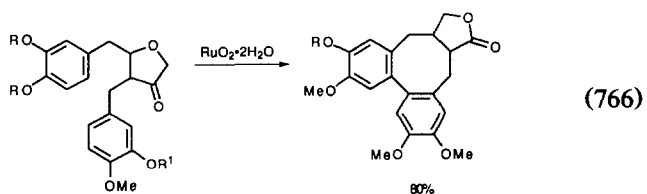
$(\text{CH}_2)_5$ $(\text{CH}_2)_{10}$



$R = \text{C}_6, \text{Br}, \text{Ph}, \text{IBu, } \sigma\text{TIPS}$

allyl = $\text{CH}_2=\text{CHCH}_2$, $\text{MeO}_2\text{CCH}=\text{CHCH}_2$, $\text{CH}_2=\text{CHCH}_2$, $\text{CH}_2=\text{CHCH}_2$

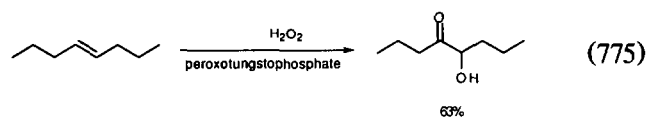
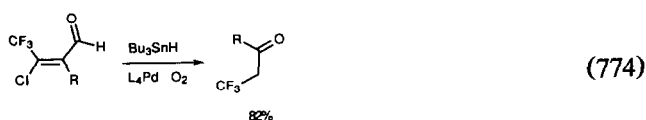
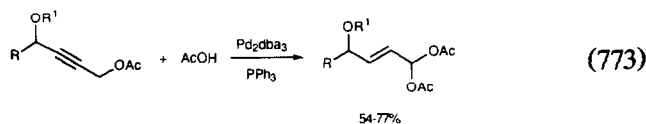
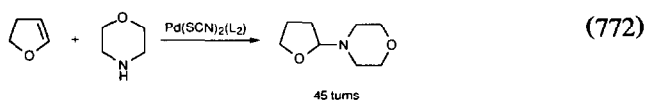
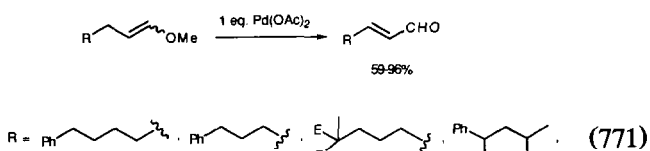
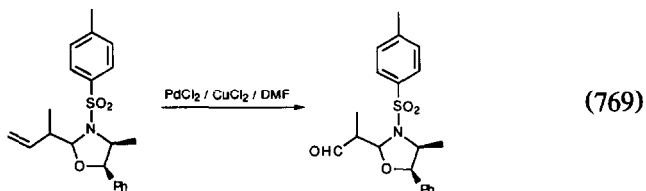
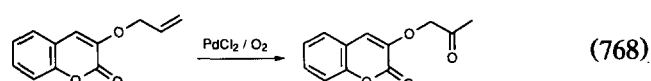
$\text{CH}_2=\text{CHCH}_2$, $\text{PhCH}=\text{CHCH}_2$



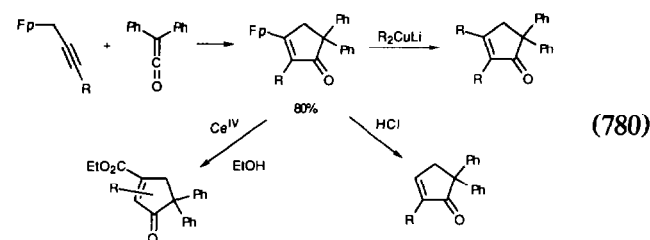
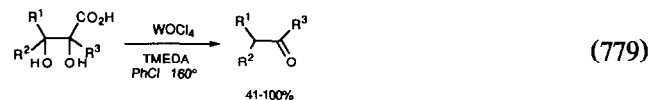
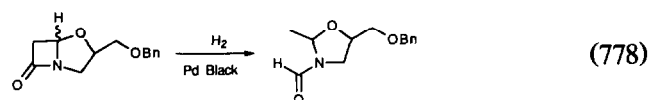
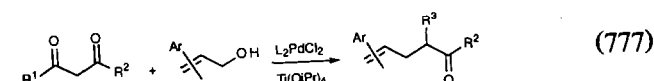
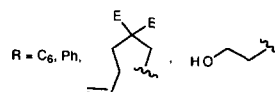
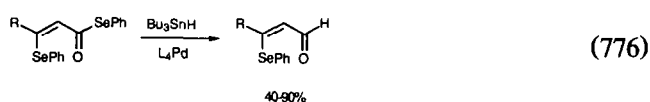
Grignard reagents converted thiols to alkenes in the presence of nickel catalysts (eqn. (761) [900] and eqn. (762) [901]). Hydrozirconation of long chain olefin sulfur compounds resulted in loss of the functional group (eqn. (763) [902]). Alkynes were reduced to alkenes by reduced tantalum or niobium species (eqn. (764) [903]). Functionalized alkenes were prepared by coupling alkyl zirconium to allylic halides (eqn. (765) [904]). Ruthenium complexes catalyzed phenolic coupling (eqn. (766) [905]). A catalyst system for decarbonylation of aldehydes was devised (eqn. (767) [906]).

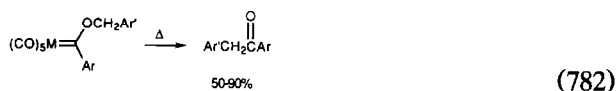
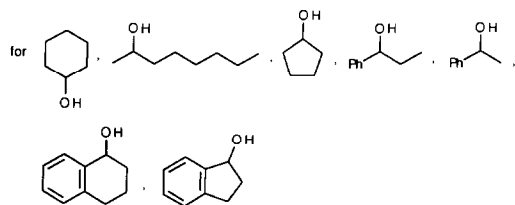
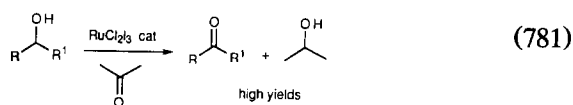
3.7. Ketones, aldehydes

Electrode mediated Wacker oxidation of cyclic olefins has been developed [907]. Palladium catalyzed the oxidation of alkenes to carbonyl compounds efficiently (eqn. (768) [908], eqn. (769) [909], eqn. (770) [910], and eqn. (771) [911]). It catalyzed the amination of dihydropyran (eqn. (772) [912]), the acetoxylation of propargyl acetates (eqn. (773) [913]), and the funny reaction in eqn. (774) [914]). Peroxy ditungstate oxidized alkenes to hydroxyketones (eqn. (775) [915]). Ruthenium catalyzed the cleavage of electron rich olefins to aldehydes [916].



Palladium catalyzed the conversion of selenoesters to aldehydes (eqn. (776) [917]), the coupling of allyl alcohols to diketones (eqn. (777) [918]), and the cleavage of β -lactams to formamides (eqn. (778) [919]). Tungsten oxychloride converted α -hydroxyacids to ketones (eqn. (779) [920]). Iron acetylene complexes reacted with diphenyl ketene to give cyclopentenones (eqn. (780) [921]). Ruthenium complexes catalyzed the oxidation of alcohols to ketones (eqn. (781) [922]). Carbene complexes were converted to ketones thermally (eqn. (782) [923]).



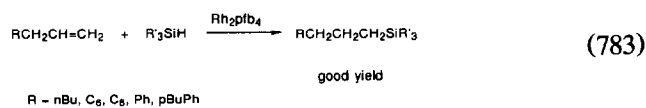
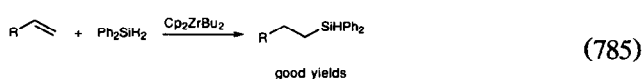
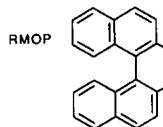
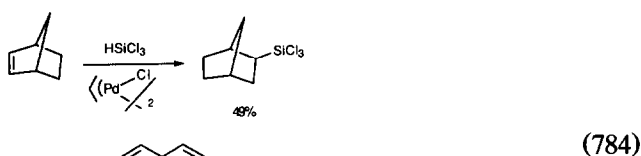
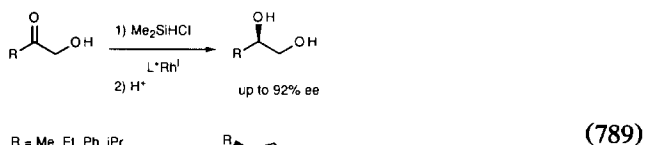
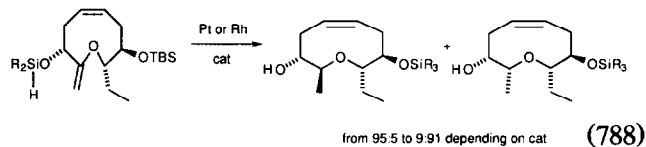
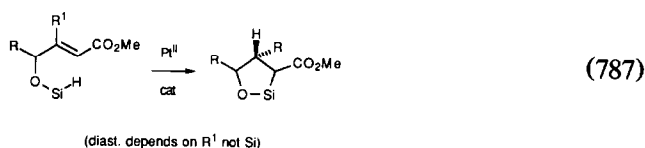
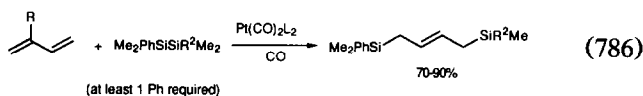


Ar = Ph, pMePh, pMeOPh

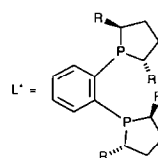
Ar = pMeOPh,

3.8. Organosilanes

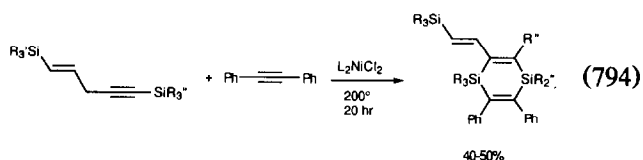
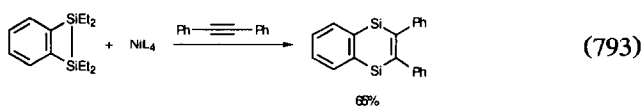
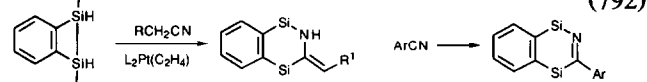
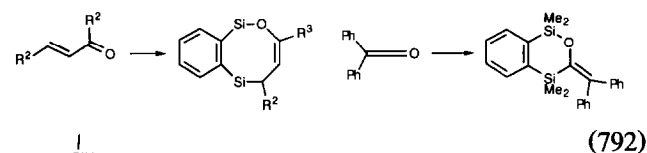
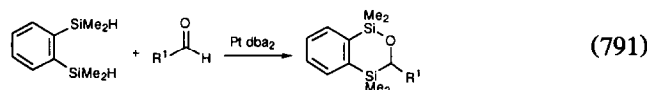
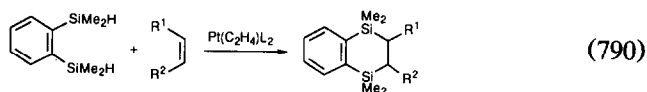
Rhodium(II) catalyzed hydrosilylation was discussed in a current review (375 references) [924]. Rhodium (eqn. (783) [925]), palladium (eqn. (784) [926]) and zirconium (eqn. (785) [927]) catalyzed hydrosilylation of alkenes. Platinum catalyzed the disilylation of dienes (eqn. (786) [928]). Intramolecular hydrosilylation was developed in several systems (eqn. (787) [929], eqn. (788) [930], and eqn. (789) [931]).

R = nBu, C₆, C₈, Ph, pBuPhR = Ph, nBu, nC₆, 

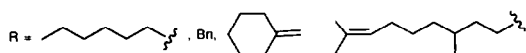
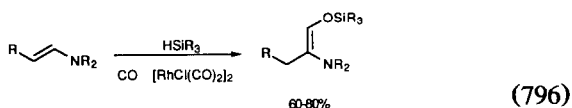
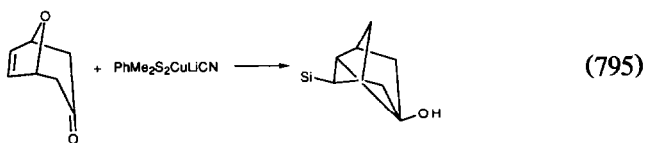
R = Me, Et, Ph, iPr



Cyclic disilanes were synthesized by metal catalyzed addition of disilanes to alkenes (eqn. (790) [932], eqn. (791) [933], eqn. (792) [934], eqn. (793) [935], and eqn. (794) [936]).

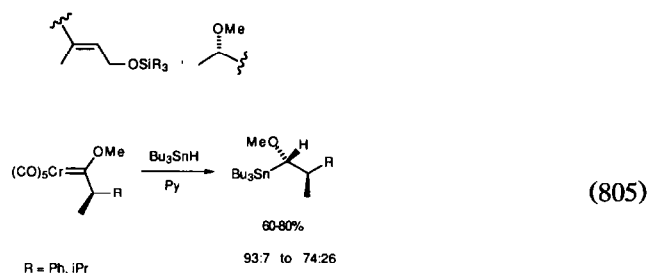
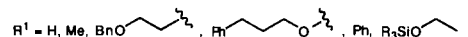
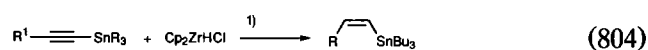
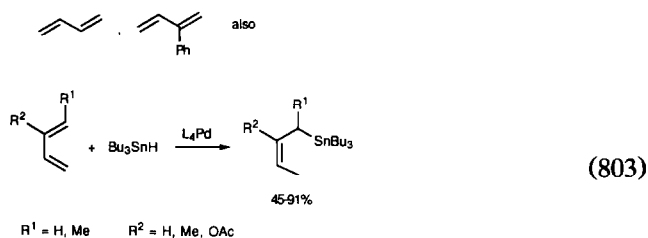
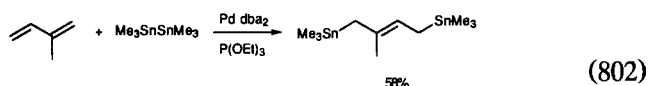
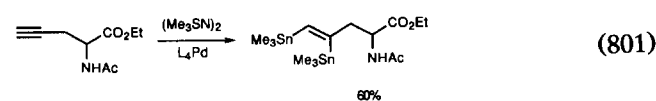
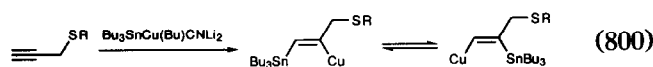
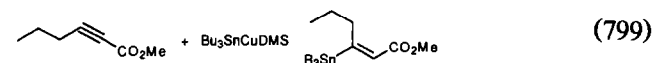
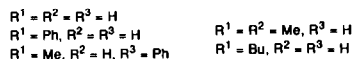
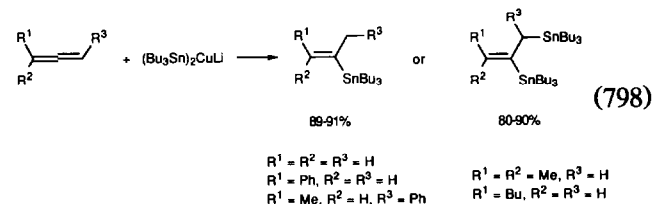
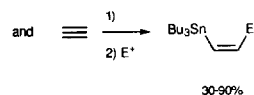
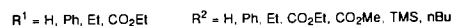
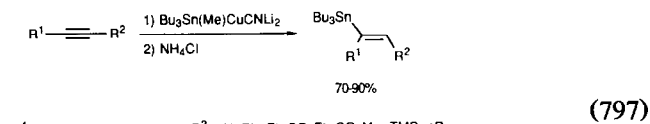


Other silylations are seen in eqn. (795) [937] and eqn. (796) [938].

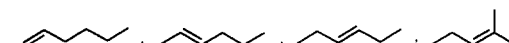
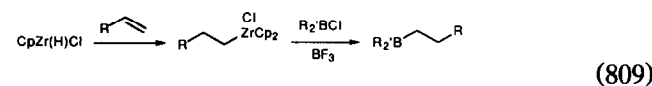
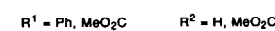
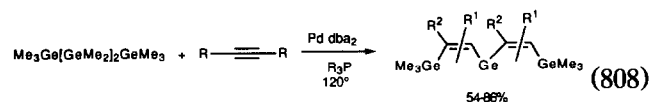
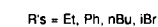
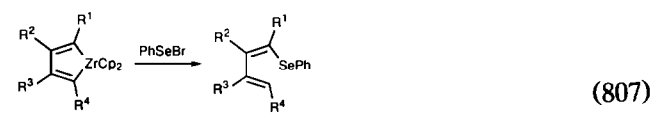
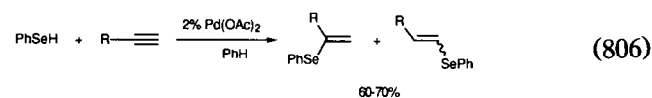


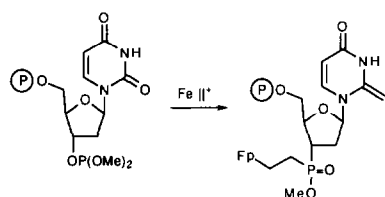
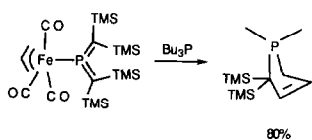
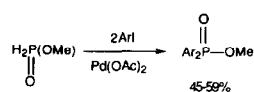
3.9. Miscellaneous

Herein are found all reactions which fit no other classification. Organotin compounds were made *via* organocopper chemistry (eqn. (797) [939], eqn. (798) [940], eqn. (799) [941], eqn. (800) [942]), organopalladium chemistry (eqn. (801) [943], eqn. (802) [944], eqn. (803) [945]), organozirconium chemistry (eqn. (804) [946]) and organochromium chemistry (eqn. (805) [947]).

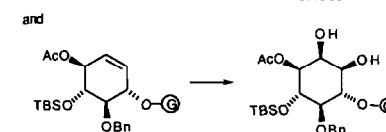
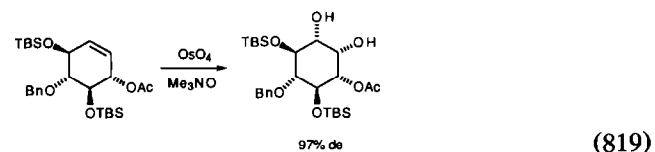
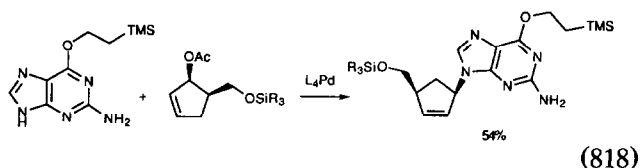
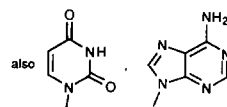
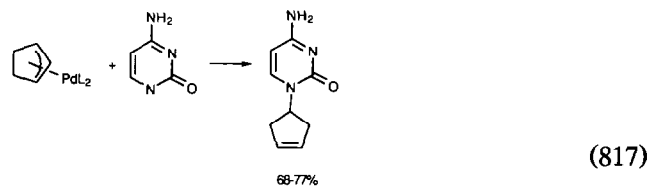
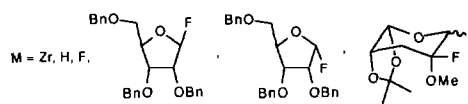
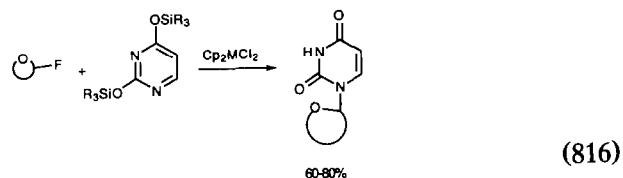
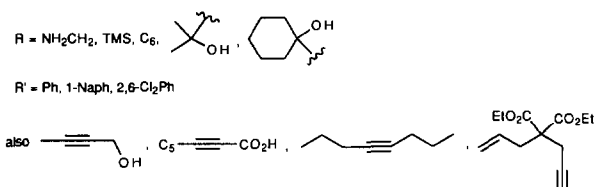
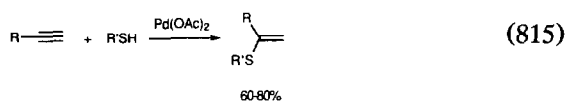
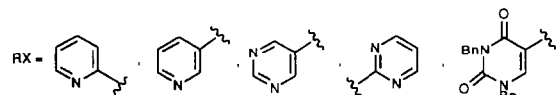
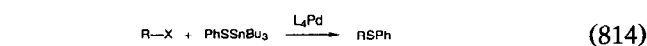
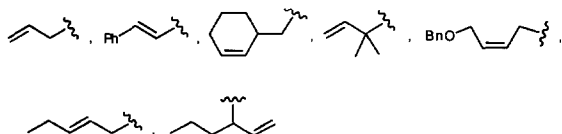
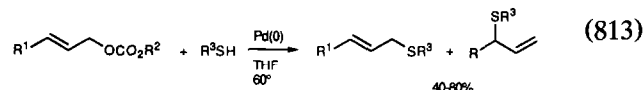


Palladium catalyzed the selenation of alkynes (eqn. (806) [948]). Zirconacycles underwent reaction with PhClBr (eqn. (807) [949]). Transition metals were used to introduce germanium (eqn. (808) [950]), boron (eqn. (809) [951]), phosphorous (eqn. (810) [952]), eqn. (811) [953]), and iron (eqn. (812) [954]) into organic substrates.





Palladium catalyzed the thiation of allyl carbonates (eqn. (813) [955]), aryl halides (eqn. (814) [956]), and alkynes (eqn. (815) [957]). Nucleosides were synthesized *via* zirconium (eqn. (816) [958]) and palladium chemistry (eqn. (817) [959] and eqn. (818) [960]). Hexaalkoxycyclohexanes were made by osmate oxidation (eqn. (819) [961]).



4. Reviews

The following reviews have appeared.

- Dicobalt hexacarbonyl complexes as chirality relay for the differentiation of acetylenic diastereomers [962];
- Selected applications to organic synthesis of intramolecular C-H activation reactions by transition metals (12 references) [963];
- Stereoselective synthesis with zirconium complexes (14 references) [964];
- Biomimetic oxidation in organic synthesis using transition metal catalysts (31 references) [965];
- Enzyme models and organometallic chemistry (15 references) [966];

- The chemistry of polyfunctional organozinc and copper reagents (15 references) [967];
- Applications of nickel(II)/chromium(II)-mediated coupling reactions to natural products syntheses (24 references) [968];
- Synthesis and reactivity of iron tricarbonyl complexes of vinylketenes, vinylketenimines and vinylallenes (12 references) [969];
- Asymmetric hydrogen transfer reactions promoted by homogeneous transition metal catalysts (more than 79 references) [970];
- Enantioselective rhodium(II) catalysts (18 references) [971];
- Organotitanium reagents in organic synthesis (21 papers) [972];
- Synthesis of enantiomerically pure 2° α halo allylic alcohols and their use in the synthesis of leukotrienes (41 references) [973];
- New aspects of organocopper reagents: 1,3- and 1,2-chiral induction and reaction mechanism (59 references) [974];
- Remote substituent effects on regioselectivity of Rh(I) cat. hydroboration of norbornene [975];
- Ruthenium oxo complexes as organic oxidants [976];
- Transition metals in organic synthesis – hydroformylation, reduction, and oxidation (897 references) [977];
- Catalytic asymmetric synthesis by means of secondary interaction between chiral ligands and substrates (129 references) [978];
- Synthesis and application of chiral Cp metal complexes (122 references) [979];
- Asymmetric hydrogen transfer reactions promoted by homogeneous transition metal catalysts (79 references) [980];
- Transition metals in organic synthesis annual survey 1990 (1048 references) [981];
- Fluorinated organometallics, perfluoroalkyl, and functionalized organometallic reagents in synthesis (297 references) [982];
- Transition metal catalyzed reactions involving reactions with ammonia and amines (278 references) [983];
- Recent developments in the synthesis of optically active α -arylpropanoic acids: an important class of non-steroidal anti-inflammatory agents (48 references) [984];
- Synthesis of organophosphorous compounds via coordination spheres of transition metals (63 references) [985];
- Synthesis and applications of binaphthyl C_2 -symmetry derivatives as chiral auxiliaries in enantioselective reactions (86 references) [986];
- Catalytic synthesis of optically active alcohols via hydrosilylation of olefins with a chiral monophosphine-palladium catalyst (19 references) [987];
- Development of organopalladium(IV) chemistry: fundamental aspects and systems for studies of mechanism in organometallic chemistry and catalysis (many references) [988];
- Transition metal catalyzed reactions of organozinc reagents (144 references) [989];
- Recent advances in the asymmetric dihydroxylation of alkenes (101 references) [990];
- Organic derivatives of manganese(II) in organic synthesis (99 references) [991];
- Selectivity control in cobalt(0) catalyzed cycloaddition reactions (29 references) [992];
- Organotitanium reagents in reduction reactions (41 references) [993];
- Organometallic chemistry. Part 1. The transition elements (226 references) [994];
- Organometallic chemistry. Part 2. Main-group elements (211 references) [995];
- Synthesis of optically active natural products based on palladium-catalyzed stereoselective reduction of alkenyloxiranes (9 references) [996];
- Organic synthesis via organometallics. Proceedings of the 3rd symposium in Marburg, July 11–14, 1990 [997];
- Aminocarbene complexes of chromium and molybdenum: initiators for cascade reactions with alkynes leading to new heterocyclic compounds via nitrogen ylides (60 references) [998];
- Palladium and BINAP, a strong pair (3 references) [999];
- Regio-, stereo-, and enantioselectivity in palladium- and platinum-catalyzed organic reactions (more than 77 references) [1000];
- Metals as selective coupling agents (4 references) [1001];
- Titanium metallacycles as intermediates in the synthesis of acyclic and heterocyclic compounds (37 references) [1002];
- Synthesis of optically active alcohols using ferrocene catalysts (29 references) [1003];
- Ligand-controlled catalysis: chemo- to stereoselective syntheses from olefins and dienes over nickel catalysts (more than 40 references) [1004];
- Basic principles in carbene chemistry and applications to organic synthesis (more than 42 references) [1005];
- Chemical synthesis of optically pure compounds. 3. Asymmetric catalysis (42 references) [1006];
- Carbonylchromium(0) complexes in organic synthesis (37 references) [1007];
- Homogeneous catalysis by transition metal complexes (370 references) [1008];
- Organo-iron complexes in aromatic synthesis (more than 14 references) [1009];
- Regio- and stereocontrol in allylation (11 references) [1010];

Chemistry of organochromium(III) complexes (more than 85 references) [1011];
 Organometallics in synthesis. The transition elements (89 references) [1012];
 Transition metals which changed organic synthesis: fundamentals and applications [1013];
 The allyl ether as a protecting group in carbohydrate chemistry (23 references) [1014];
 Studies in organo-transition metal chemistry [1015];
 Synthesis and reactivity patterns of (arene)manganese carbonyl and (cyclohexadienyl)manganese carbonyl derivatives [1016];
 Application of organic copper compounds (87 references) [1017];
 The chemistry of iron imido clusters and synergistic bimetallic iron/ruthenium catalysts for the carbonylation of nitroaromatics [1018];
 Homogeneous catalytic oxidations using metalloporphyrin complexes with emphasis on ruthenium systems (67 references) [1019];
 Allyl based side-chain protection for SPSS [1020];
 Transition metals in organic synthesis: hydroformylation, reduction, and oxidation. Annual survey covering the year 1990 (897 references) [1021];
 Palladium(II)-catalyzed oxidation, isomerization and exchange of olefins, allylic alcohols and allylic ethers in water and methanol solvents [1022];
 Palladium cluster catalysts supported on chelate resin-metal complexes (13 references) [1023];
 Organic derivatives of manganese(II) in organic synthesis (99 references) [1024];
 A new development of ruthenium complex catalysts. Carbon-carbon bond forming reactions (25 references) [1025];
 Homogeneous metal-catalyzed oxidations by molecular oxygen (71 references) [1026];
 Photocatalysis by transition-metal complexes (164 references) [1027];
 Iron- and cobalt-induced activation of hydrogen peroxide and dioxygen for the selective oxidation-dehydrogenation and oxygenation of organic molecules (209 references) [1028];
 Transition metal-promoted reactions of main group species and main group-promoted reactions of transition metal species (more than 76 references) [1029];
 New routes to biologically active heterocyclic natural products (12 references) [1030];
 New development in catalytic asymmetric hydrogenation (50 references) [1031];
 Cyclizations made easy by transition metal catalysts (20 references) [1032];
 High-velocity palladium catalysis (10 references) [1033];

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