

**TRANSITION METALS IN ORGANIC SYNTHESIS**  
**ANNUAL SURVEY COVERING THE YEAR 1988**

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## I. General Comments

This annual survey covers the literature for 1988 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.

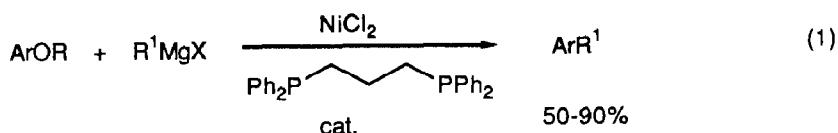
The papers in this survey are grouped primarily by reaction type rather than by organometallic reagent, since the reader is likely to be more interested in the organic transformation effected than the metal causing it. Oxidation, reduction, and hydroformylation reactions are specifically excluded, and 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.

## II. Carbon-Carbon Bond-Forming Reactions

### A. Alkylation

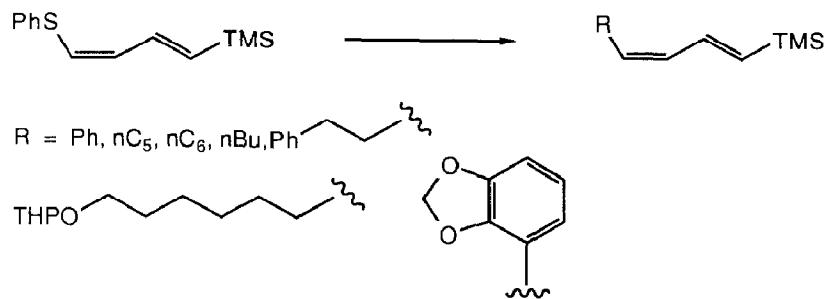
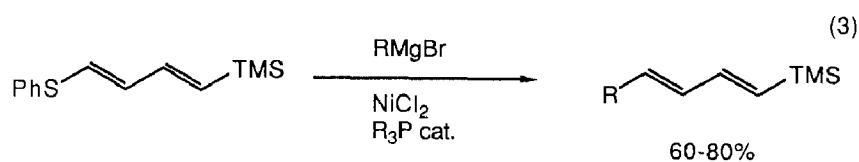
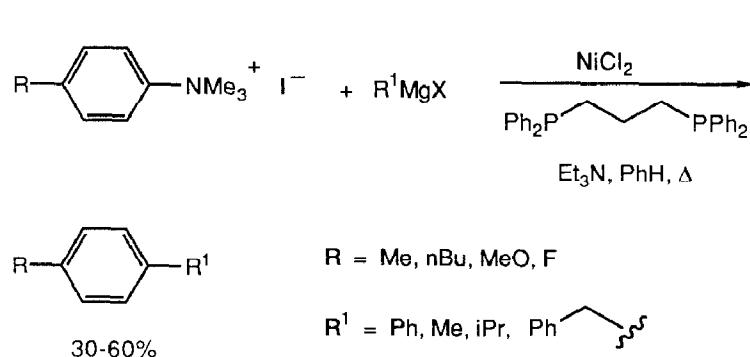
#### 1. Alkylation of Organic Halides, Tosylates, Triflates, Acetates and Epoxides

Transition metal catalyzed reactions of Grignard reagents with organic halides continues to be a useful synthetic method. Substituted styrenes were prepared by the palladium(0) catalyzed reactions of vinyl Grignard reagents with aryl iodides [1]. Nickel(II) and palladium(II) salts catalyzed the reactions of Grignard reagents with 1-bromo-2-phenylthioethane to produce E or Z olefins stereospecifically [2]. Aryl ethers were alkylated by Grignard reagents in the presence of nickel phosphine complexes (equation 1) [3], as were quaternary anilinium salts (equation 2) [4]. 1-Phenylthio-4-trimethylsilyldienes were alkylated by Grignard reagents in the presence of transition metal catalysts (equation 3) [5].



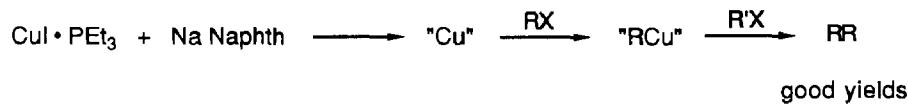
$\text{Ar} = \text{Ph}, 2\text{-Naphth}, \text{pMeOPh}$

$\text{R} = \text{Et}, \text{nPr}, \text{iPr}, \text{secBu}, \text{nBu}, \text{Ph}$

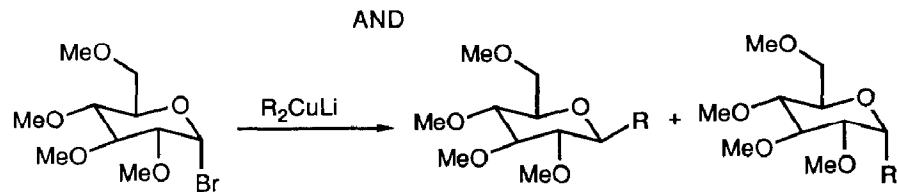
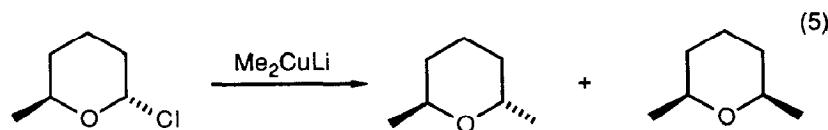
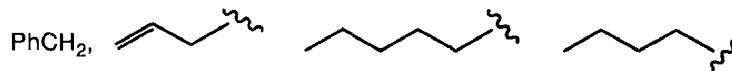
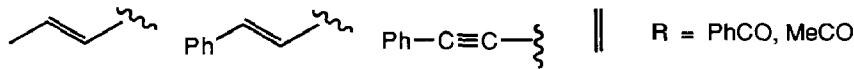


Organocupper chemistry continues to be extensively used for the alkylation of halides. Functionalized organocuprates were prepared from functionalized halides and active copper metal produced by the reduction of CuI-PEt<sub>3</sub> with sodium naphthalenide. These reagents alkylated organic halides in excellent yield (equation 4) [6].  $\alpha$ -Chloro and  $\alpha$ -bromoethers were cleanly alkylated by alkylcuprates (equation 5) [7]. Perfluorinated halocyclohexenes were converted to cuprates and alkylated by functionalized organic halides (equation 6) [8]. Functionalized benzyl halides were converted to cuprates via their zinc salts, and alkylated by halides (equation 7) [9].

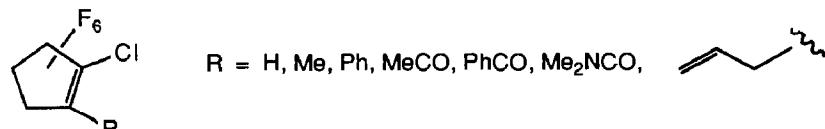
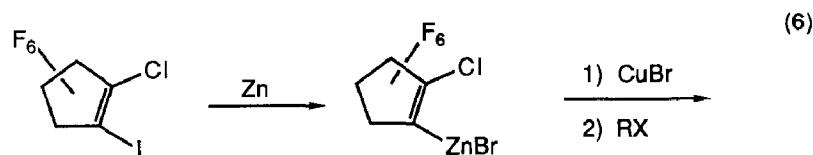
(4)



$\text{R} = \text{Ph}, \text{oNO}_2\text{Ph}, \text{oNCPH}, \text{C}_6\text{F}_5, \text{pMeO}_2\text{CPh}, \text{pPhCOPh}, \text{mMePh}$

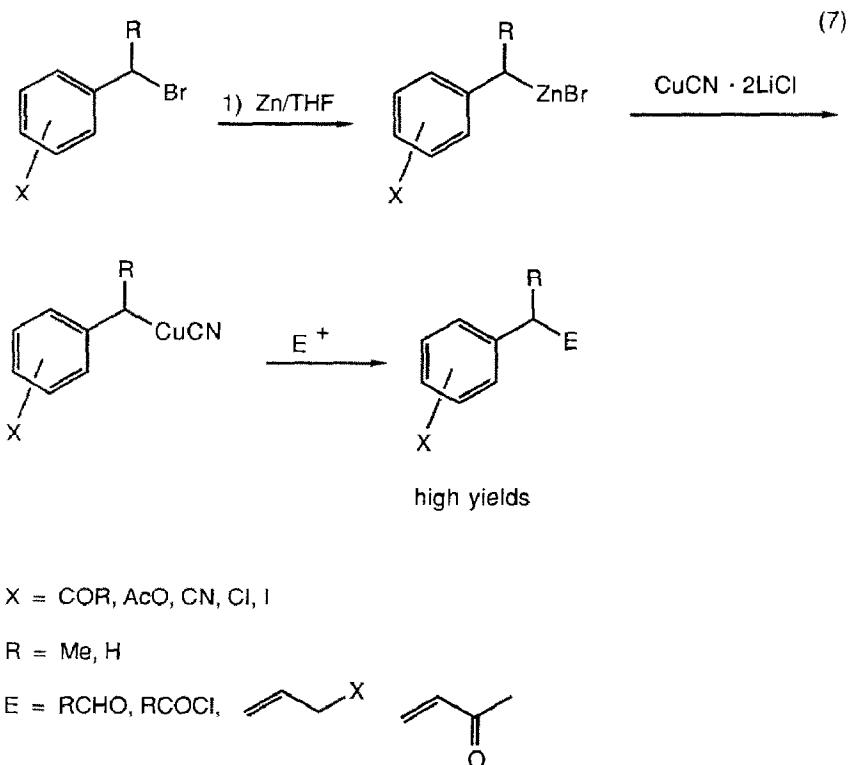


$\text{R} = \text{Me}$	46%	20 : 1
$\text{Bu}$	40%	20 : 1
$\text{Ph}$	25%	20 : 1

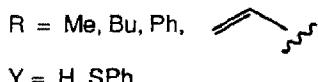
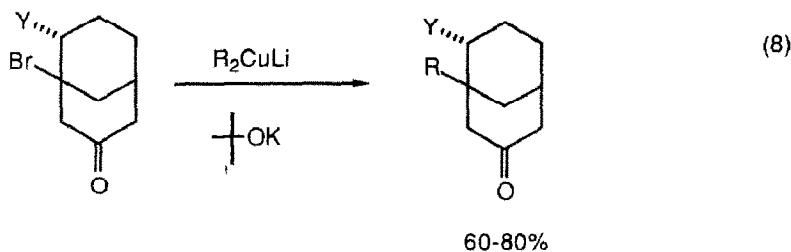


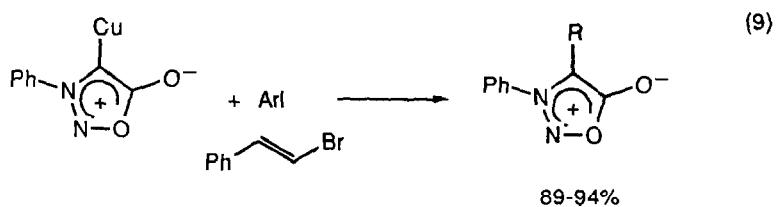
50-90%

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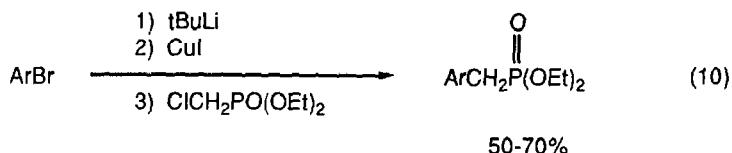


Bridgehead bromides were alkylated by organocuprates (equation 8) [10]. Heterocyclic cuprates were arylated by aryl iodides (equation 9) [11], and aryl halides were coupled to chloromethylphosphonates via organocupper intermediates (equation 10) [12]. 1,4-Dicopperbutanes formed spirocyclic compounds when treated with  $\beta$ -chloroenones (equation 11) [13]. Insect sex pheromones were synthesized by reactions of triethylamine-coordinated vinyl cuprates [14].

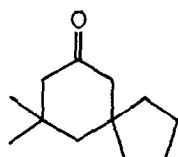
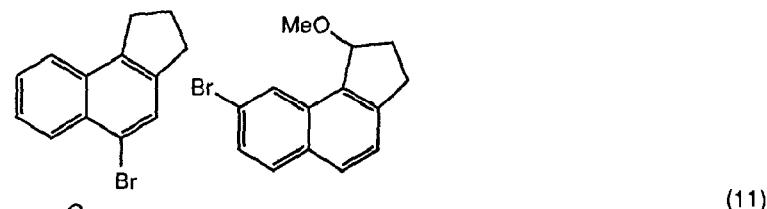




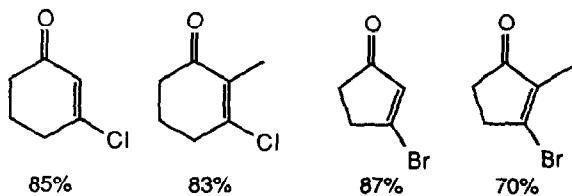
$\text{R} = \text{Ph}, \text{pNO}_2\text{Ph}, \text{pMeOPh}, \text{Ph}-\text{CH}_2-\text{CH}_2-\text{CH}_2-$



$\text{Ar} = \text{Ph}, 1\text{-Naphth}, 2\text{-Naphth},$

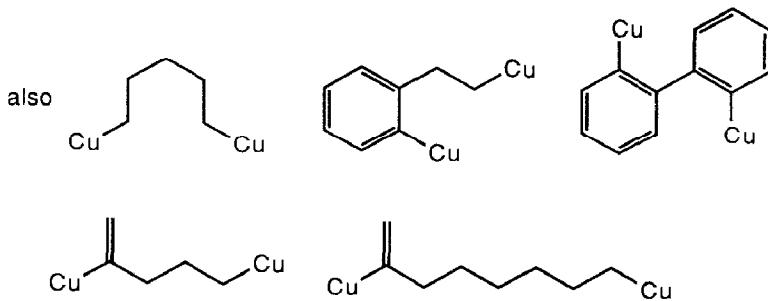


96%

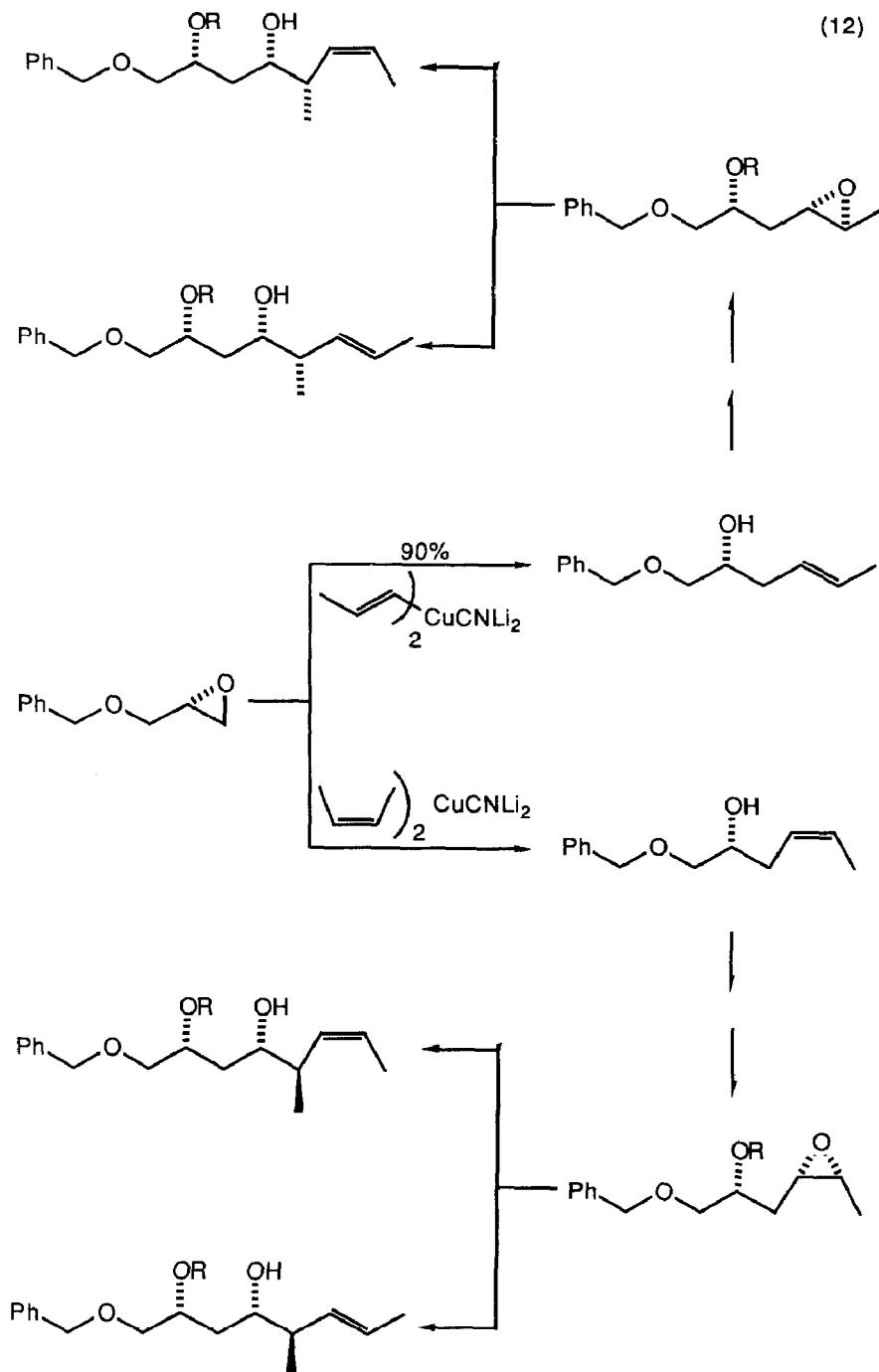


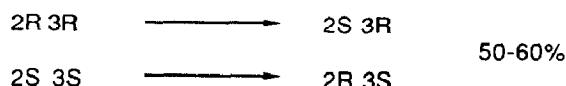
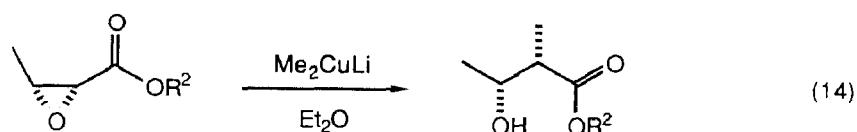
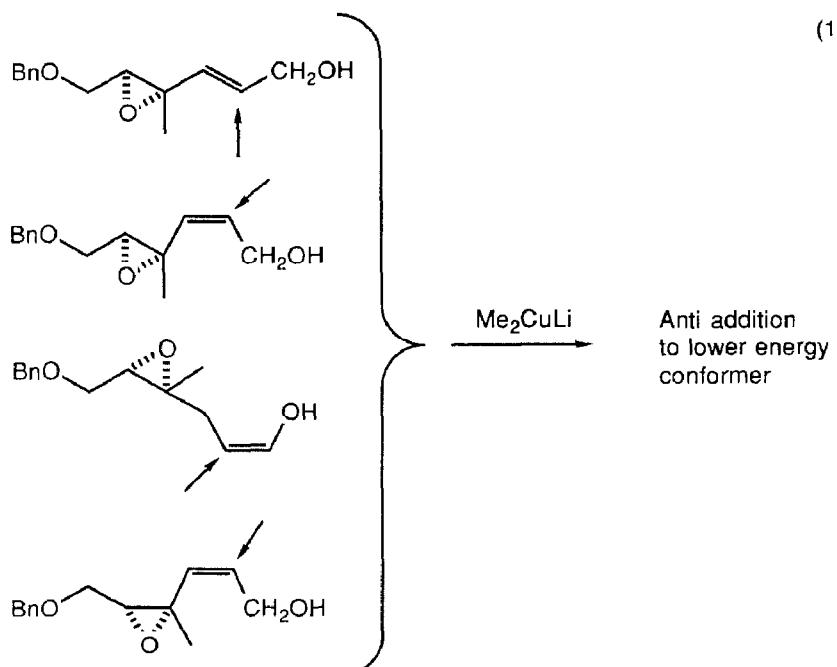
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(equation 11 continued)

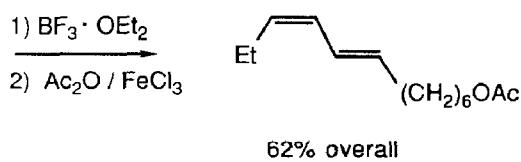
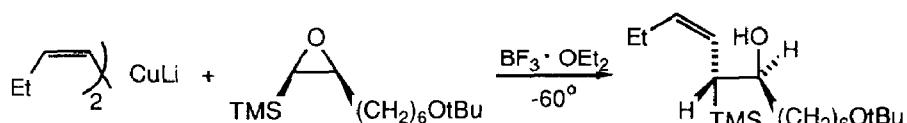


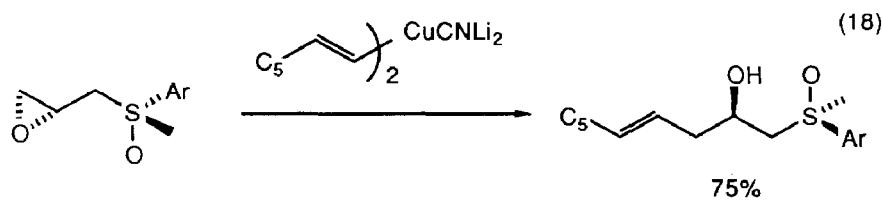
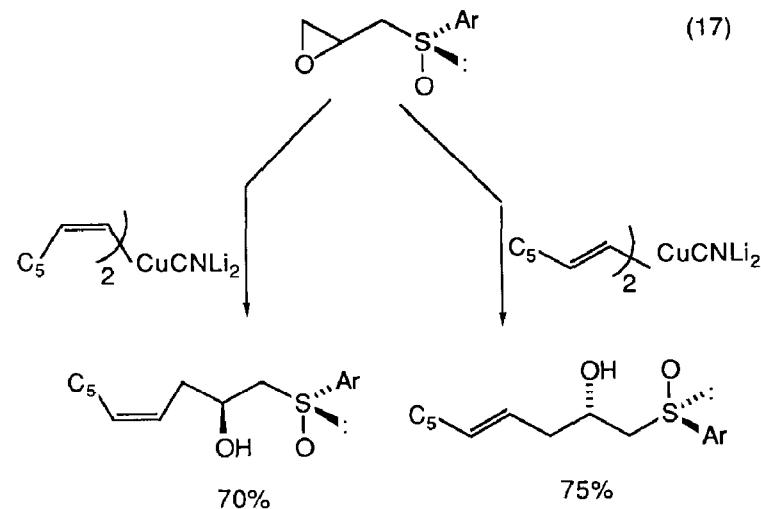
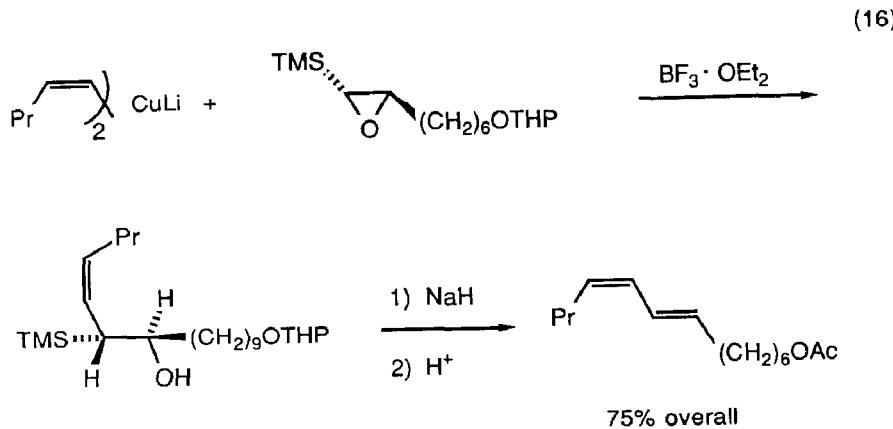
The reaction of chiral epoxides with organocopper species has been used to synthesize a number of complex systems (equation 12) [15], (equation 13) [16,17], (equation 14) [18], (equations 15 and 16) [19]. Stereoselectivity was also high with chiral epoxy sulfoxides (equations 17 and 18) [20].





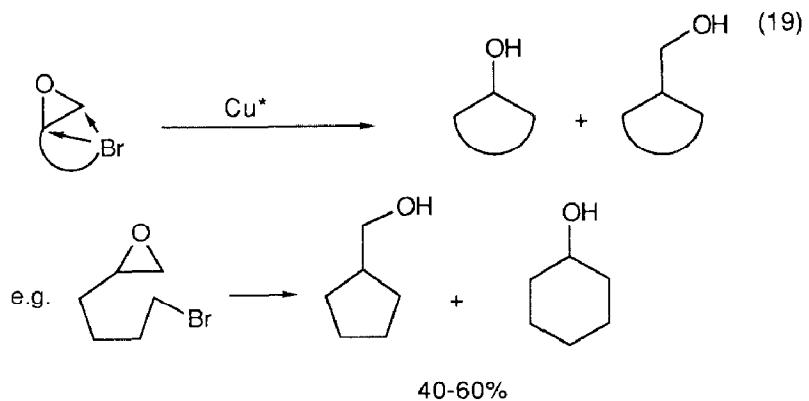
(15)



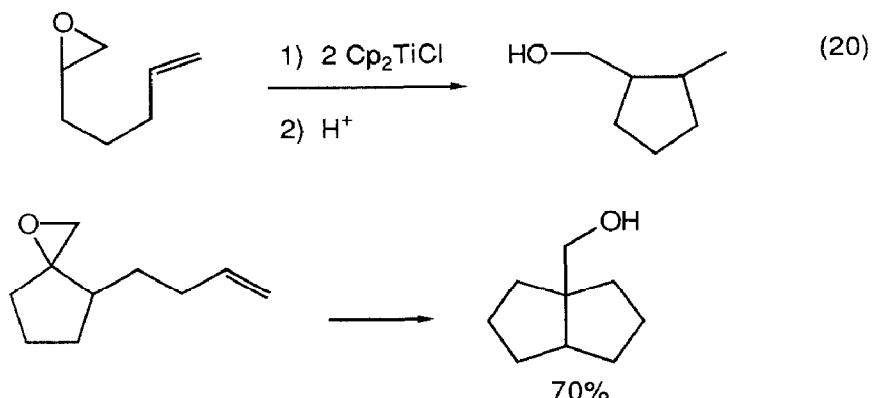
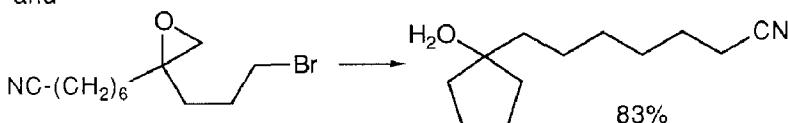


Treatment of  $\omega$ -bromo epoxides with activated copper resulted in cyclization (equation 19) [21].  $\omega$ -Olefinic epoxides underwent a related cyclization when treated with  $Cp_2TiCl$  (equation 20) [22]. Nickel(0) complexes promoted the alkylation of epoxides by methyl iodide (equation 21) [23].

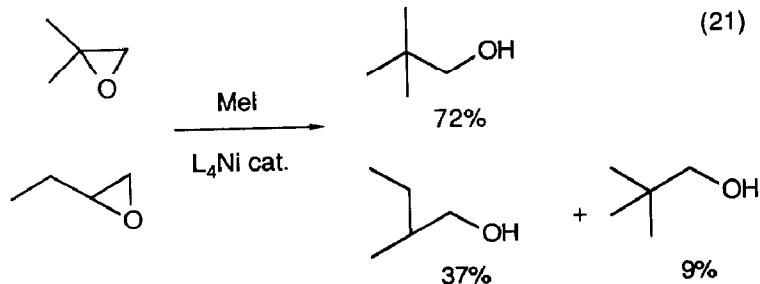
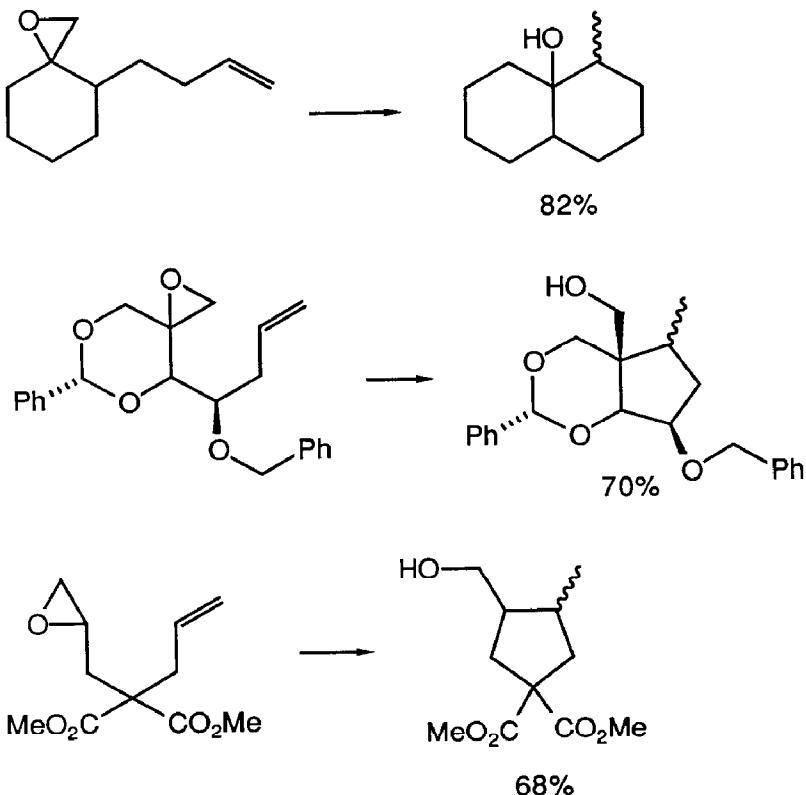
Palladium(0) complexes catalyzed the alkylation of aryl halides (equations 22 and 23) [24] and heteroaromatic halides (equation 24) [25,26] by stabilized carbanions. Aryl iodides were coupled to transition metal carbonyl anions using palladium(0) catalysts [27].

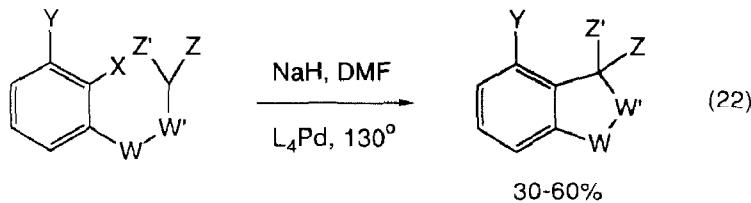


and



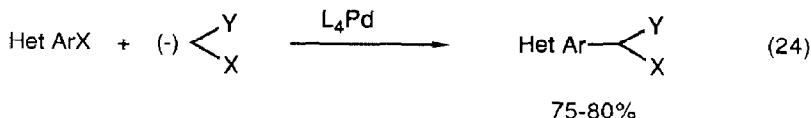
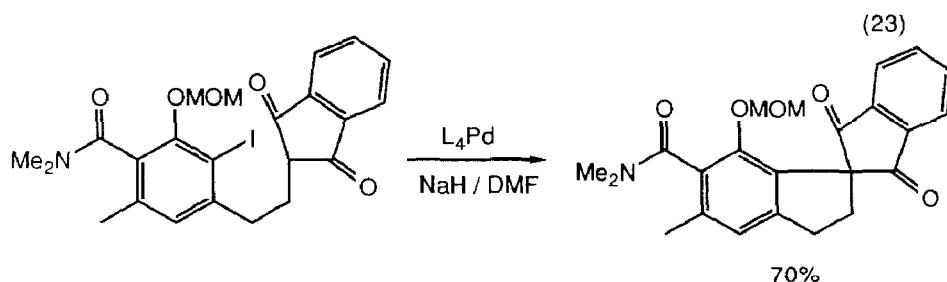
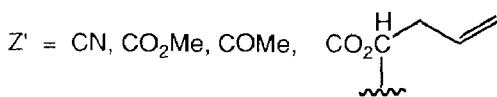
(equation 20 continued)



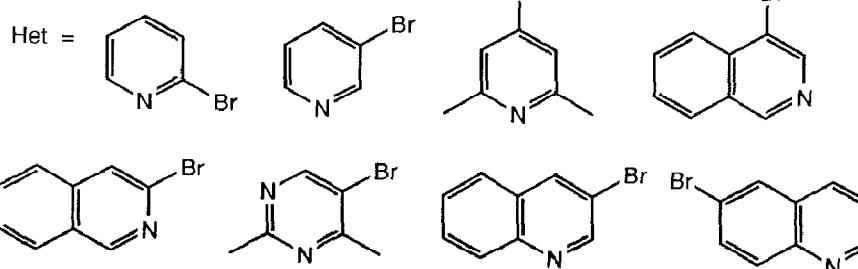


$W = \text{CH}_2, \text{NMe}; W' = \text{CH}_2, (\text{CH}_2)_2, \text{SO}_2, \text{CO}$

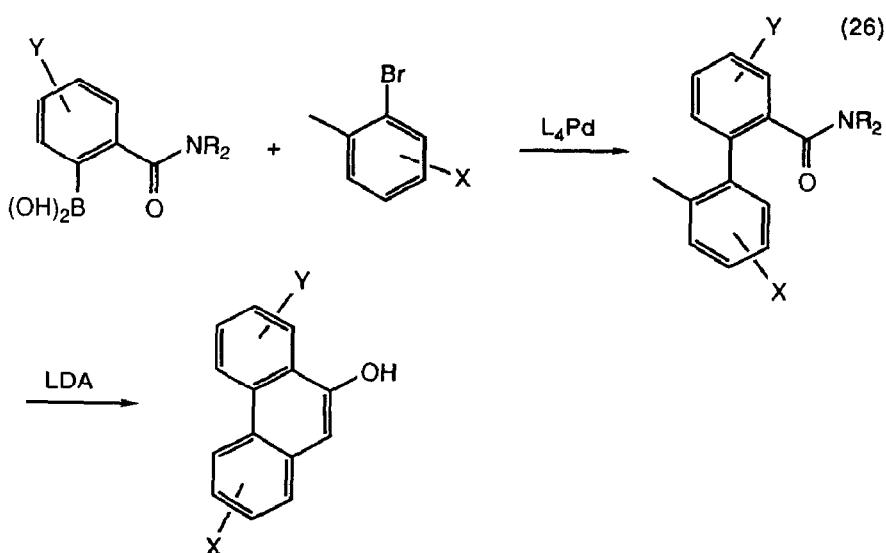
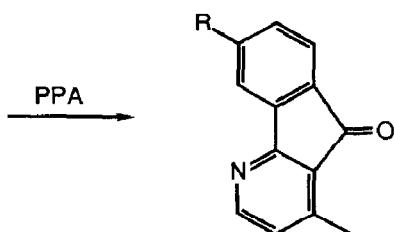
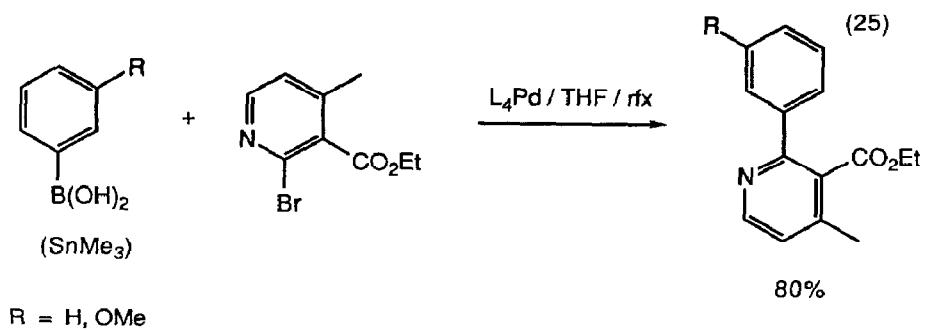
$X = \text{Br}, \text{I}; Z = \text{CN}, \text{CO}_2\text{Me}, \text{CO}_2\text{Et}, \text{H}, \text{Me}$

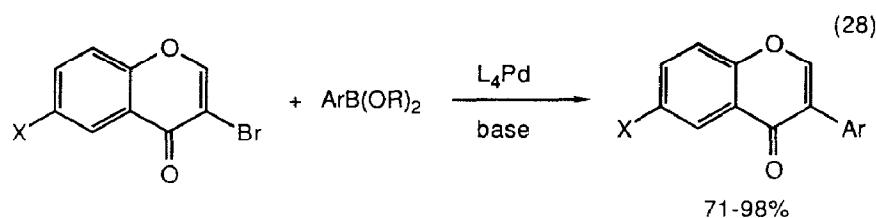
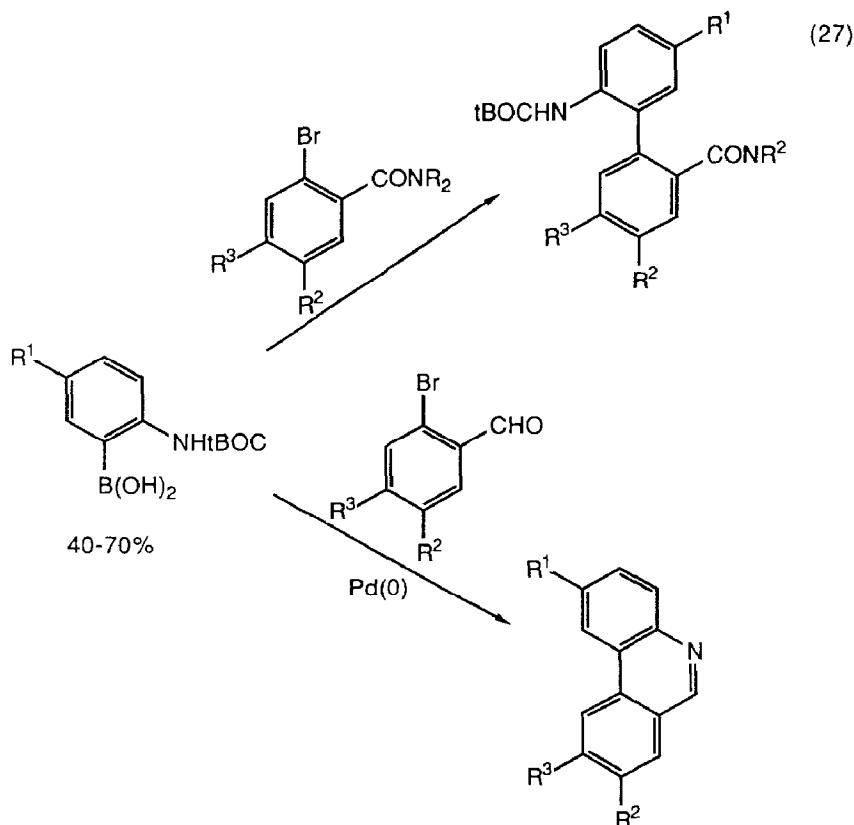


$X, Y = \text{CN}, \text{CO}_2\text{Et}, \text{SO}_2\text{Ph}, \text{PO}(\text{OEt})_2$



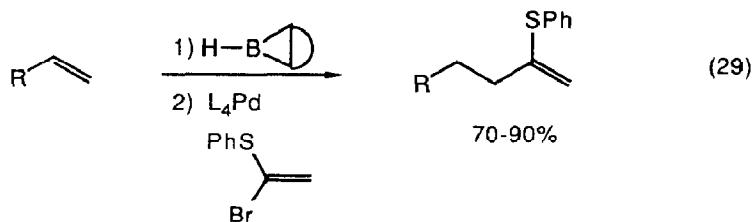
The choice of reagents in palladium-complexed catalyzed reactions of organometallic compounds with organic halides has been investigated [28]. Palladium(0) catalyzed the coupling of aryl boranes with aryl halides (equation 25) [29], (equation 26) [30], (equation 27) [31], and vinyl halides (equation 28) [32]. Alkyl (equation 29) [33] and vinyl boranes (equation 30) [34] underwent a similar coupling.

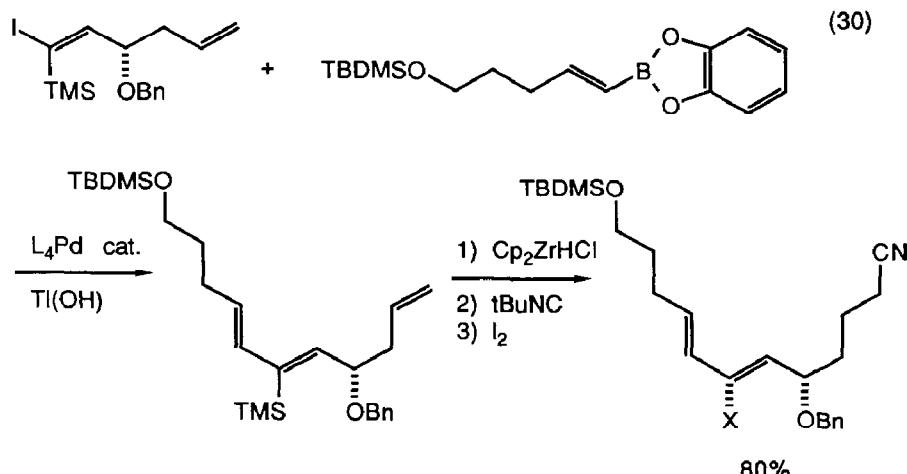




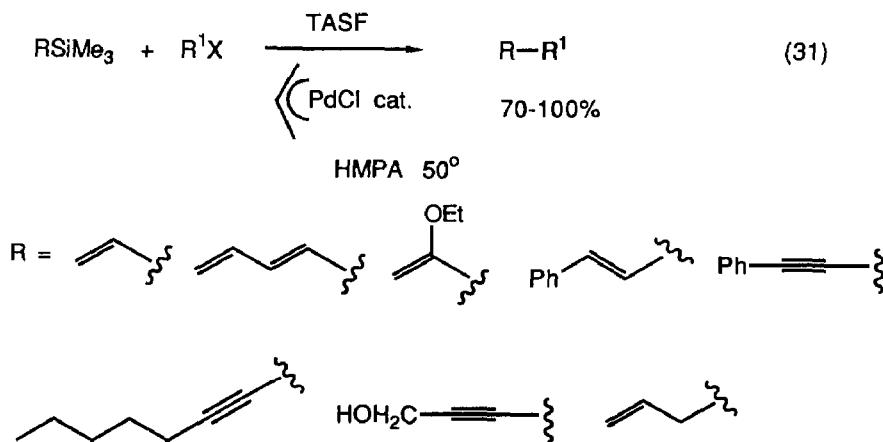
X = H, Cl, MeO

Ar = pMePh, pMeOPh, oMePh, pFPh, pNO<sub>2</sub>Ph, 1-naphth  
2,4,6-Me<sub>3</sub>Ph (47%)

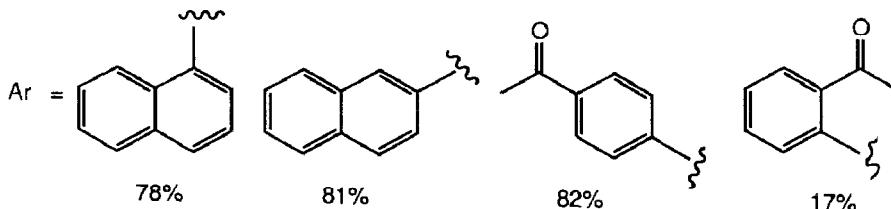
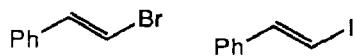
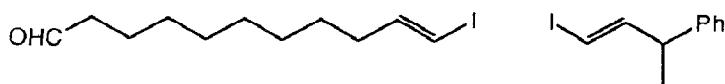
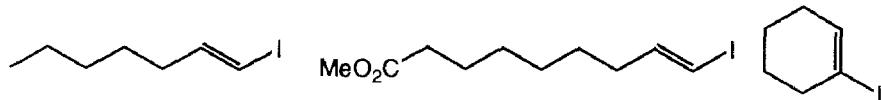
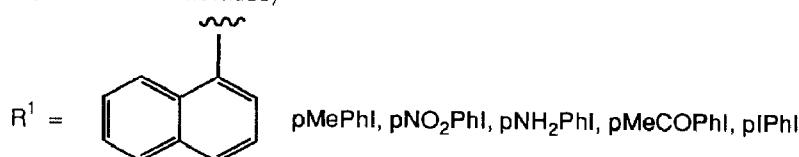




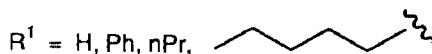
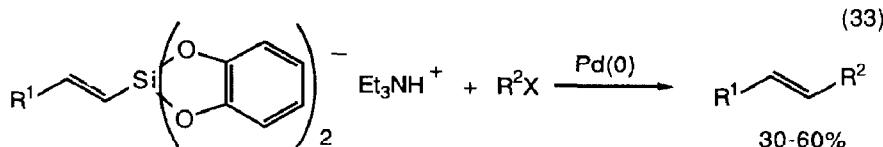
Transmetalation from silicon to palladium has finally been achieved, and a number of palladium(0) catalyzed alkylations of halides by silanes have been developed. Treatment of aryl and vinyl halides with trimethyl(alkenyl) silanes in the presence of a palladium(0) catalyst resulted in efficient alkylation (equation 31) [35]. Aryl halides were methylated by  $\text{Me}_3\text{SiF}_2^-$  in the presence of palladium(0) catalysts (equation 32) [36]. Vinyl silyl "ate" complexes also efficiently alkylated aryl and vinyl halides under palladium(0) catalysis (equation 33) [37]. Organoaluminum reagents alkylated halogenonucleosides in the presence of palladium(0) catalysts [38].



(equation 30 continued)



pNO<sub>2</sub>Ph, 86%; pMeO<sub>2</sub>CPh, 84%; mMeO<sub>2</sub>CPh, 67%; mMeO<sub>2</sub>CCH<sub>2</sub>Ph, 59%

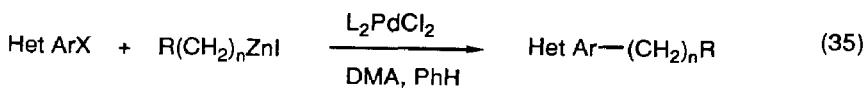
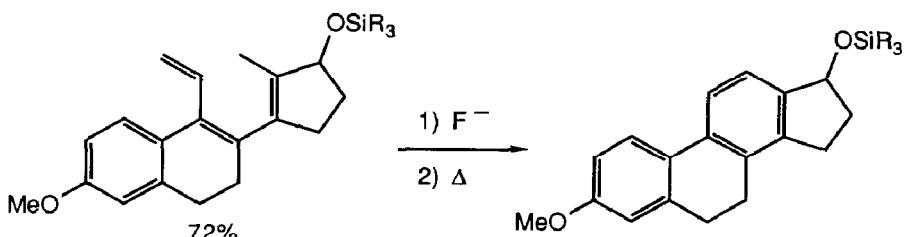
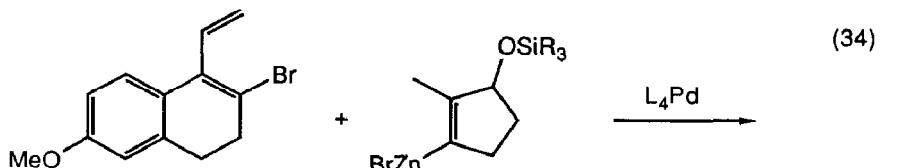


$R^2 = Ph, pNO_2Ph, pMePh, pMeOPh, pCH_3COPh, pCHOPh, 1\text{-Naphth}$

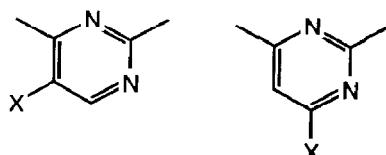
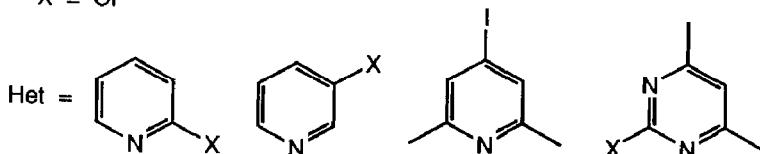


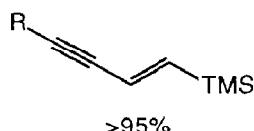
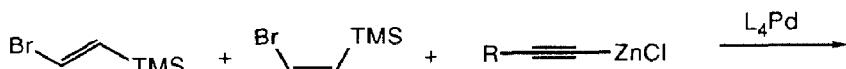
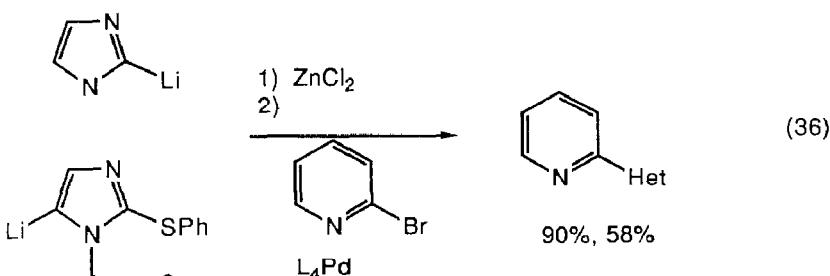
$X = I, OTf$

Coupling by transmetalation from zinc to palladium has also been further developed. The steroid nucleus was assembled using palladium(0) catalyzed alkylation of a vinyl halide by a vinyl zinc reagent (equation 34) [39]. Alkyl zinc reagents alkylated heteroaromatic halides in the presence of palladium catalysts (equation 35) [40], (equation 36) [41]. Alkynyl zinc reagents alkylated vinyl halides containing trimethylsilyl groups (equation 37) [42] and alkylborane groups (equation 38) [43], (equation 39) [44] in the presence of palladium catalysts. Aryl iodides were alkylated by perfluorovinyl zinc reagents in the presence of palladium(0) catalysts (equation 40) [45].

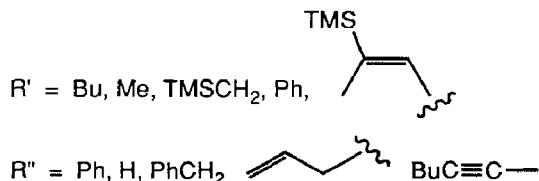
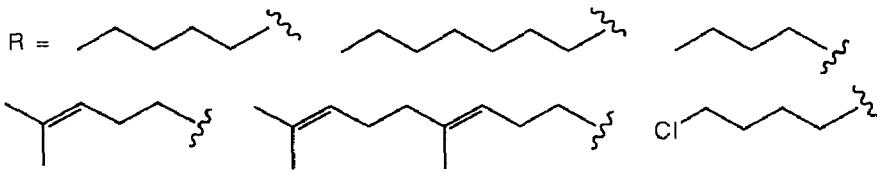
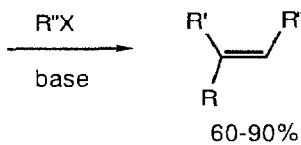
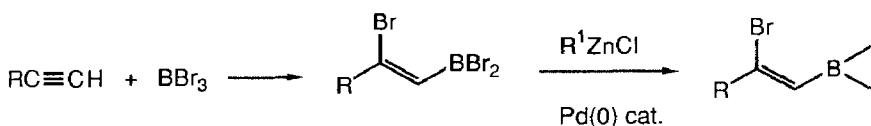


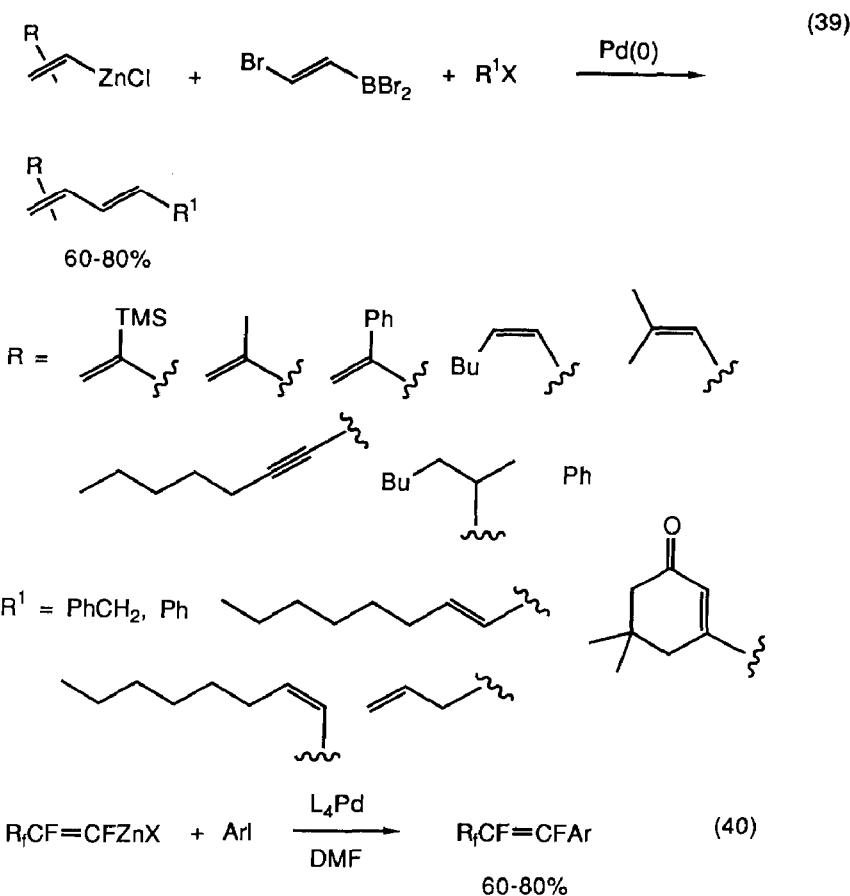
$n = 1, 2, 3$   
 $X = Cl$





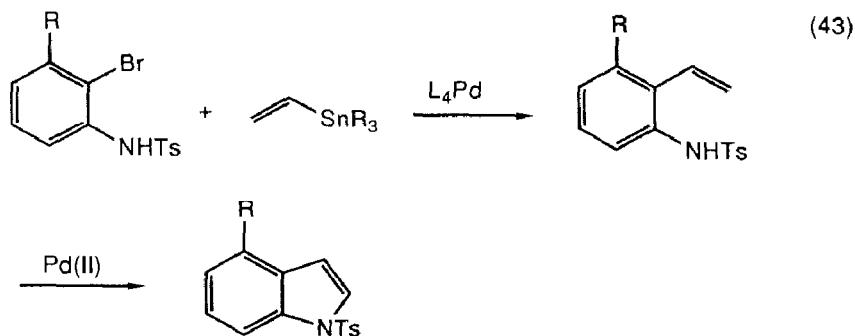
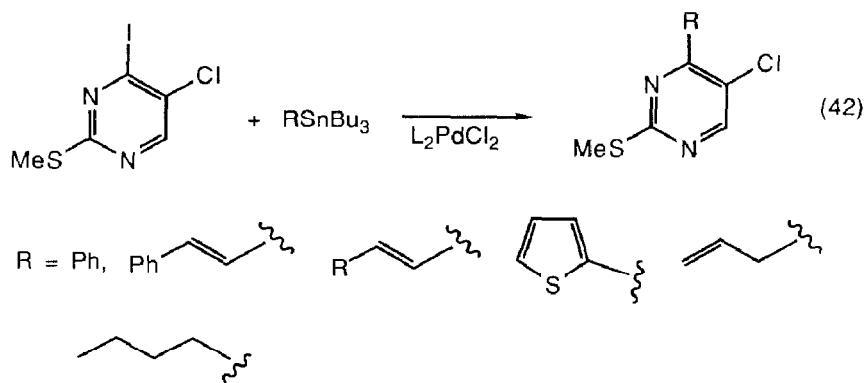
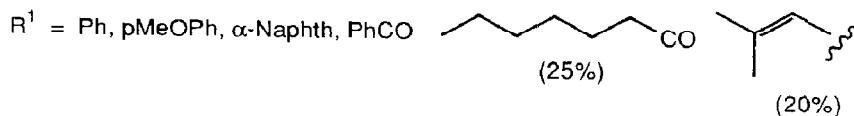
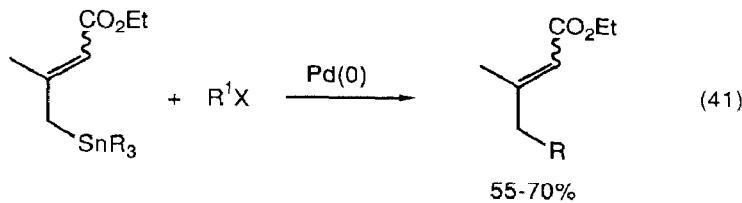
(38)

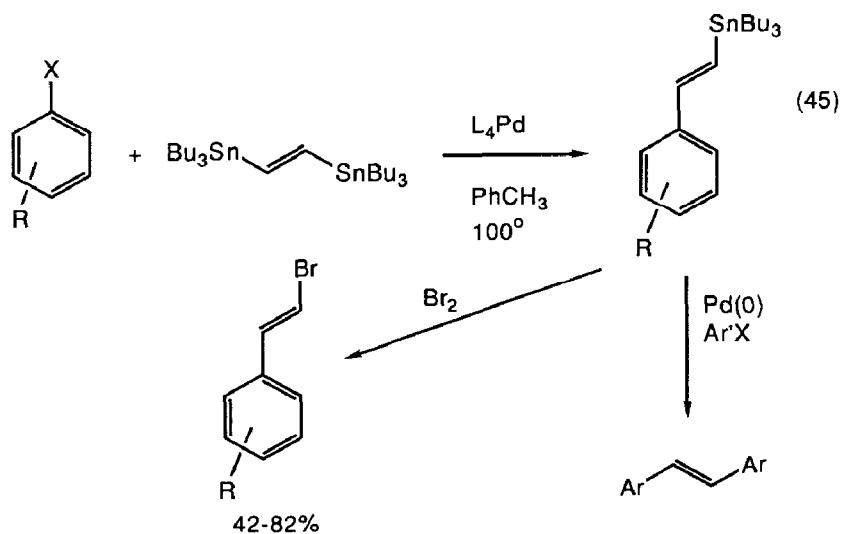
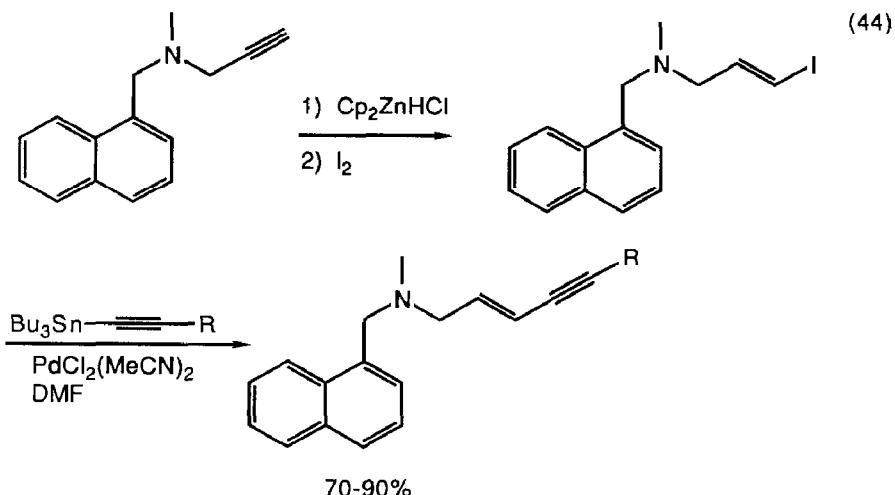




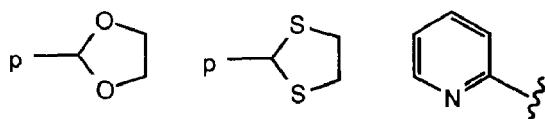
$\text{Ar} = \text{Ph}, \text{oNO}_2\text{Ph}, \text{pMeOPh}, \text{iPrPh}, \text{oCF}_3\text{Ph}, 2,5\text{Cl}_2\text{Ph}, \text{mNO}_2\text{Ph}, \text{pClPh}, 1,4\text{Cl}_2\text{Ph}$

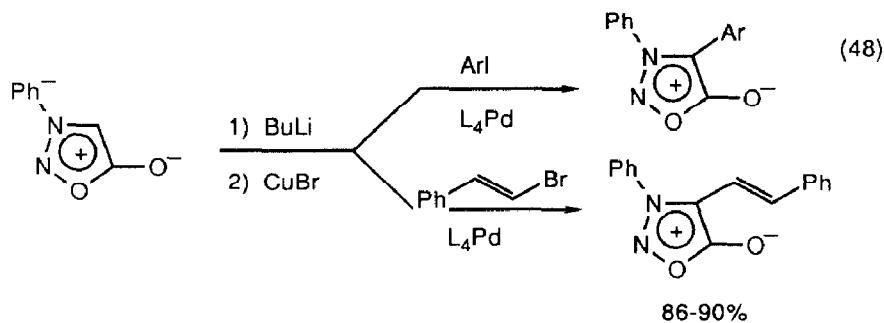
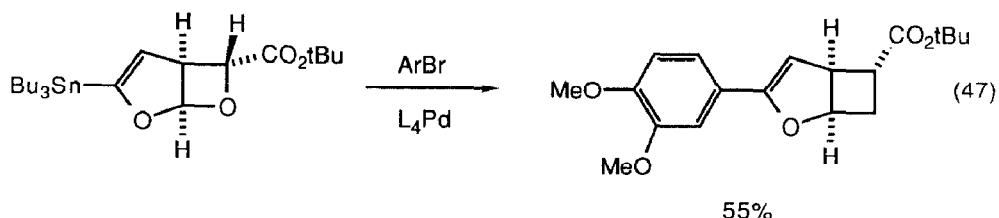
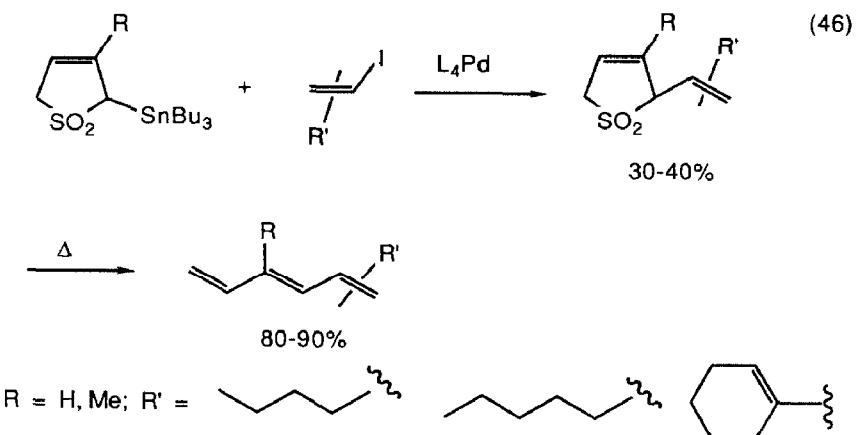
Transmetallation from tin to palladium has also accounted for a number of processes for alkylating organic halides.  $\gamma$ -Stannylo enoate esters were alkylated by aryl and acyl halides using a palladium(0) catalyst (equation 41) [46]. 4-Iodopyrimidines were alkylated by tin reagents in the presence of palladium(0) catalysts (equation 42) [47]. Indoles were prepared from o-bromoanilines by a two step palladium catalyzed process (equation 43) [48]. Vinyl iodides were alkynylated by acetylenic tin reagents again using palladium catalysis (equation 44) [49]. Unsymmetrical stilbenes were prepared by the diarylation of 1,2-distannylolefins (equation 45) [50]. Substituted trienes were synthesized from vinyl iodides and tin derivatives of the  $\text{SO}_2$  adduct butadiene or isoprene (equation 46) [51]. Palladium catalyzed the arylation of vinyl tin (equation 47) [52] and heteroaromatic copper reagents (equation 48) [53].



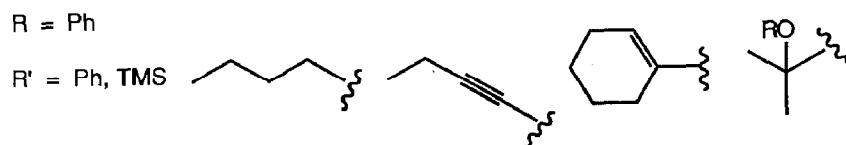
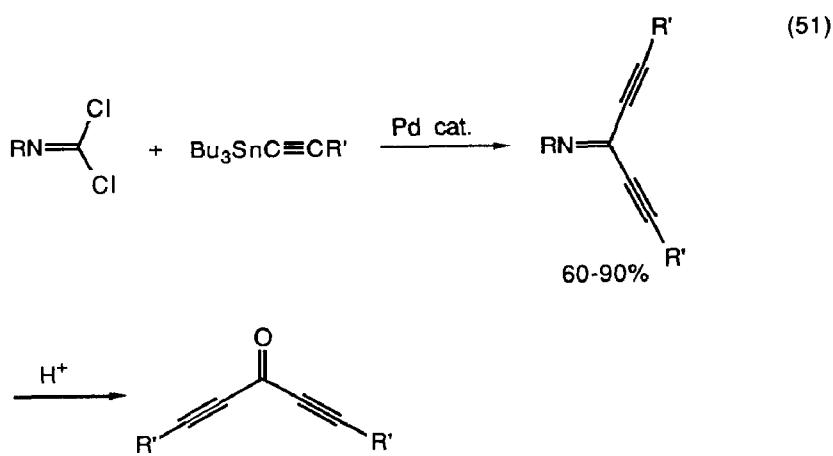
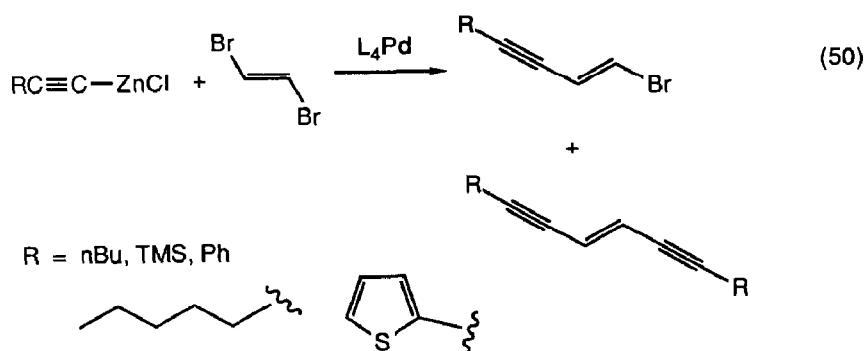
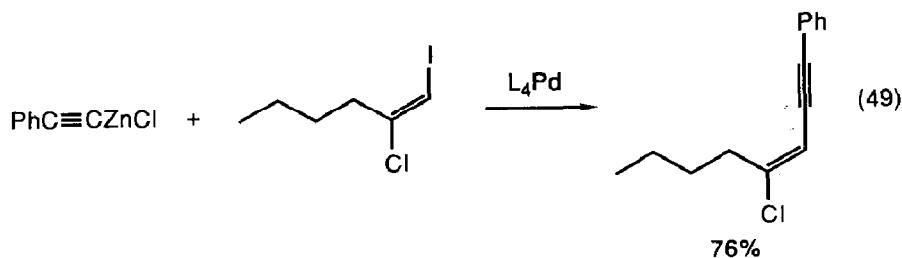


R = oAc, pAc, oOMe, mOMe, pOMe, pNO<sub>2</sub>, pCF<sub>3</sub>, pCH<sub>2</sub>OH, pCH<sub>2</sub>CN, pCO<sub>2</sub>Et, oPH

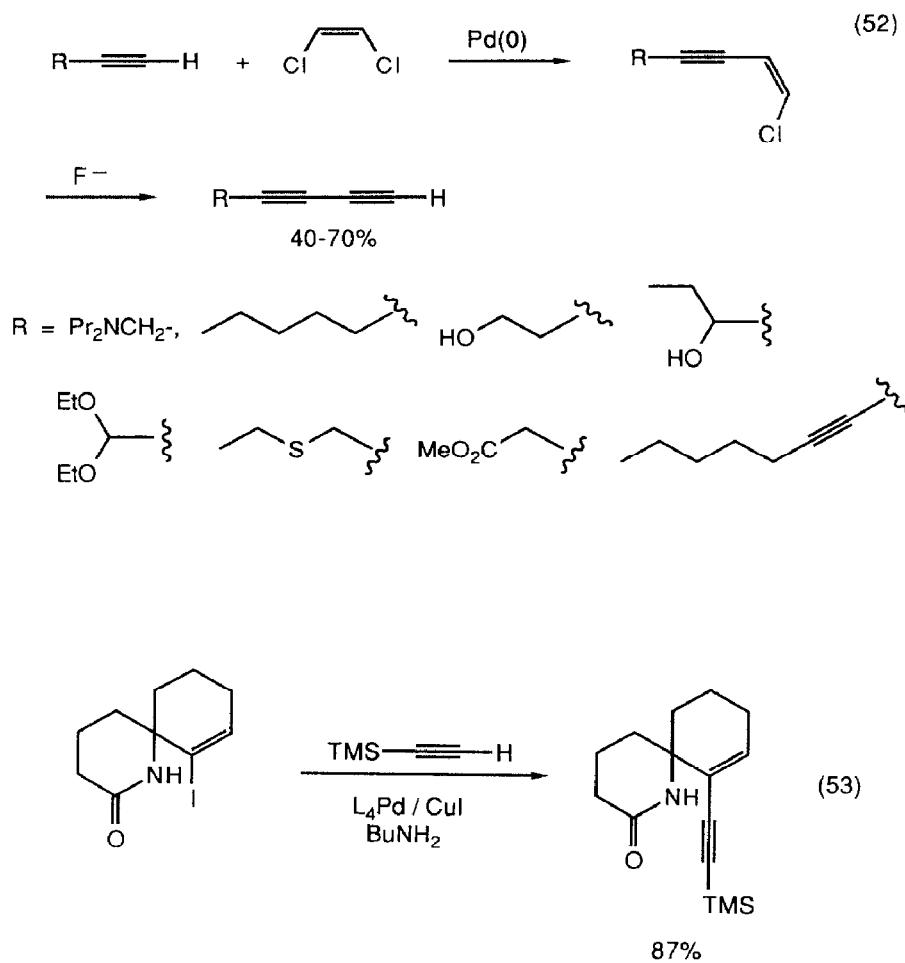


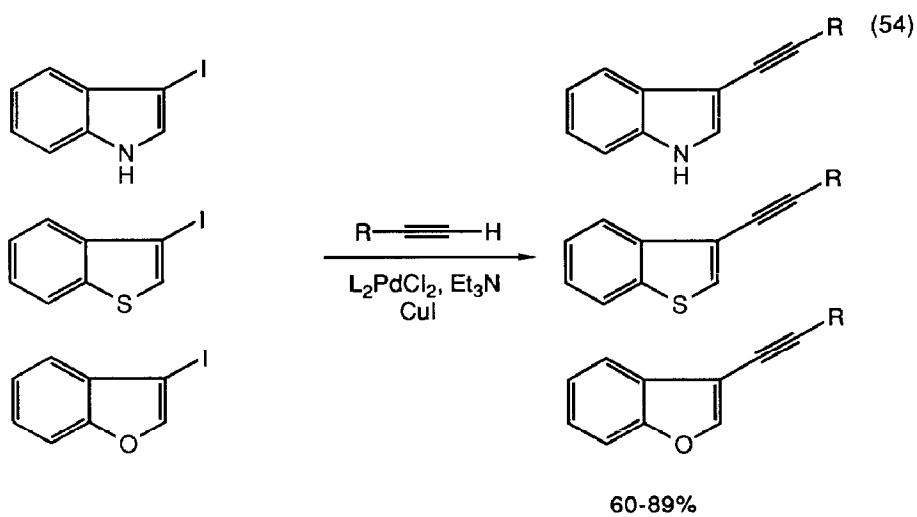


Alkynylzinc chlorides alkylated vinyl iodides in the presence of palladium catalysts (equation 49) [54]. Vinyl bromides (equation 50) [55] and imidoyl chloride (equation 51) [56] underwent a similar reaction.



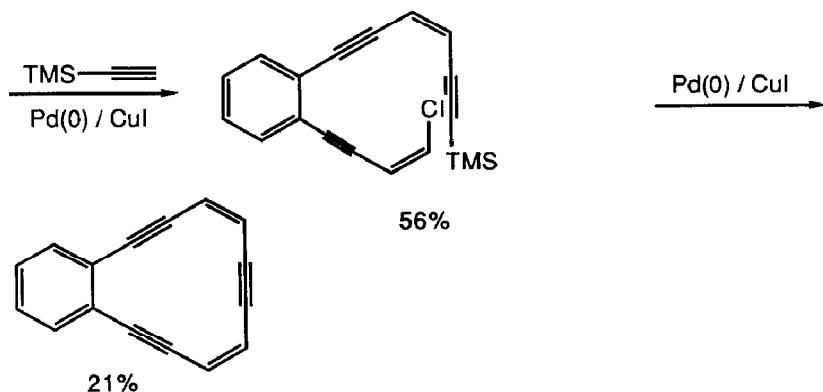
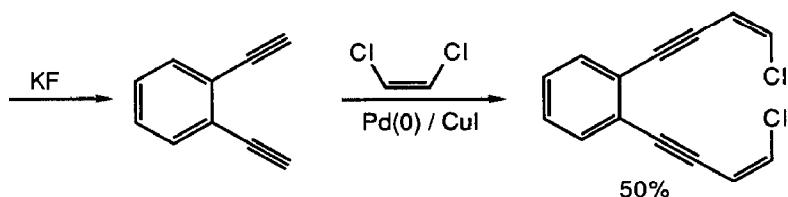
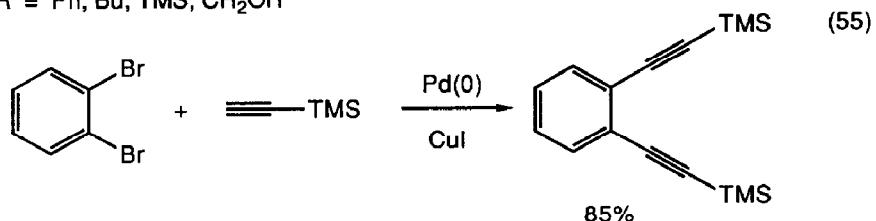
The palladium/copper catalyzed direct coupling of terminal alkynes with vinyl halides has found a number of interesting synthetic applications (equation 52) [57], (equation 53) [58], (equation 54) [59], (equation 55) [60], (equation 56) [61], (equation 57) [62]. Phase transfer conditions were used to develop the palladium catalyzed arylation of terminal alkynes by aryl iodides [63].

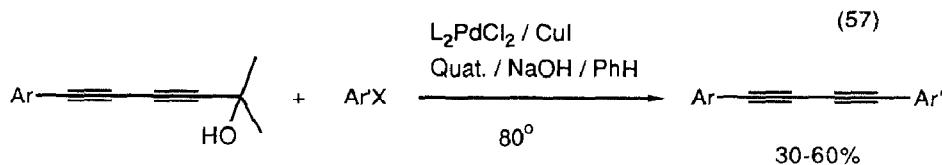
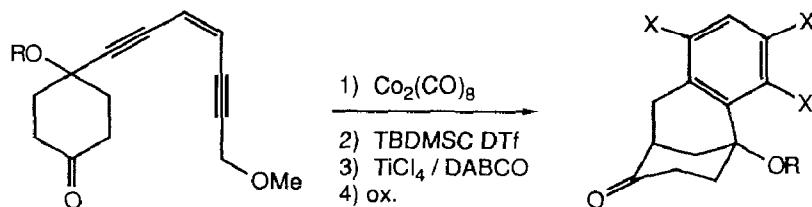
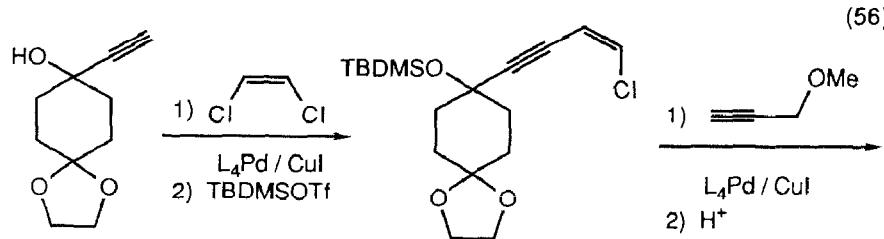




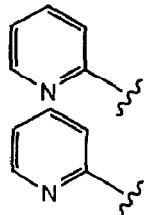
$\text{R} = \text{Ph}, \text{Bu}, \text{TMS}, \text{CH}_2\text{OH}$

60-89% (54)

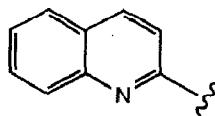
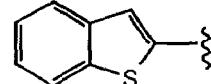




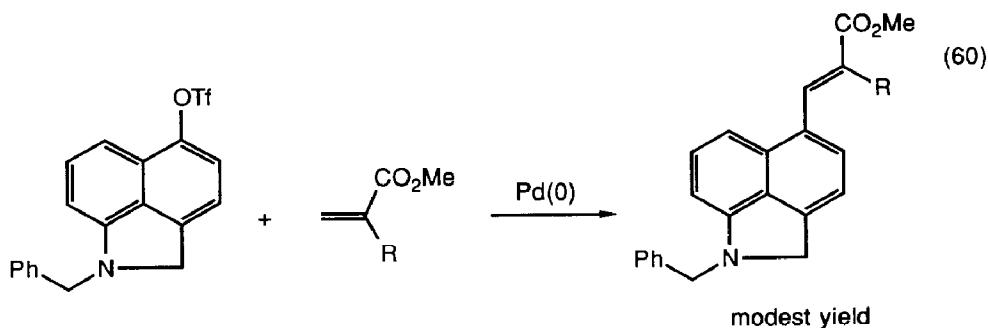
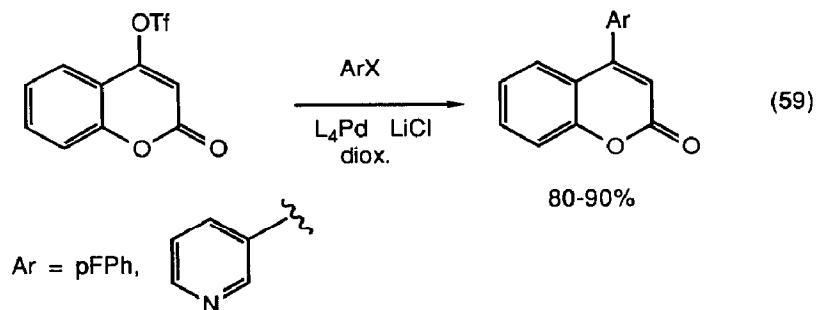
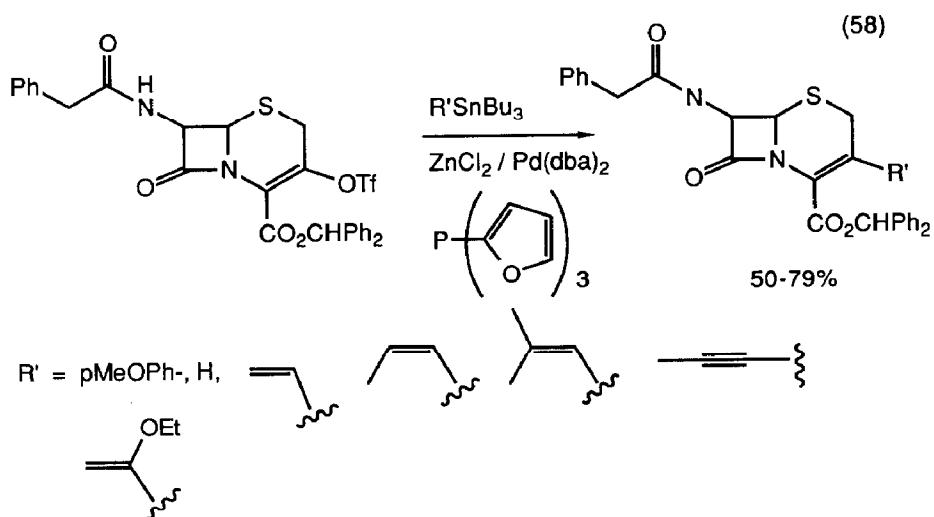
$\text{Ar} = \text{Ph}, p\text{MePh}, p\text{MeOPh},$

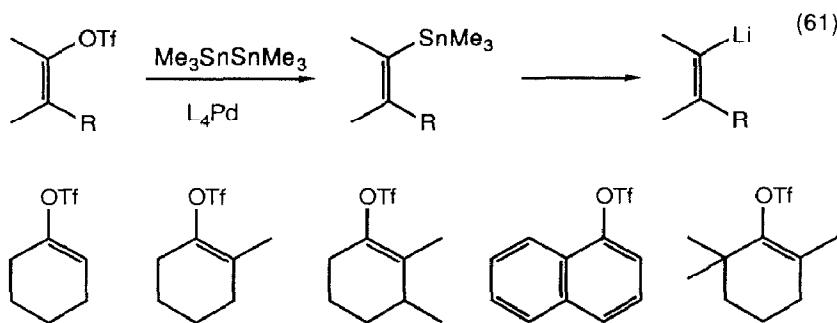


$\text{Ar}' = \text{Ph}, p\text{MePh}, p\text{MeOPh},$



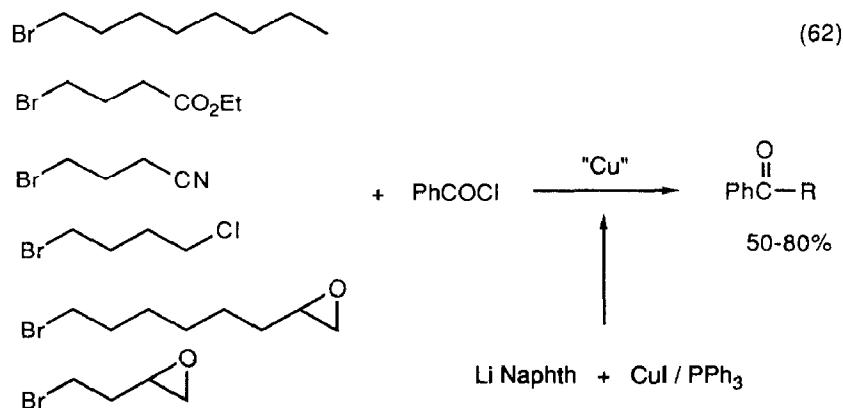
Olefin synthesis via organometallic coupling reactions of enol triflates has been reviewed (46 references) [64]. Palladium(0) complexes catalyzed the reactions of fluoroalkanes sulfonates with allyltributyl tin [65]. Palladium catalyzed alkylation of vinyl triflates by alkyltin reagents was used to synthesize a number of complex organic solvents (equation 58) [66], (equation 59) [67], (equation 60) [68]. Vinyl triflates were converted to vinyl stannanes by a palladium(0) catalyzed reaction of hexamethyl ditin. These were then converted to vinylolithium reagents (equation 61) [69].

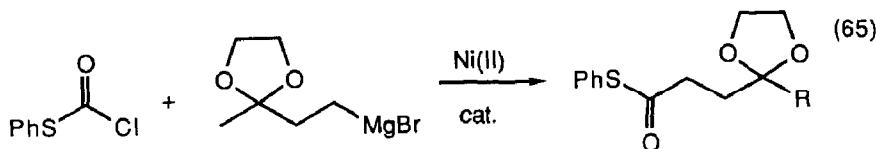
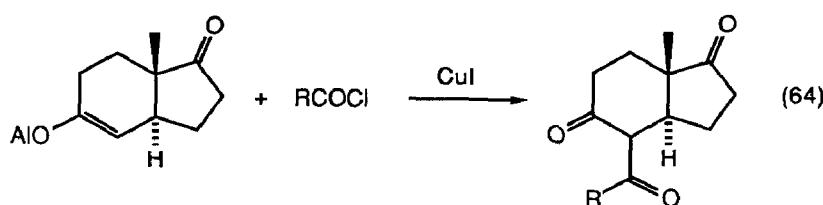
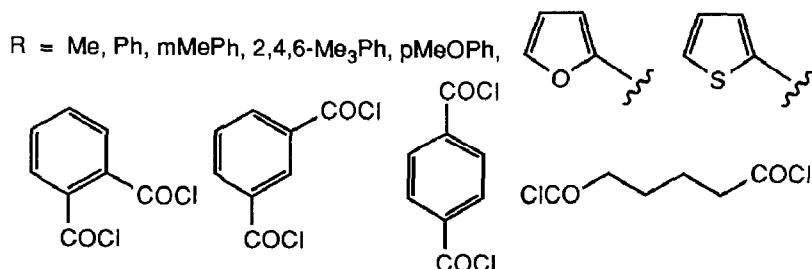




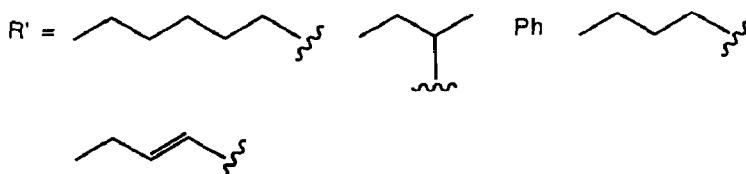
## 2. Alkylation of Acid Derivatives

Benzoyl chloride was alkylated by a wide variety of functionalized organocupper species, prepared from functionalized halides and activated copper metal (equation 62) [70]. Trimethylsilyl copper converted acid halides to acyl silanes (equation 63) [71]. A complex aluminum enolate alkylated acid chlorides in the presence of copper(I) salts (equation 64) [72]. Thiochloroformates were alkylated by Grignard reagents in the presence of nickel(II) salts (equation 65) [73].

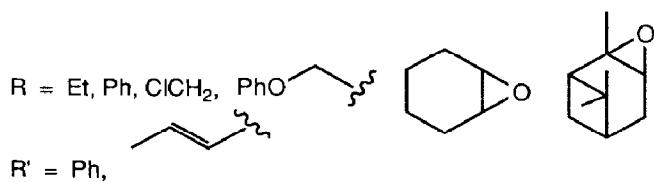
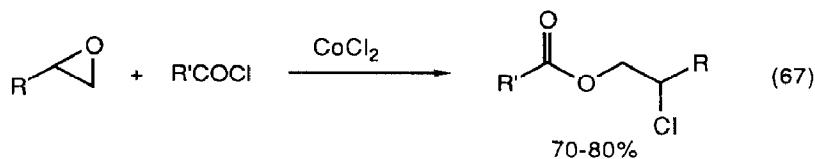
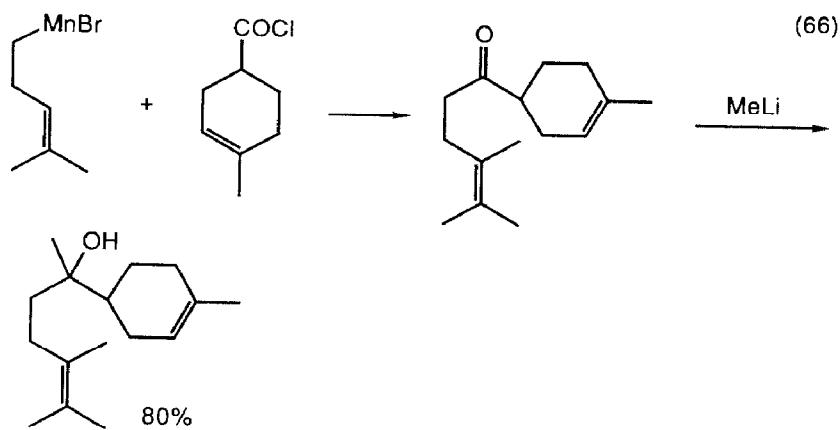


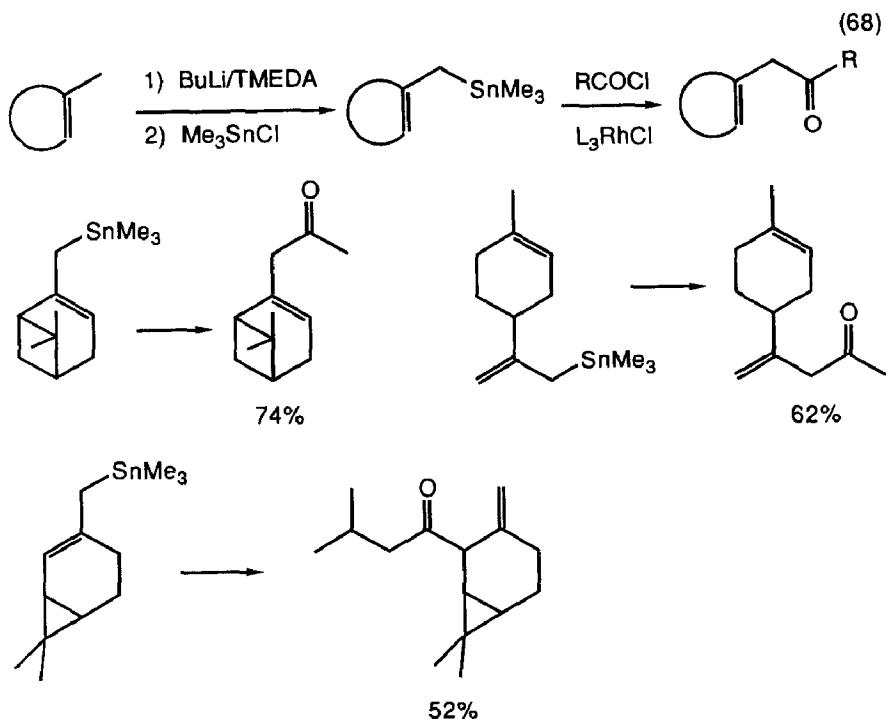


$\text{R}' = \text{H, Me}$



Alkylmanganese complexes alkylated acid halides (equation 66) [74]. Cobalt(II) chloride catalyzed the addition of epoxides to acid chlorides (equation 67) [75]. Rhodium(I) complexes catalyzed the alkylation of acid halides by allyl and benzyl tin reagents (equation 68) [76].



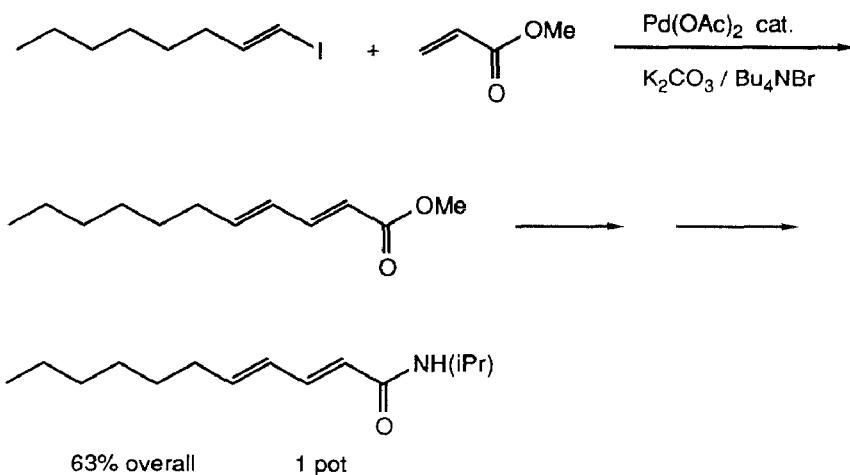


A dissertation entitled "Aniterative approach to the synthesis of enantiomerically pure polypropionate compounds based upon the Claisen rearrangements and Pd(0)-catalyzed alkylation of  $\gamma$ -butyrolactone templates" has appeared [77]. Palladium(0) complexes catalyzed the vinylation of phenylpropioloyl chloride [78], and of arylsulfonyl chlorides [79]. Fluorinated carbanions were produced by palladium catalyzed decomposition of allyl perfluoro-2-methyl-3-oxohexanoate [80].

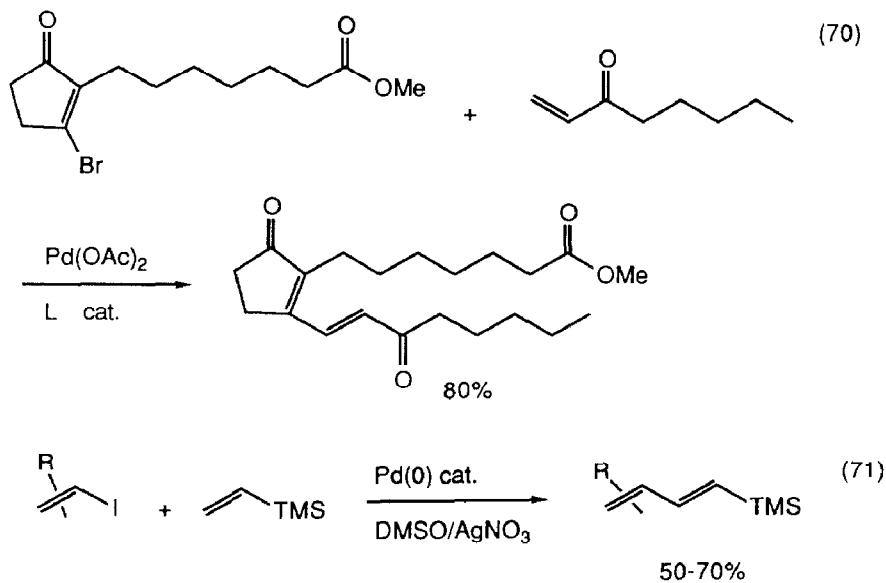
### 3. Alkylation of Olefins

Oxidative addition of aryl and vinyl halides to palladium(0) complexes followed by olefin insertion continues to be the method of choice for the alkylation of olefins. 1-Iodooctane alkylated methyl acrylate in the presence of palladium(II) acetate/amine catalysts (equation 69) [81]. Prostanoids were prepared by alkylation of  $\beta$ -bromocyclopentenones with enones (equation 70) [82]. Trimethylsilyldienes were prepared by palladium catalyzed coupling of vinyl silanes with vinyl iodides (equation 71) [83]. Vinyl lactones were produced by the palladium catalyzed alkylation of  $\omega$ -olefinic acids with vinyl halides (equation 72) [84]. Vinyl tin compound alkylated enol triflates under palladium(0) catalysis (equation 73) [85].

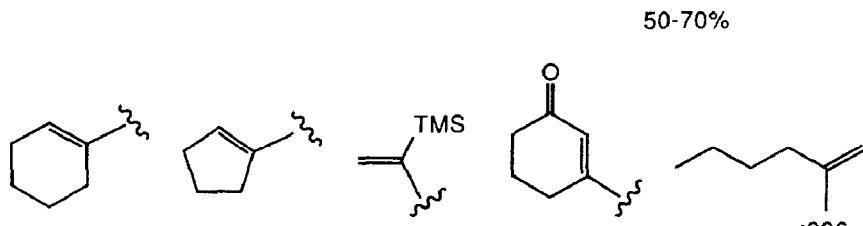
(69)



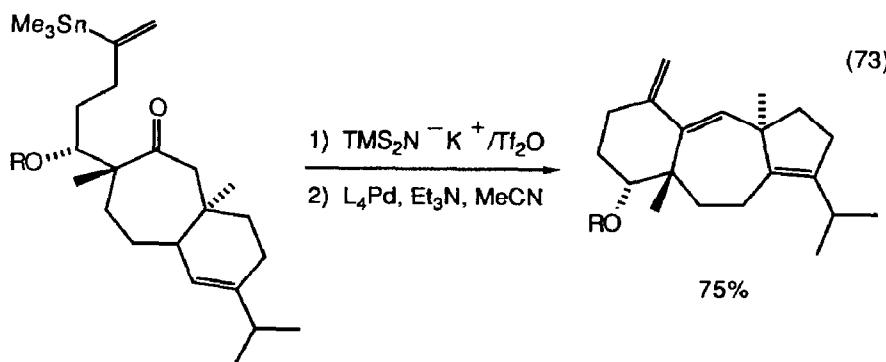
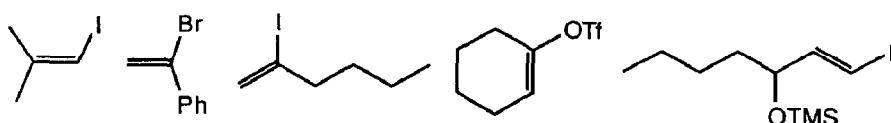
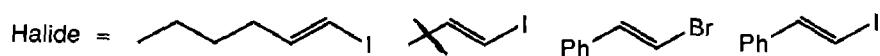
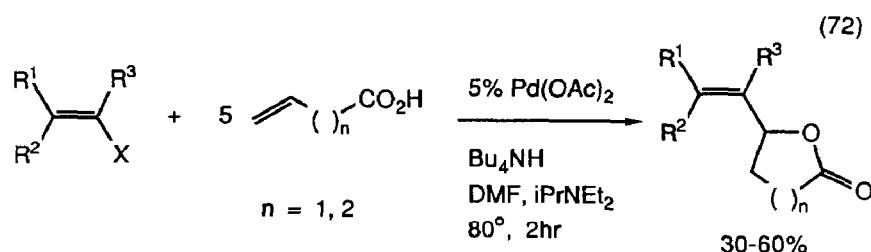
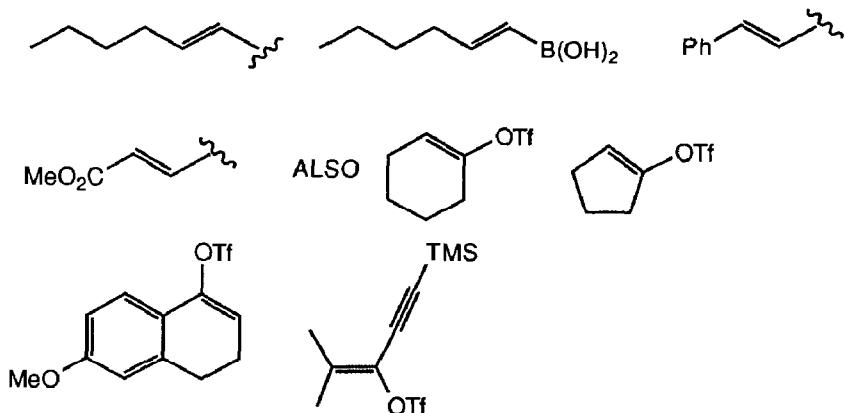
(70)



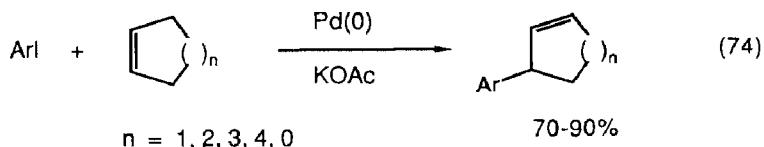
(71)



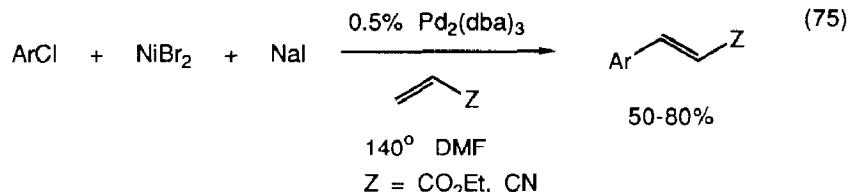
(equation 71 continued)



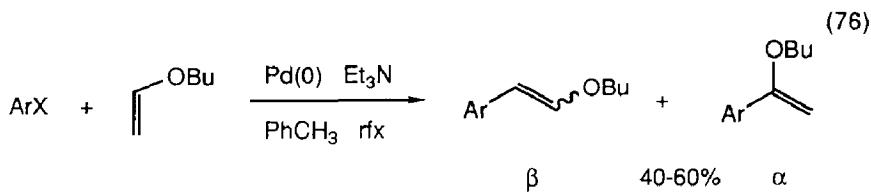
Cycloalkenes were arylated by aryl halides in the presence of palladium(0) catalysts (equation 74) [86]. Aryl chlorides, normally unreactive in these palladium(0) catalyzed olefin arylation reactions, were activated by the addition of nickel(II) bromide and sodium iodide (equation 75) [87]. The regiochemistry of arylation of enol ethers depended strongly on the starting aryl halide (equation 76) [88]. Bromopyridines were alkylated by electron deficient olefins in the presence of palladium(0) catalysts (equation 77) [89], (equation 78) [90]. Bromoquinolines (equation 79) [91] and bromofurans and thiophenes (equation 80) [92] underwent similar reactions.



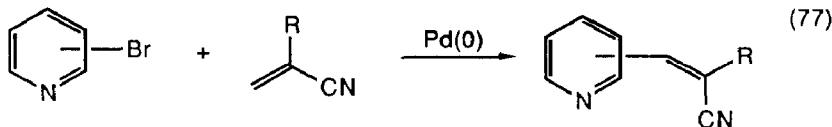
$\text{Ar} = \text{Ph}, \text{oEtO}_2\text{CPh}, \text{pCO}_2\text{EtPh}, \text{mMeOPh}, \text{oMeOPh}$



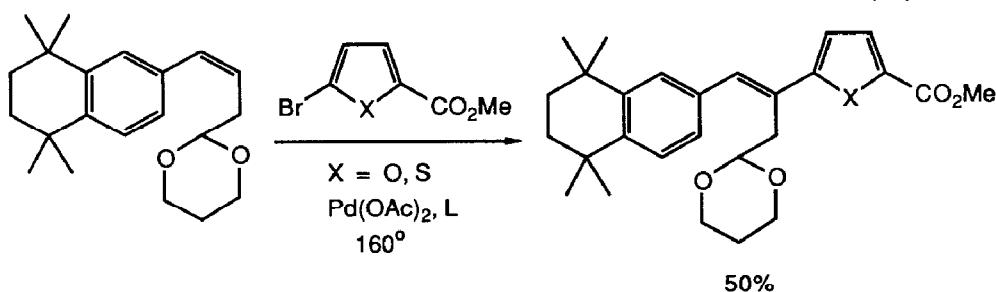
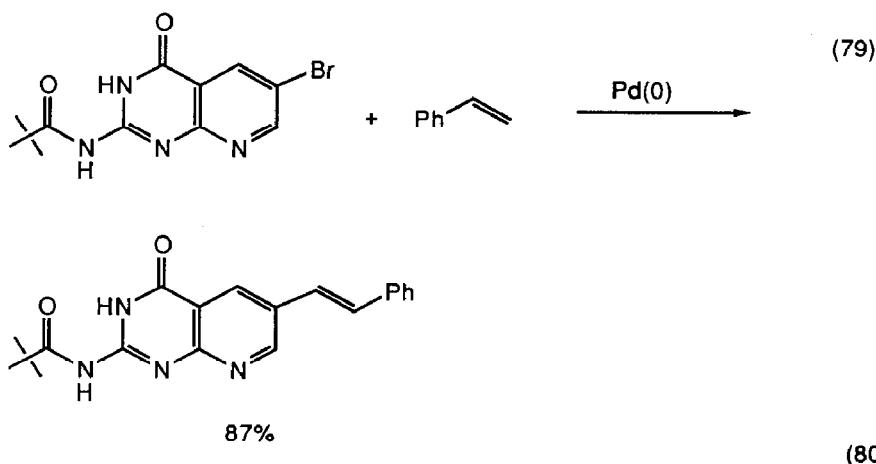
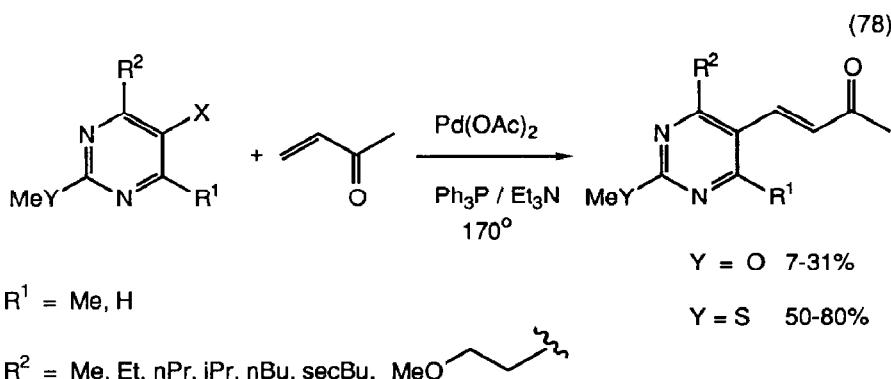
$\text{Ar} = \text{Ph}, \text{pMeO}_2\text{CPh}, \text{pMeOPh}, \text{pMePh}, \text{pNCPH} \parallel \text{pNO}_2\text{Ph}$  failed.



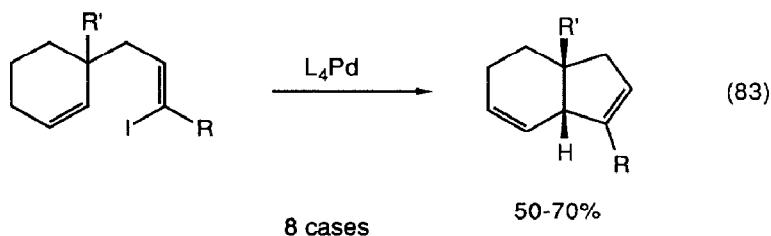
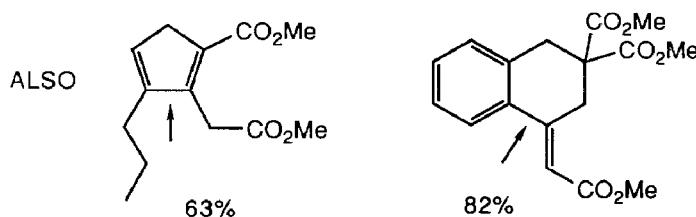
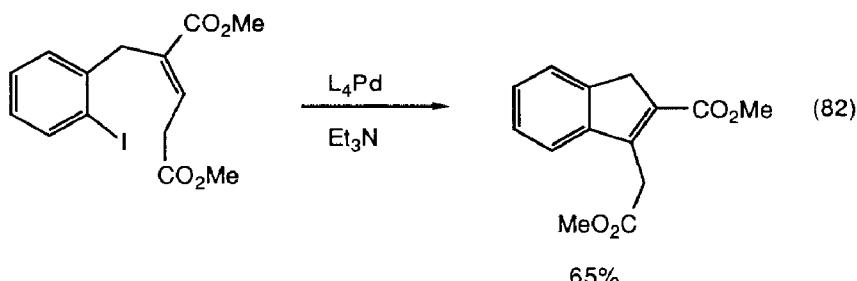
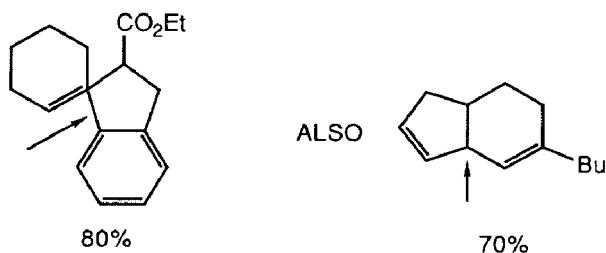
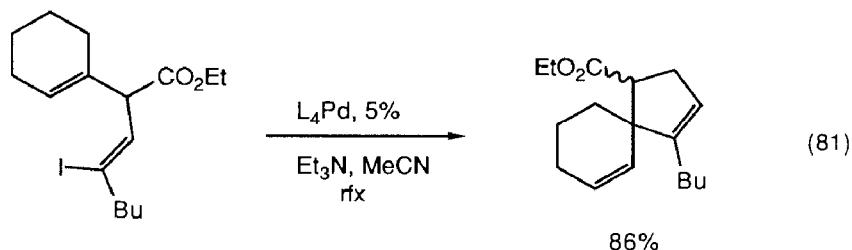
for  $\text{pMeOPhI} \rightarrow 100\% \alpha$   
for  $\text{pNO}_2\text{PhCOCl} \quad 10:1 \beta/\alpha$

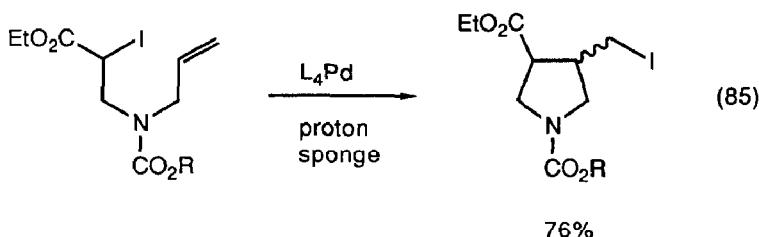
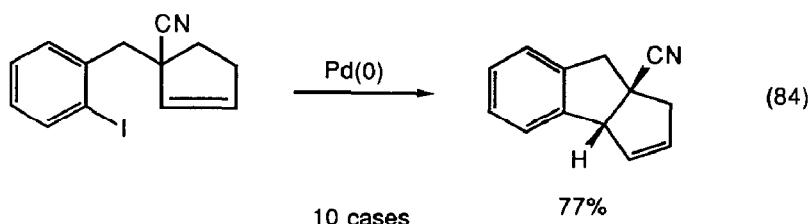


$\text{R} = \text{PhSO}_2, \text{P(O)(OEt)}_2$



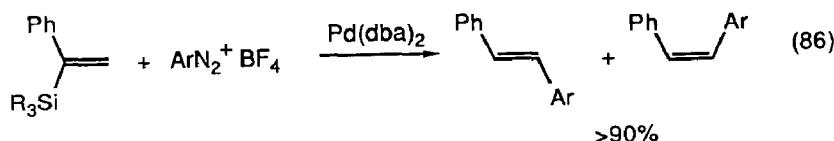
Many intramolecular versions of the palladium catalyzed olefin alkylation process have been reported. These are detailed in equation 81 [93], equation 82 [94], equation 83 [95], equation 84 [96], and equation 85 [97]. It is likely that even more variations on this same theme will be forthcoming.



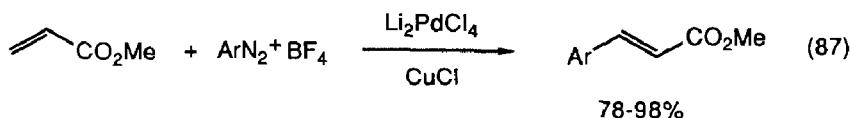


many cases to make 5- and 6-membered rings

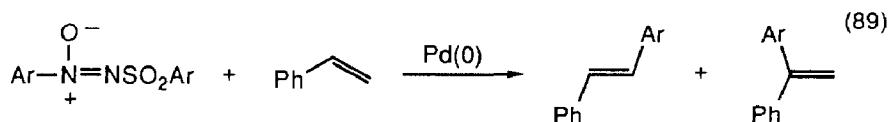
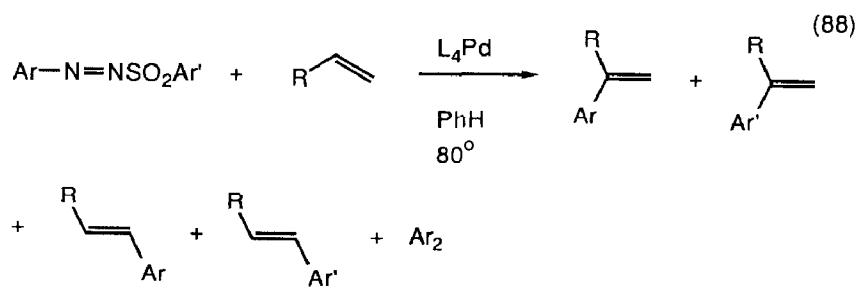
Arenediazonium salts (equation 86) [98], (equation 87) [99] and azosulfones (equation 88) [100], (equation 89) [101] can also serve as sources of aryl groups for the arylation of olefins, although the last two cases are inefficient.



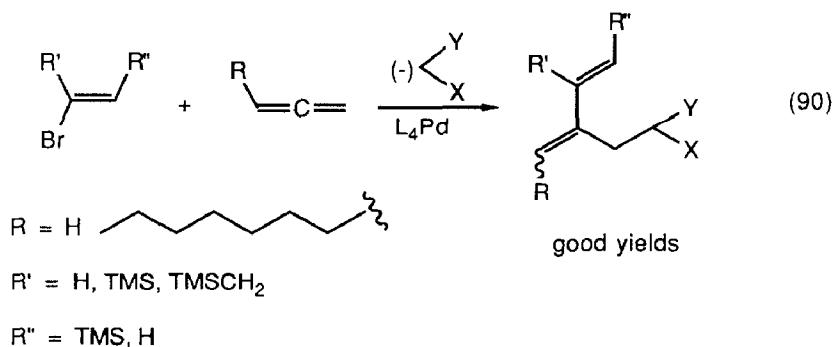
$\text{Ar} = \text{Ph}, \text{pMePh}, \text{pBrPh}, \text{pNO}_2\text{Ph}$

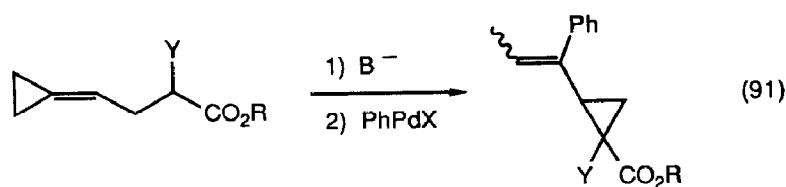


$\text{Ar} = \text{Ph}, \text{pMePh}, \text{pMeOPh}, \text{oClIPh}, \text{mClIPh}, \text{pClIPh}, \text{oNO}_2\text{Ph}, \text{mNO}_2\text{Ph}, \text{oCOOHPh}, \text{mCOOHPh}, \text{pCOOHPh}$



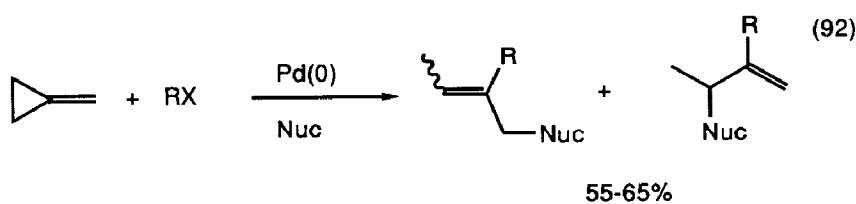
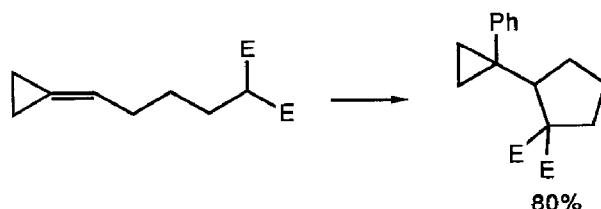
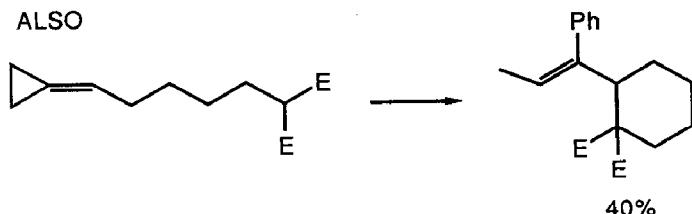
More complex processes which involve oxidative addition/insertion into an allene to give a  $\pi$ -allyl complex and nucleophilic attack on that  $\pi$ -allyl complex to form another carbon-carbon bond have also been developed (equation 90) [102], (equation 91) [103], (equation 92) [104]. The kinetics of the palladium-catalyzed arylation of allylic alcohols (equation 93) have been studied [105]. It was found that triethylamine was a ligand for palladium(0) and a 20 fold excess was best, that the alcohol reduced the palladium(II) precursor, and, with a  $\rho$  of -1.6, oxidative addition was the slow step.





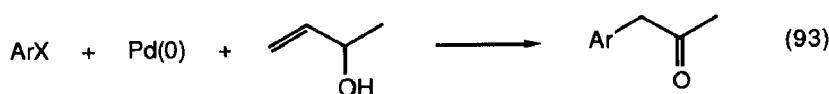
$\text{Y} = \text{CO}_2\text{R}, \text{SO}_2\text{Ph}$

ALSO

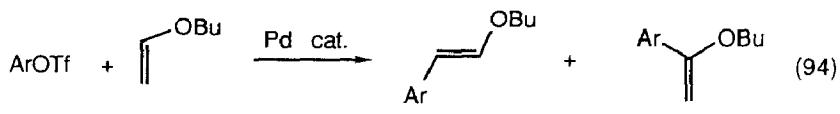


$\text{RX} = \begin{array}{l} \text{---Br} \\ | \\ \text{---PhI} \end{array}$

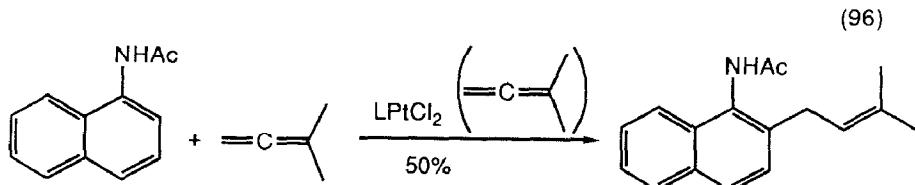
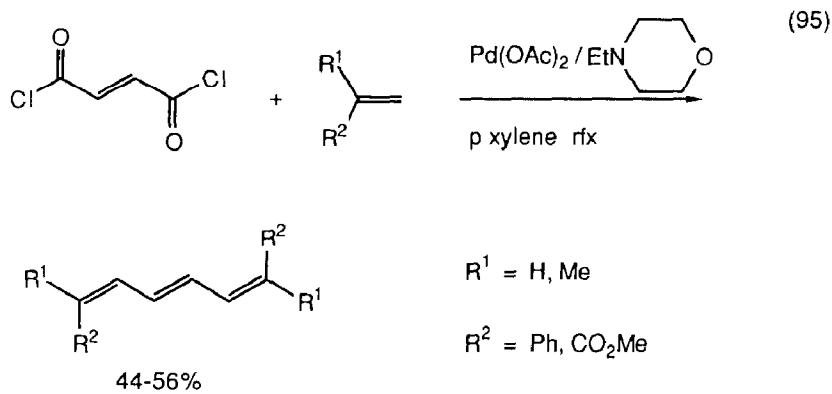
$\text{Nuc} = \begin{array}{l} \text{---} \begin{array}{l} \text{---E} \\ | \\ \text{---E} \end{array} \\ \text{---} \begin{array}{l} \text{---SO}_2\text{Ph} \\ | \\ \text{---E} \end{array} \end{array}$

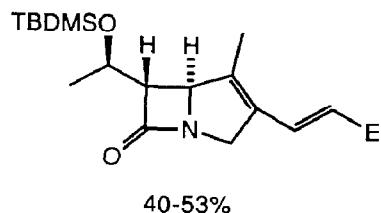
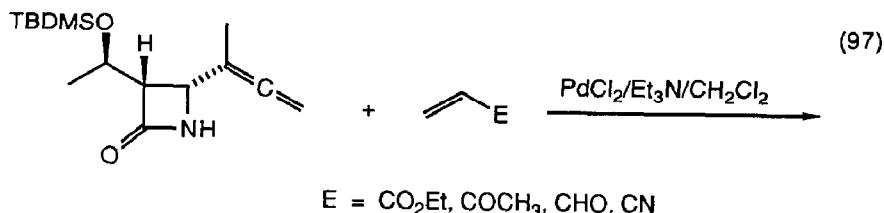


In the palladium(0) catalyzed arylation of enol ethers by aryl triflates, lithium halides had a profound influence on the regiochemistry of the reactions (equation 94) [106]. Trienes were prepared by a palladium(0) catalyzed decarbonylative coupling of olefins with alkenes (equation 95) [107]. Platinum allene complexes effected the direct isoprenylation of arenes by 1,1-dimethylallene (equation 96) [108]. A carbapenem was synthesized through a process involving a palladium catalyzed reaction between an allene and an olefin (equation 97) [109].

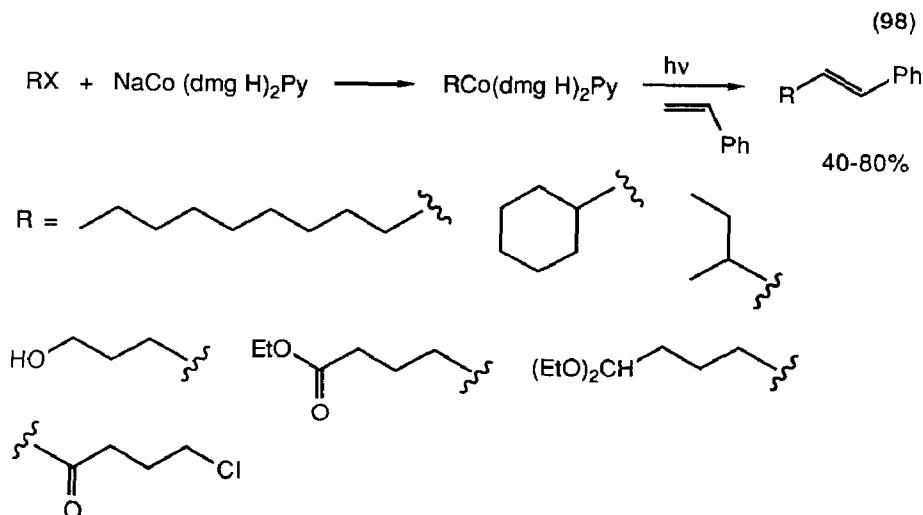


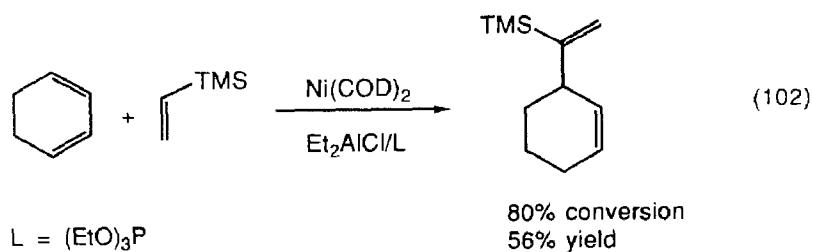
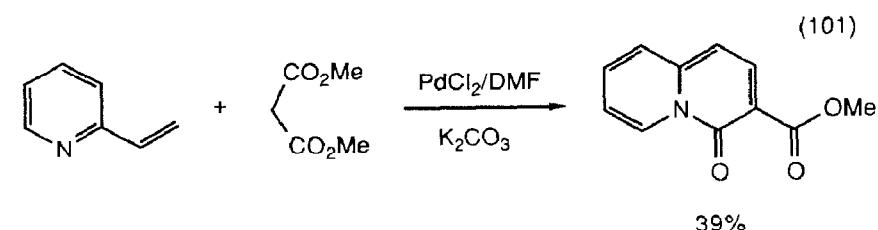
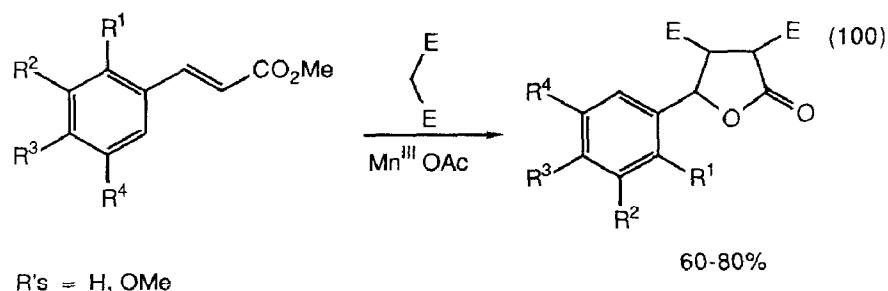
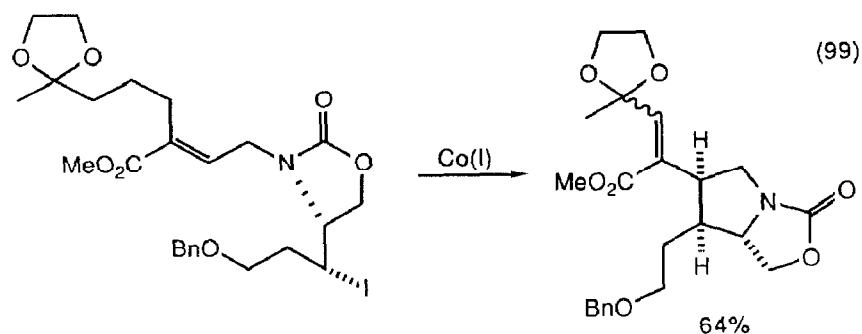
No LiX	1.5 / 1
LiX	13 / 1



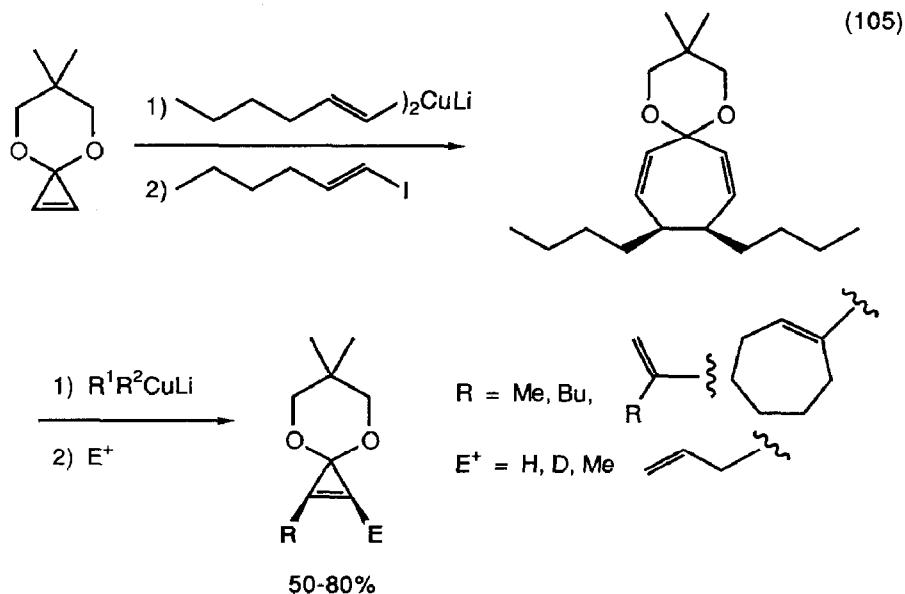
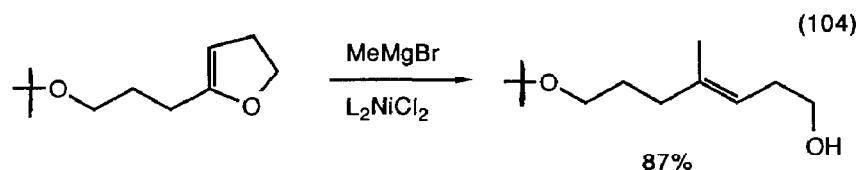
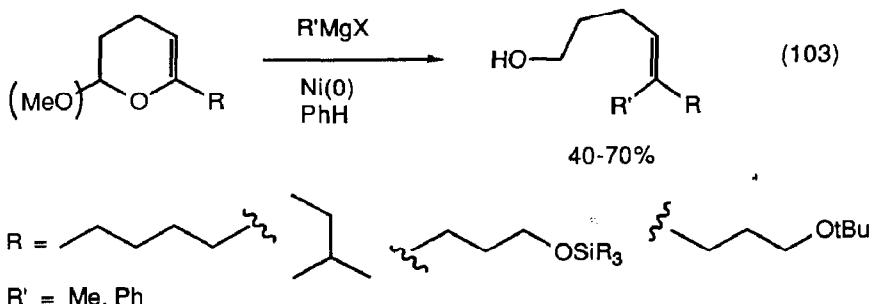


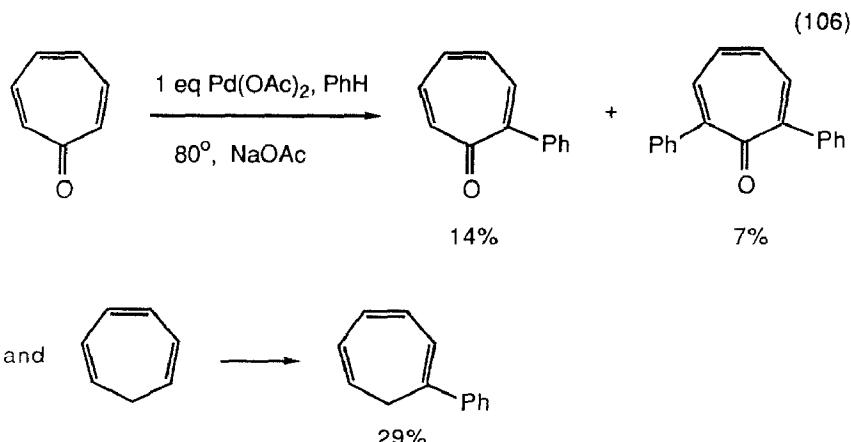
Cobalt(I) complexes promoted free radical alkylations of olefins (equation 98) [110], (equation 99) [111]. Manganese(III) (equation 100) [112] and palladium(II) (equation 101) [113] promoted addition/cyclization reactions of malonate esters to olefins. Nickel(0) complexes catalyzed the  $\alpha$ -alkylation of vinyl trimethylsilane with cyclohexadiene (equation 102) [114].



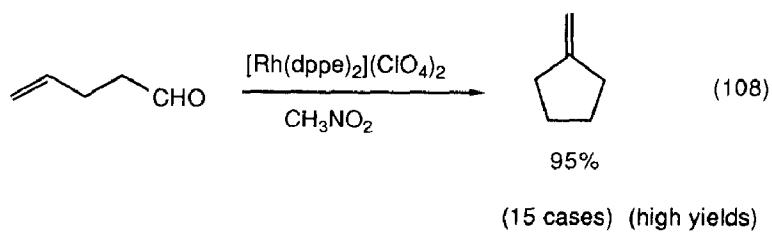
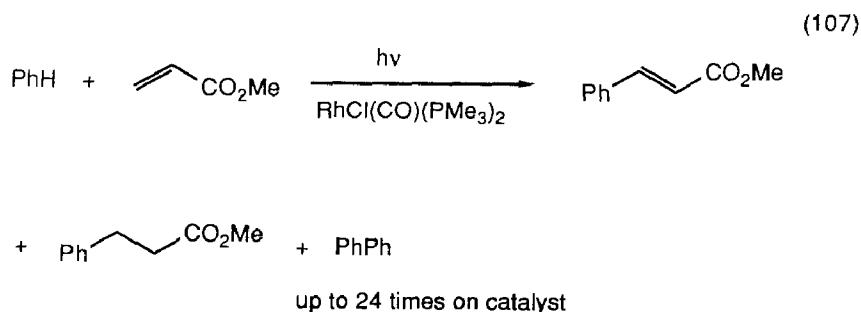


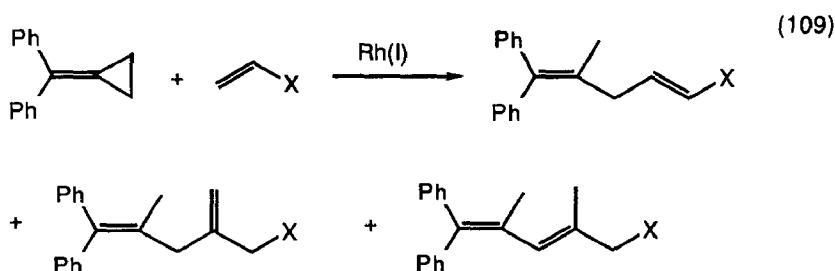
Nickel(II) salts catalyzed the ring opening of dihydropyrans and furans by Grignard reagents (equation 103) [115], (equation 104) [116]. Cyclopropenone ketal underwented direct alkylation by organocupper reagents (equation 105) [117]. Tropanes and tropanones were directly arylated by benzene in the presence of palladium acetate, although the yields were low (equation 106) [118].





Rhodium(I) complexes catalyzed the arylation of acrylate by benzene under photolytic conditions (equation 107) [119]. Rhodium(I) complexes catalyzed the intramolecular addition of aldehydes to olefins (equation 108) [120] and the reactions of vinylcyclopropanes with olefins (equation 109) [121].

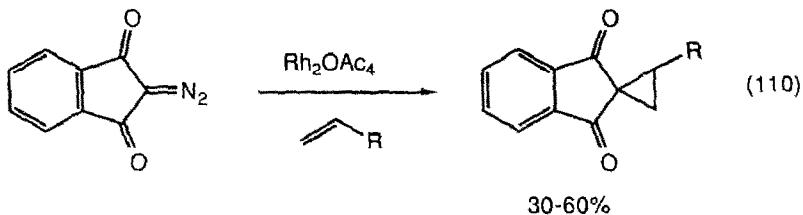




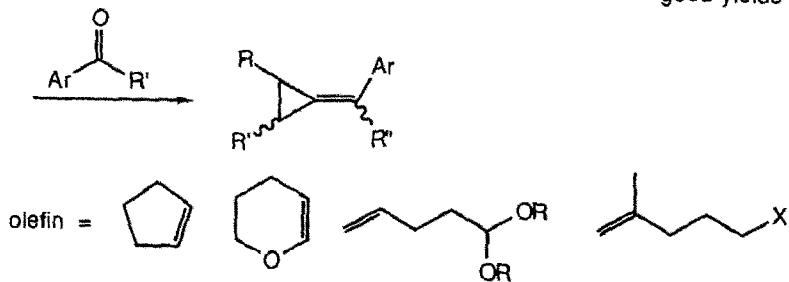
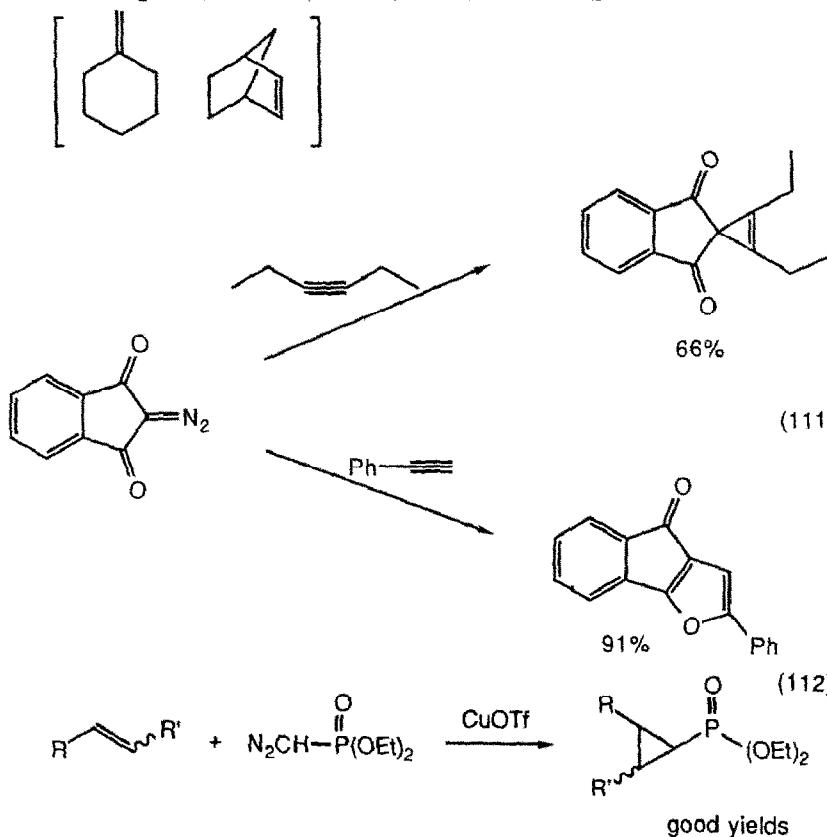
#### 4. Decomposition of Diazoalkanes and Other Cyclopropanations

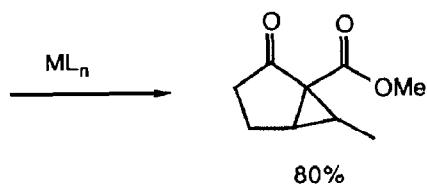
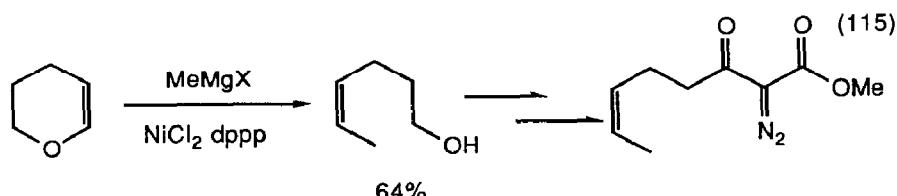
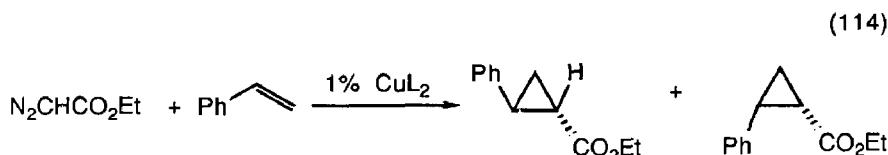
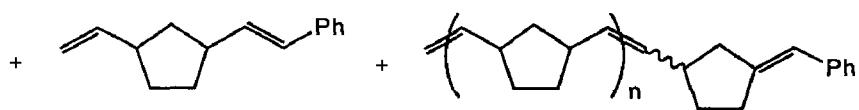
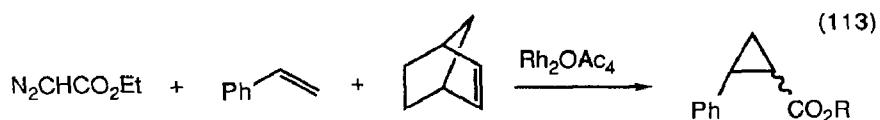
Unsaturated fatty acid methyl esters were cyclopropanated by diazomethane in the presence of palladium acetate [122]. Allyl alcohols, acetates, amines and acetals were cyclopropanated under similar conditions [123]. Copper(I) or rhodium(II) on Nafion 10 catalyzed the cyclopropanation of cyclohexene, and styrene by ethyl diazoacetate [124]. Palladium(II) catalyzed the cyclopropanation of allenes by diazomethane [125]. Palladium acetate catalyzed the polymethylenation of vinylidene cyclopropanes by diazomethane [126], while norbornadiene gave mono and biscyclopropanated products when treated with diazoethane and a palladium chloride catalyst [127]. The kinetics of the cyclopropanation of styrene by diazoacetic esters in the presence of cobalt phthalocyanine complexes has been studied [128].

Rhodium(II) acetate catalyzed cyclopropanation (equation 110) and insertion reactions (equation 111) of diazodiketones with alkenes and alkynes [129]. Copper triflate catalyzed the cyclopropanation of olefins by diazophosphonate esters (equation 112) [130]. Rhodium(II) acetate catalyzed both the cyclopropanation and ring opening polymerization of styrene and norbornene (equation 113) [131]. Chiral copper complexes catalyzed the cyclopropanation of styrene with reasonable ee (equation 114) [132]. Rhodium, ruthenium, palladium, and copper complexes catalyzed a number of intramolecular cyclopropanations by decomposition of diazodicarbonyls (equation 115) [133]. Alkynes were converted to mixtures of cyclopropenes and furans when diazoesters were decomposed by rhodium(II) acetate in their presence (equation 116) [134].

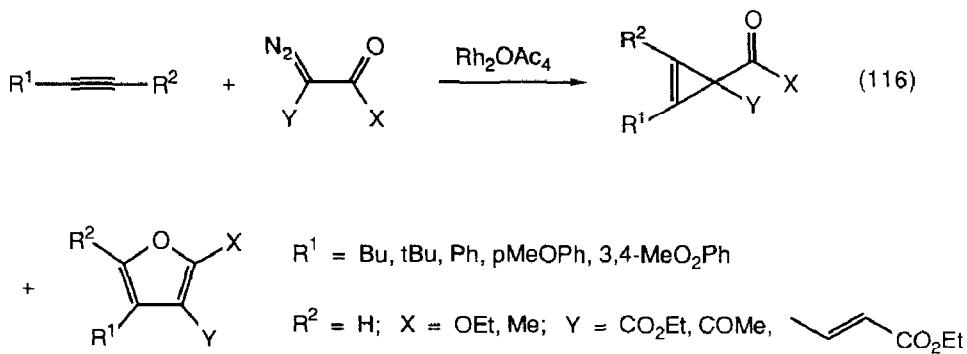


$R = \text{PhCH}_2, \text{Ph}, \text{pMeOPh}, \text{pMePh}, \text{pClPh}, \text{pBrPh}, \text{pNO}_2\text{Ph}$

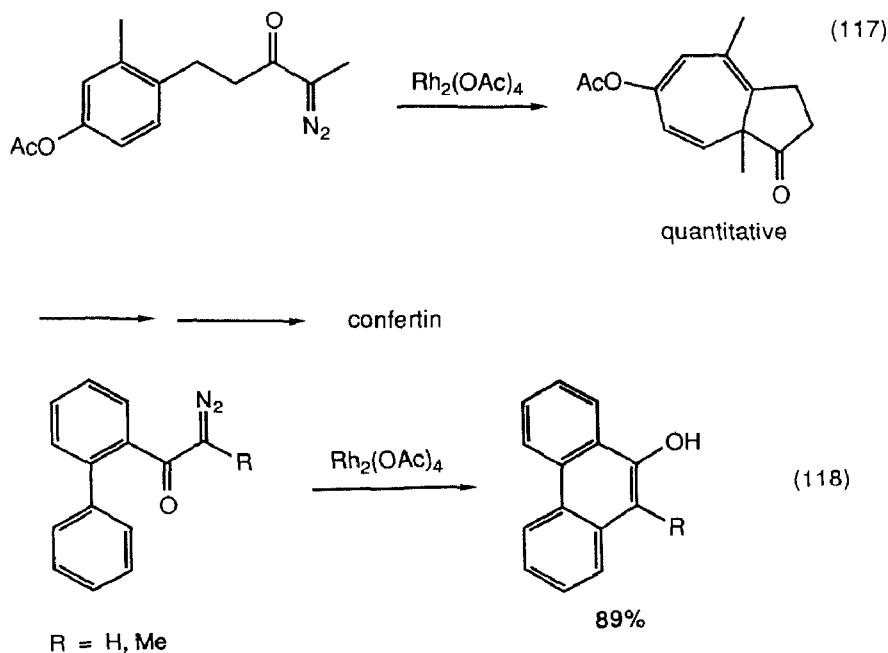




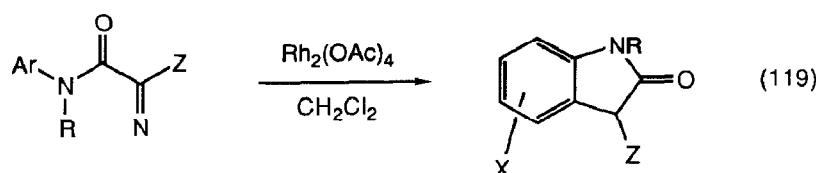
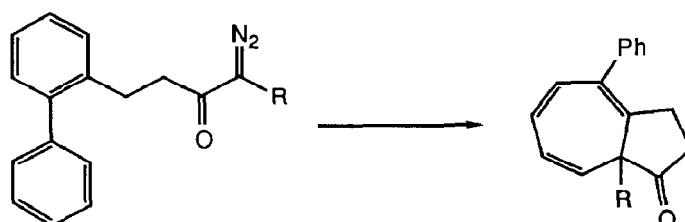
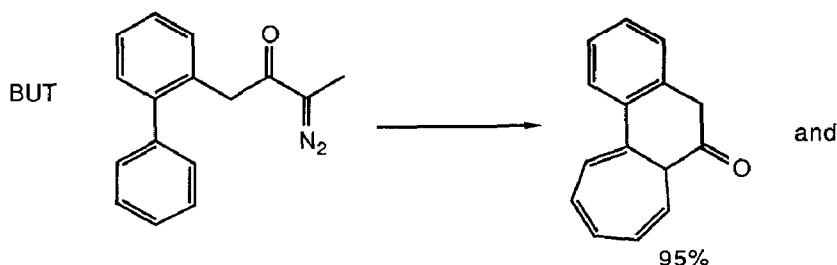
$\text{ML}_n = \text{TPPRhCl}; \text{Ru}_3(\text{CO})_{12}; \text{dpppPdCl}_2; \text{Cu}(\text{TBS})_2$



Transition metals also catalyze a variety of intramolecular C-H insertions, many of which proceed with rearrangement (equation 117) [135], (equation 118) [136]. Heterocycles have been made using metal-catalyzed diazoalkane C-H insertion processes (equation 119) [137], (equation 120) [138], (equation 121) [139]. Enol ethers underwent facile C-H insertion (equation 122) [140], and cyclopentanones were prepared by rhodium(II) catalyzed intramolecular C-H insertion (equation 123) [141].

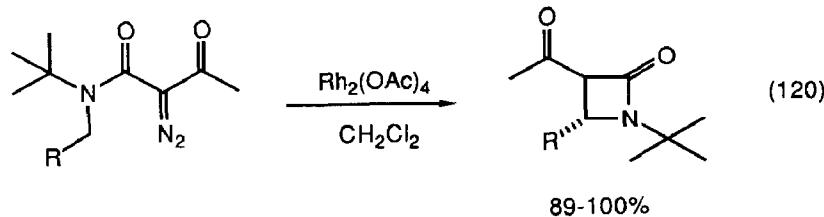


(equation 118 continued)

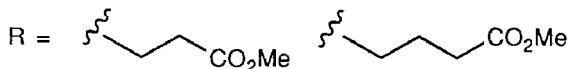
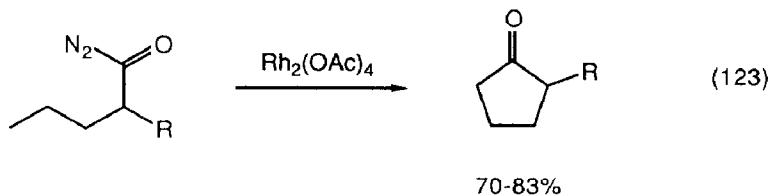
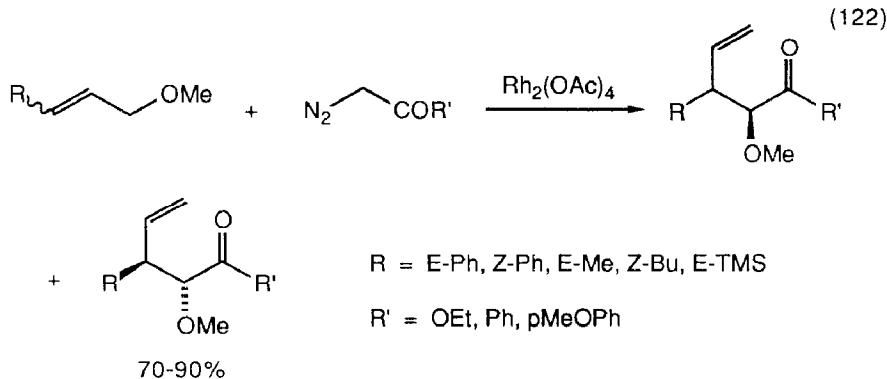
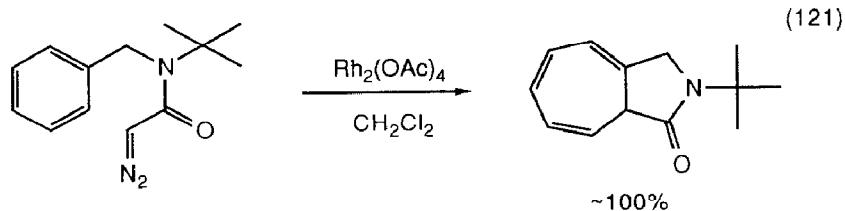


Z = H,  $\text{CH}_3\text{CO}-$ ; R = Me, Et, Bn

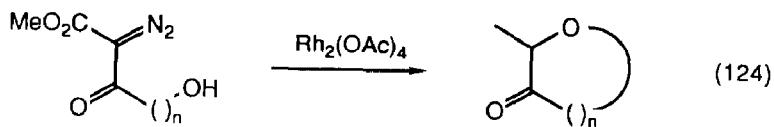
Ar = Ph, oMePh, mMePh,  $\alpha$ -Naphth, 2,3-Me<sub>2</sub>Ph, 3,4-



R = Me<sub>2</sub>,

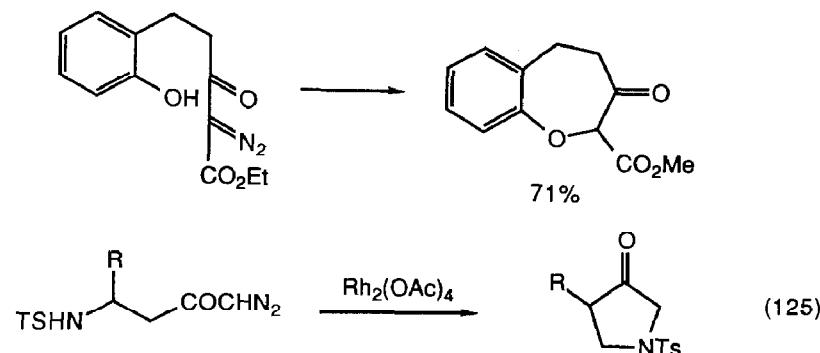
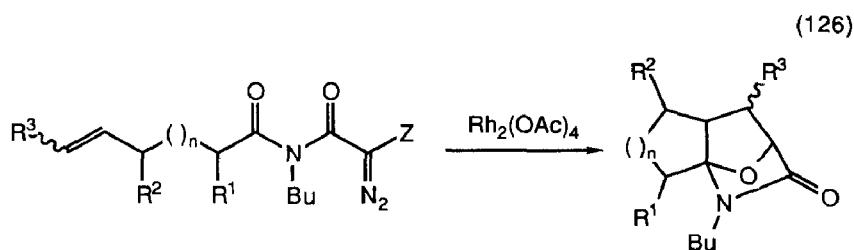


Heterocycles have also been prepared by the rhodium(II) acetate catalyzed insertion of carbenes into OH (equation 124) [142] and NH (equation 125) [143] bonds. Carbenes produced by metal-catalyzed diazo decompositions can also be trapped by carbonyl groups (equation 126) [144], alkenes and alkynes (equation 127) [145], (equations 128 and 129) [146], dienes (equation 130) [147], and vinyllogous amides (equation 131) [148].

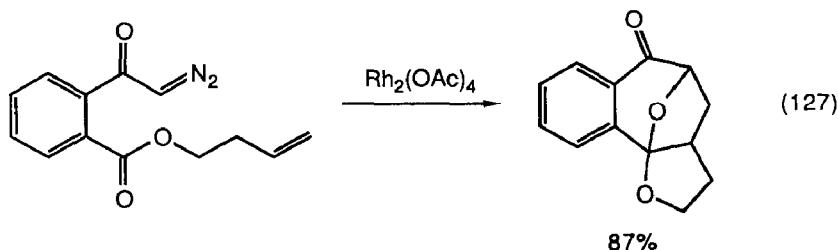


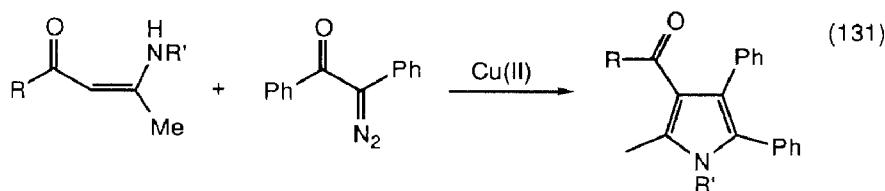
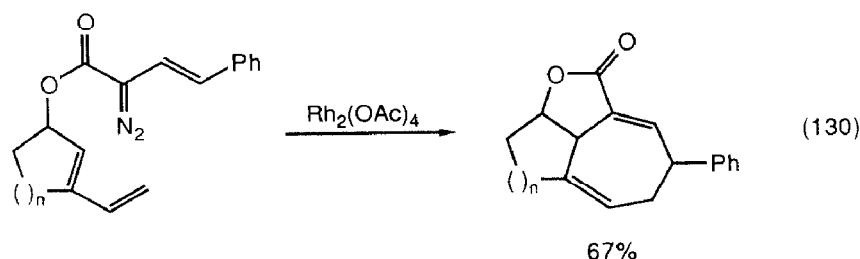
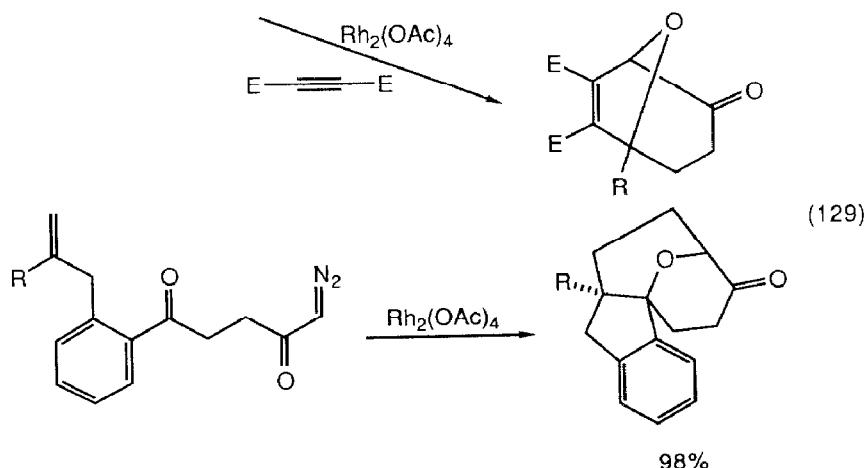
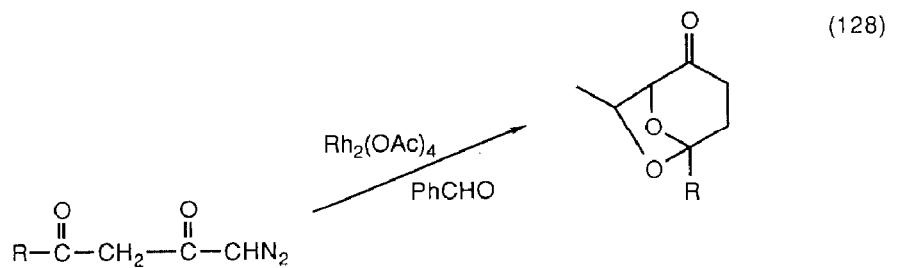
$n = 2$	0%
3	78%
4	24%
5	0%

AND

 $R = H, Me, Et \quad 100\%$ 

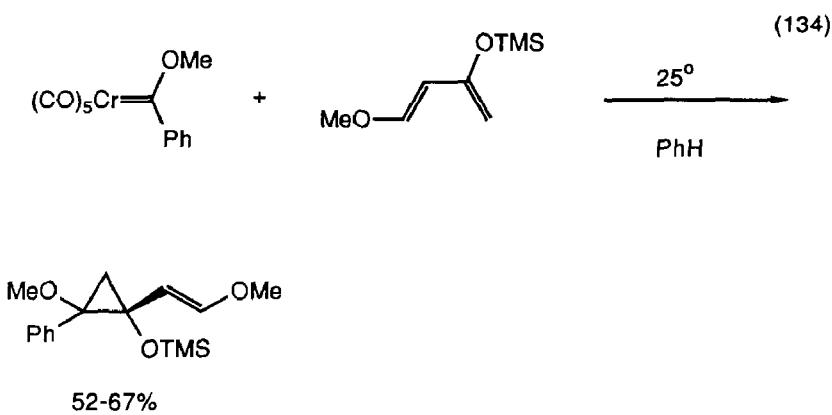
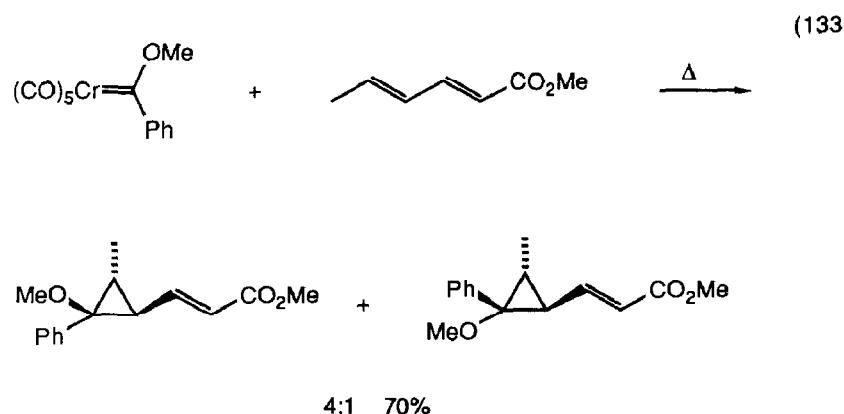
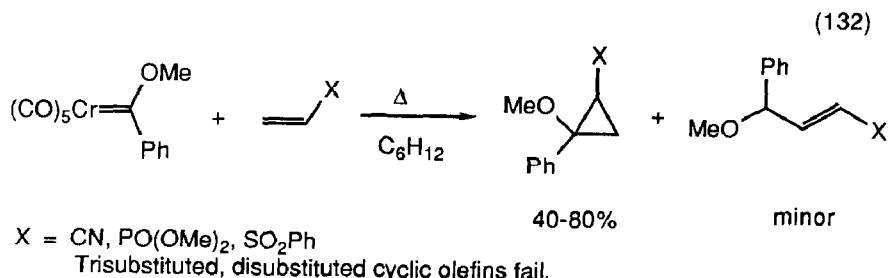
$R^1 = H, Me; R^2 = H; R^3 = H, Me$   
 $n = 1, 2; Z = C=O$

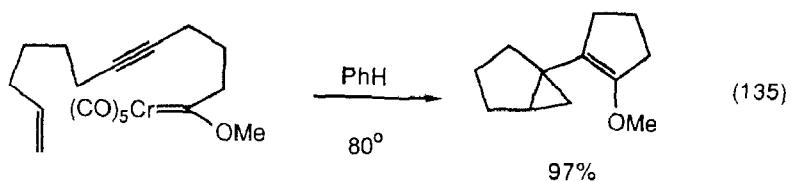




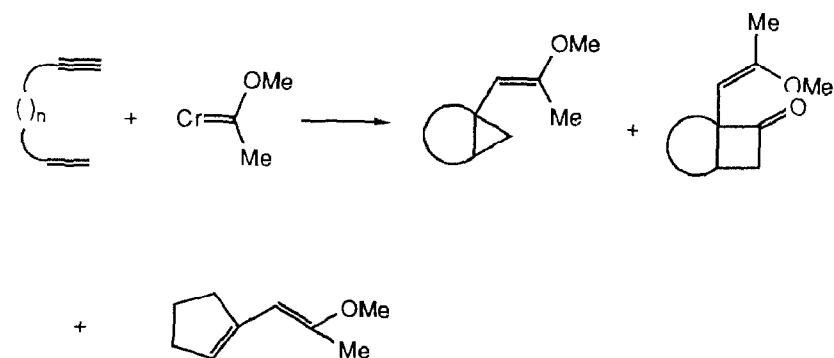
$\text{R}$	$\text{Me}$	$21\%$	$\text{Me}$	$37\%$	$\text{OEt}$	$14\%$	$\text{EtO}$
$\text{R}'$	$\text{H}$		$\text{Me}$	$\text{H}$		$\text{Me}$	$71\%$

Thermal reaction of chromium carbene reagents with electron poor olefins (equation 132) [149], dienoic esters (equation 133) [150], and electron rich dienes (equation 134) [151][152] results in cyclopropanation. Intramolecular cyclopropanation and olefin metathesis reactions of tungsten carbene complexes was the subject of a dissertation [153]. Chromium carbene complexes containing pendant olefinic groups underwent combined metathesis/ cyclopropanation processes (equation 135) [154].

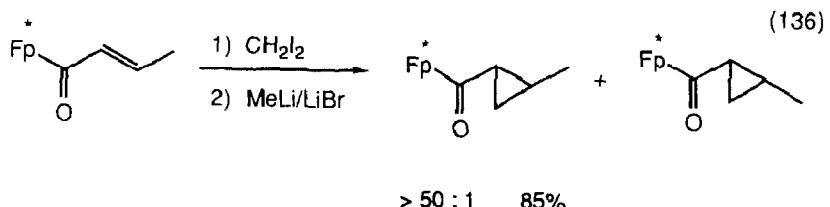




AND

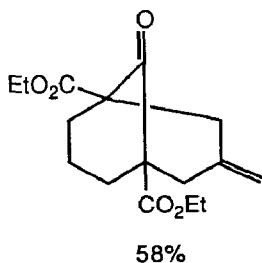
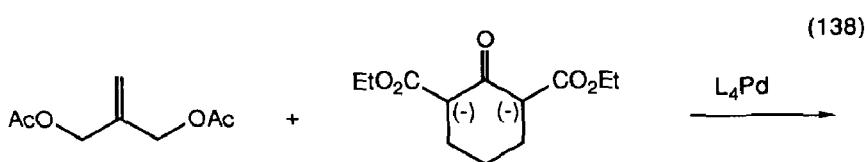
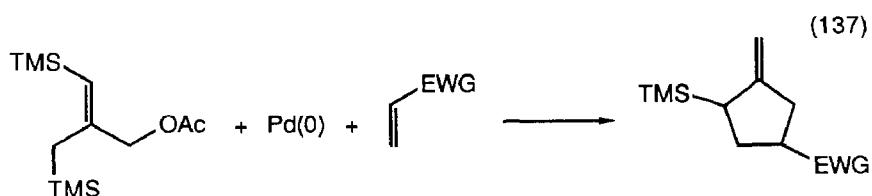


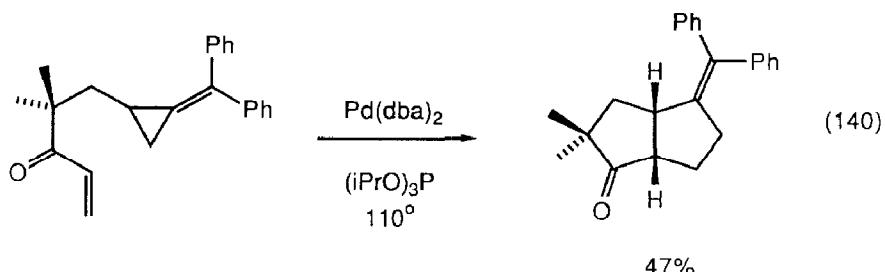
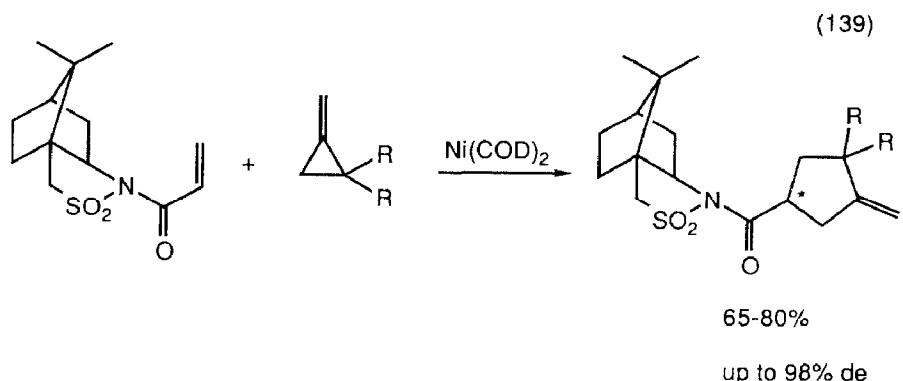
Dimethyl maleate was cyclopropanated by dibromomethane in the presence of zinc metal and cobalt(II) or nickel(II) chloride and ultrasound [155]. Chiral  $\alpha,\beta$ -unsaturated acyliron species were cyclopropanated by diiodomethane with a high degree of stereoselectivity (equation 136) [156]. Asymmetric cyclopropanation has been reviewed (26 references) [157].



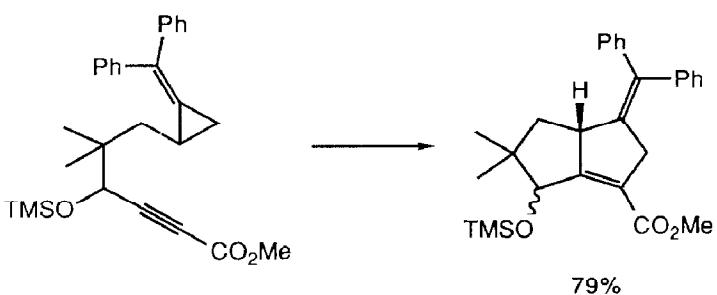
### 5. Cycloaddition Reactions

Several reviews dealing with transition metal catalyzed cycloaddition processes have appeared. They included "intramolecular carbometallation approach to cyclopentanoids (a lecture)" [158]. "Transition metal templates as guides for cycloaddition" (36 references) [159] and "Transition metal mediated cycloaddition reactions of alkynes in organic synthesis" (336 references) [160]. Palladium-complexed trimethylene methane derivatives cycloadded to conjugated enones (equation 137) [161] and stabilized dianions (equation 138) [162]. Vinyl cyclopropanes underwent nickel(0) (equation 139) [163] and palladium(0) (equation 140) [164] catalyzed cycloadditions to  $\alpha,\beta$ -unsaturated enones.

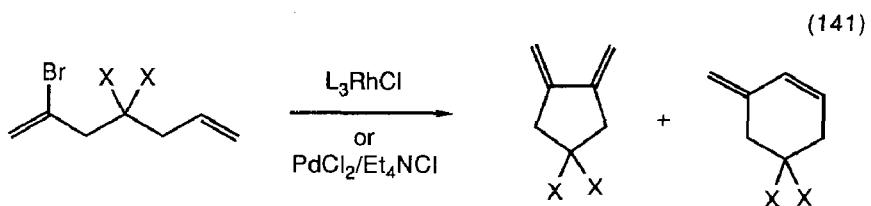




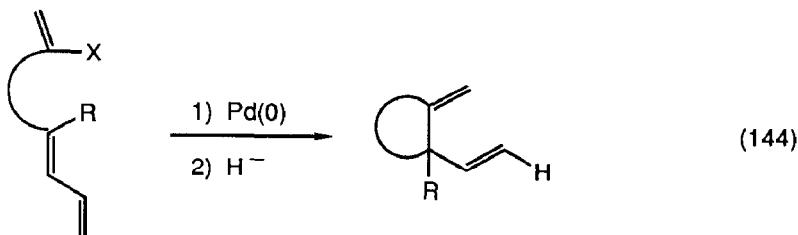
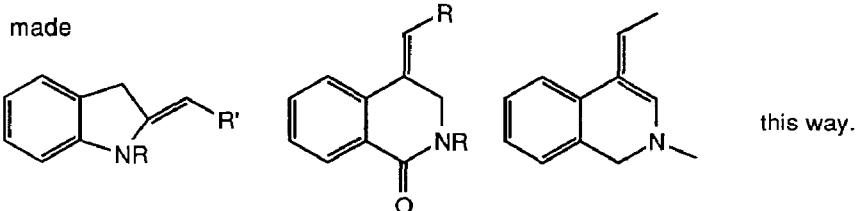
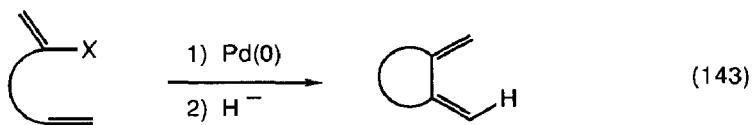
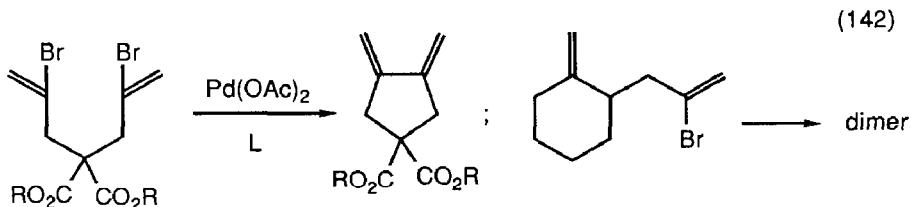
AND

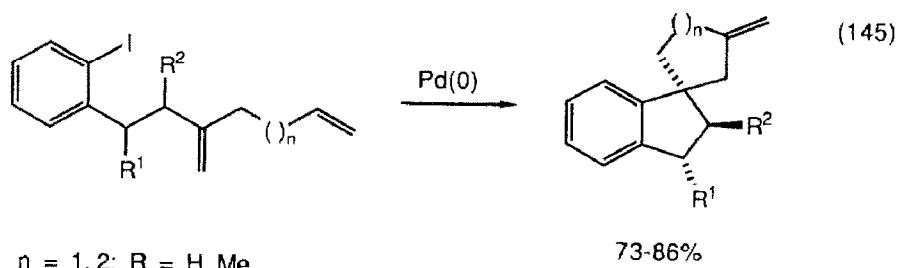


A number of cycloadditions involving oxidative addition/olefin insertion processes with vinyl or aryl halide substrates have been developed. Thus  $\omega$ -olefinic vinyl halides cyclized to exocyclic dienes (equation 141) [165] as did dibromodihalides (equation 142) [166]. Alkynes (equation 143) [167], dienes (equation 144) [168] and diolefins underwent similar cyclization (equation 145) [169], the latter producing two rings.

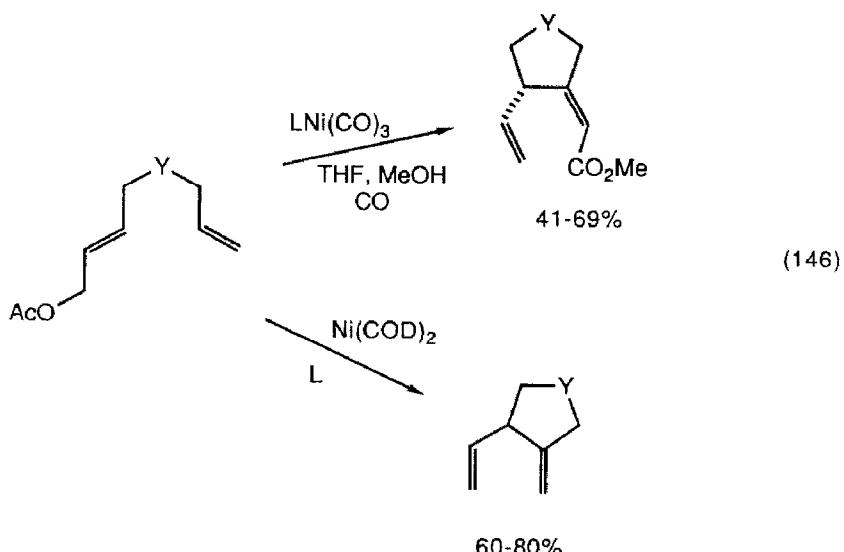


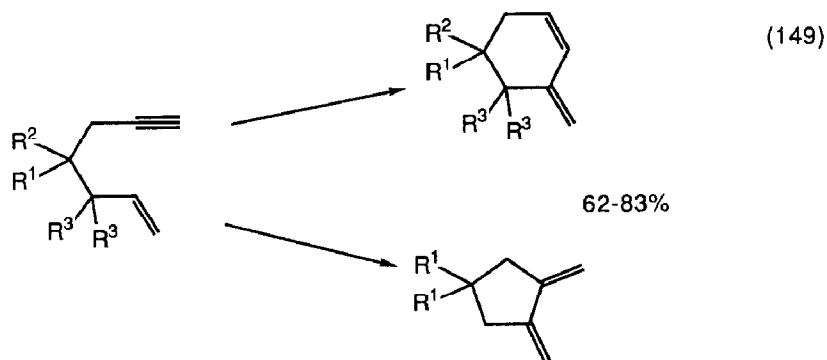
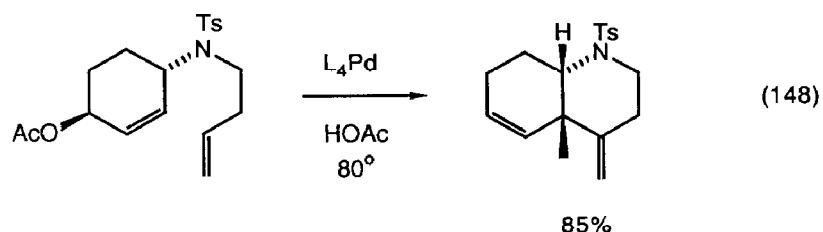
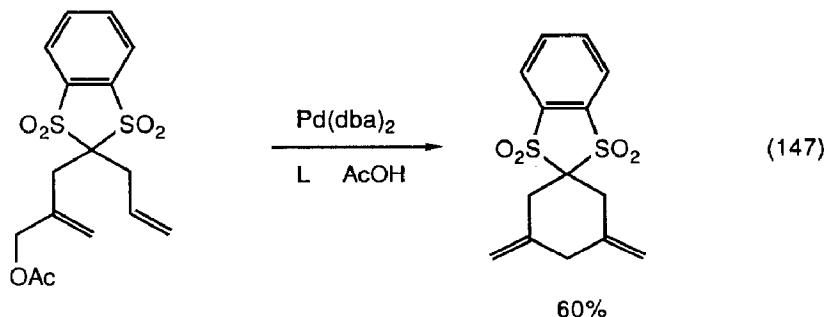
major with Rh  
No selectivity for Pd





Allylacetates cycloadded to olefins under nickel(0) catalysis (equation 146) [170] and palladium(0) catalysis (equation 147) [171], (equation 148) [172]. Enynes were cyclized by rhodium(I) and palladium(II) catalysts (equation 149) [173], (equation 150) [174], and the resulting diene could be trapped with a dienophile (equation 151) [175]. Eneallenes cyclized in the presence of chromium(II)/polymer bound nickel(II) catalysts (equation 152) [176].

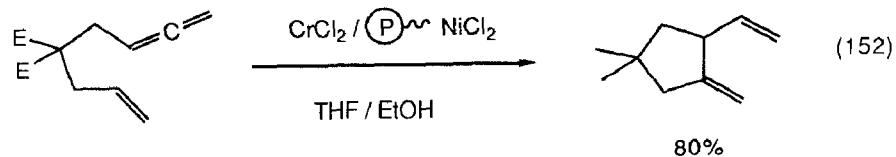
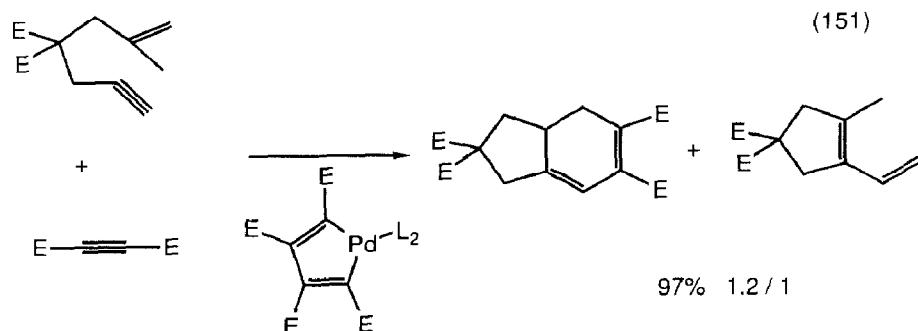
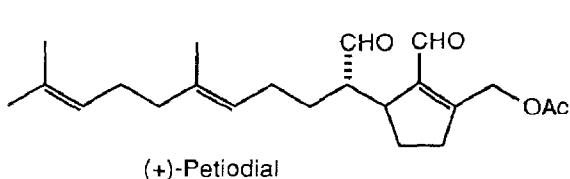
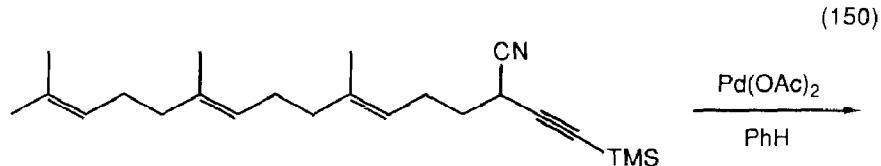




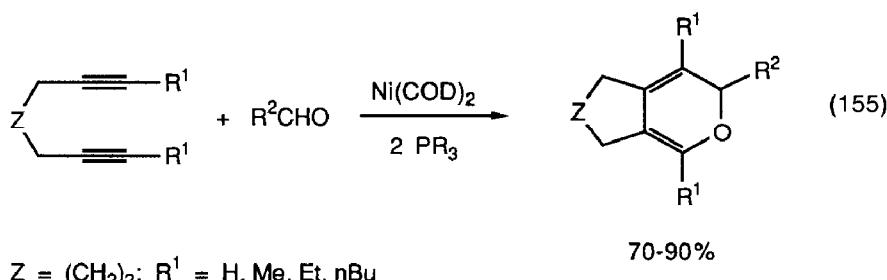
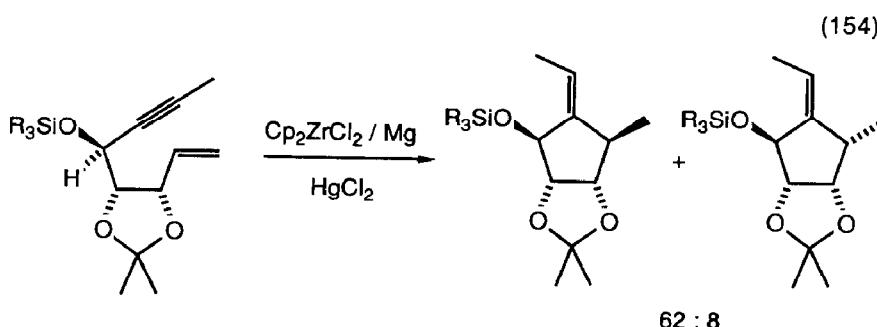
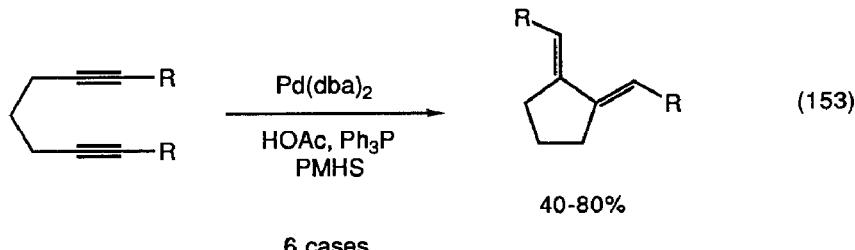
$R^1 = CO_2Et, COMe, H, SO_2Ph$

$R^2 = CO_2Et, COMe, CO_2Me, SO_2Ph$

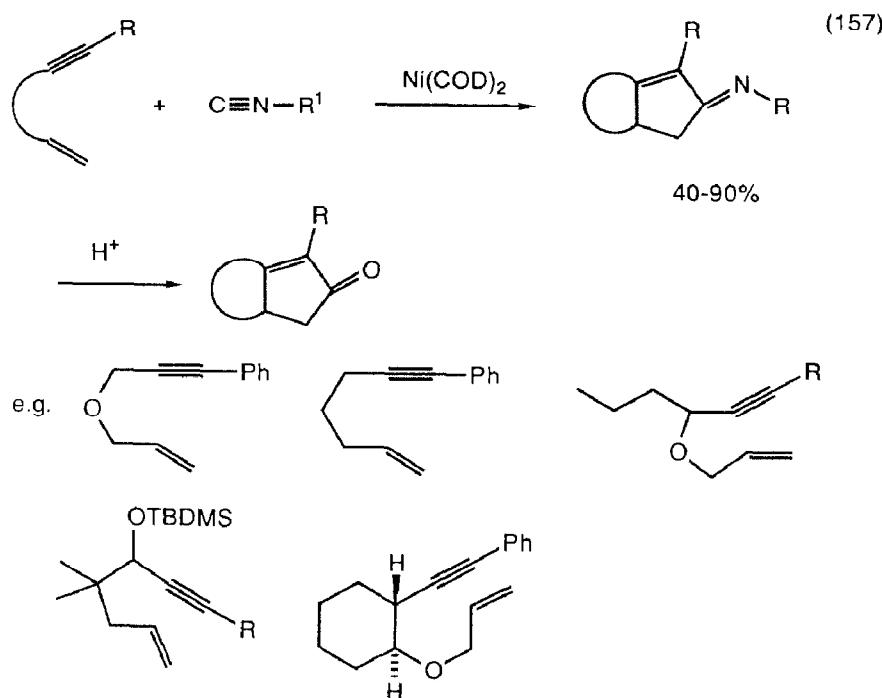
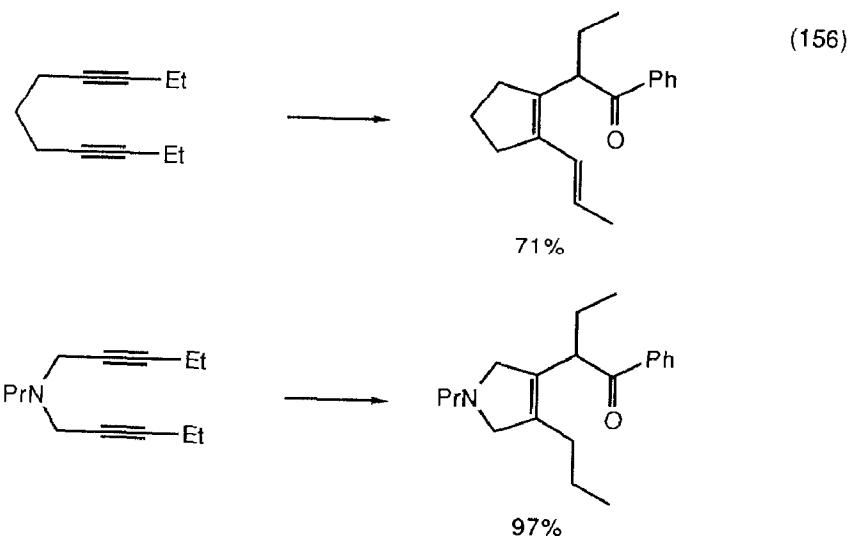
$R^3 = H$



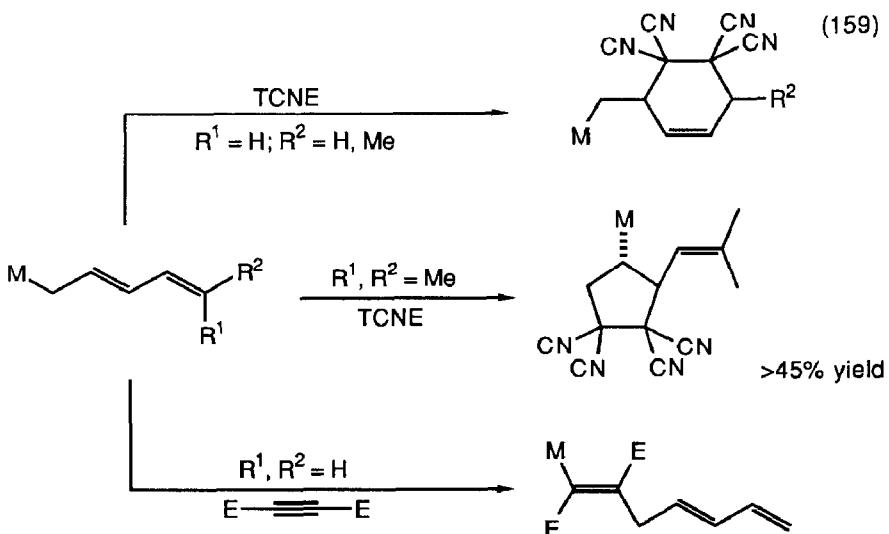
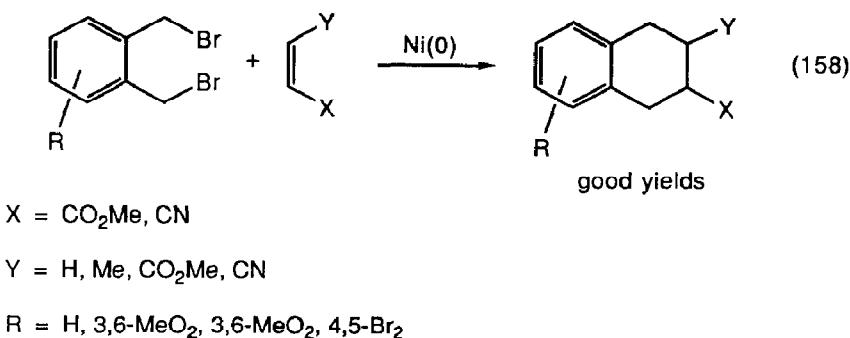
Palladium(0) complexes catalyzed the cyclization of diynes to exocyclic dienes (equation 153) [177]. Reduced zirconocenes cyclized enynes to cyclopentanones (equation 154) [178]. Nickel(0) complexes catalyzed the cocomplexation of diynes with aldehydes (equation 155) (equation 156) [179] and with isonitriles (equation 157) [180].



$Z = (\text{CH}_2)_2$ ;  $R^1 = \text{H, Me, Et, nBu}$   
 $R^2 = \text{Ph, Me, iPr}$

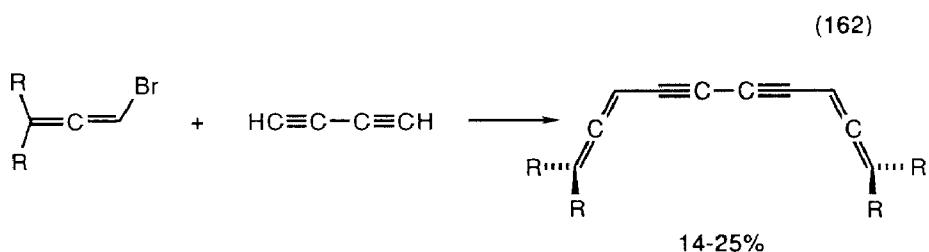
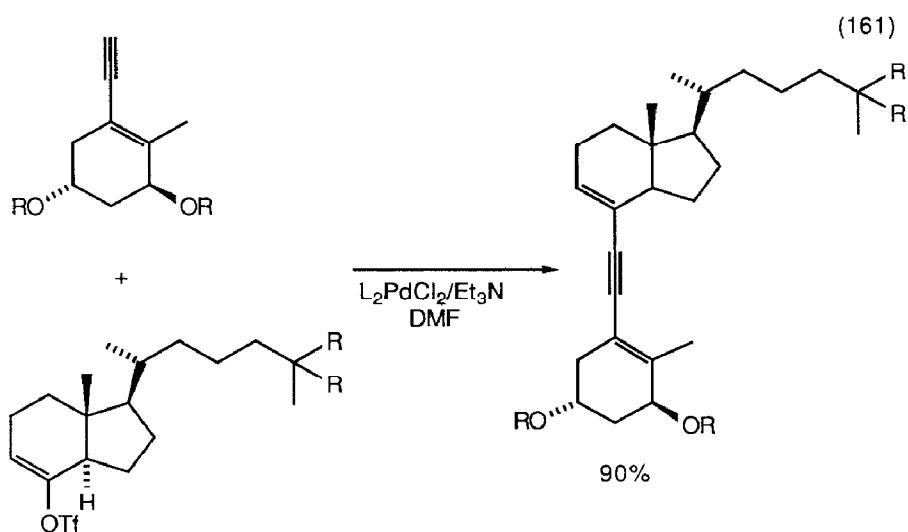
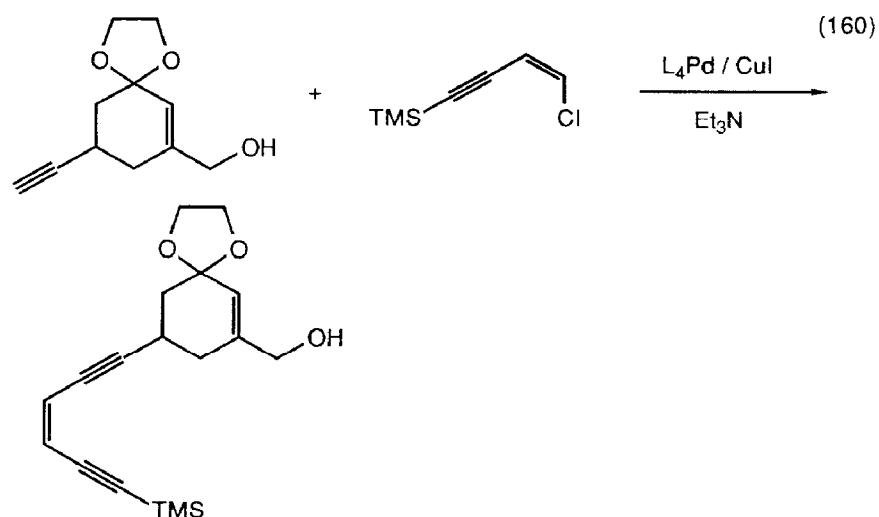


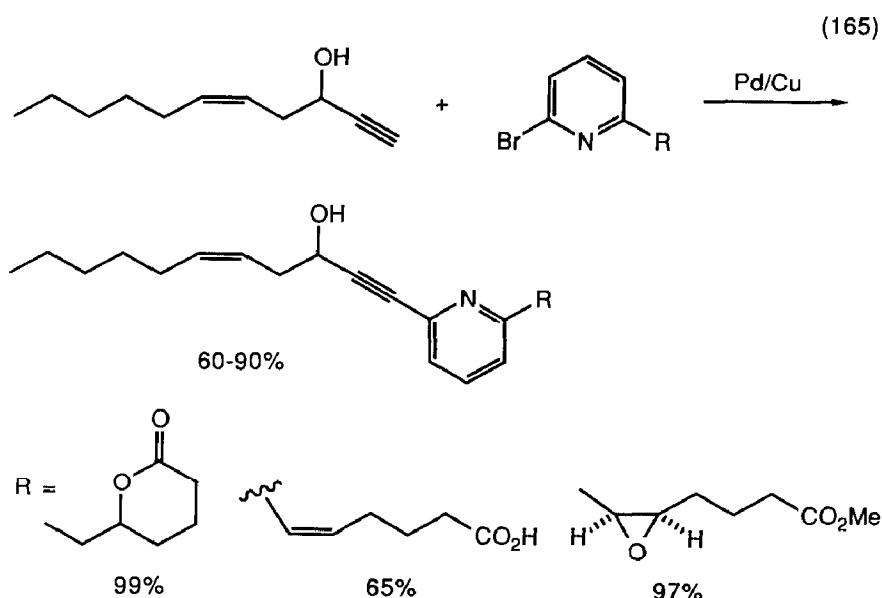
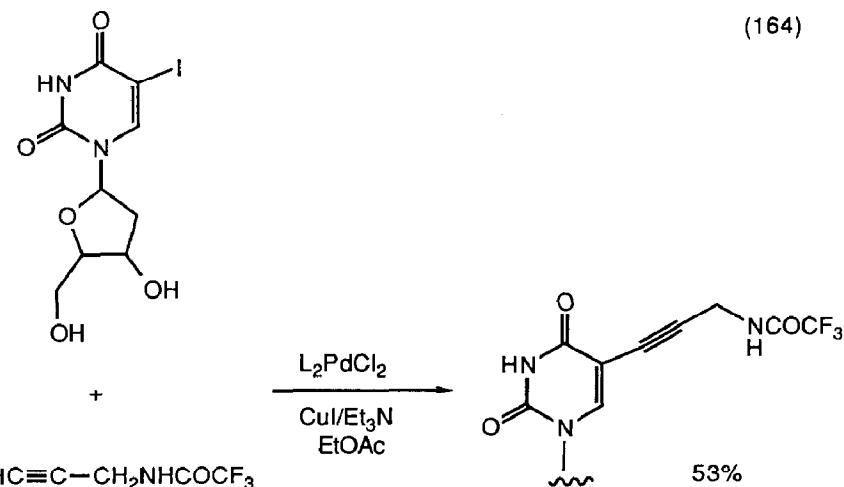
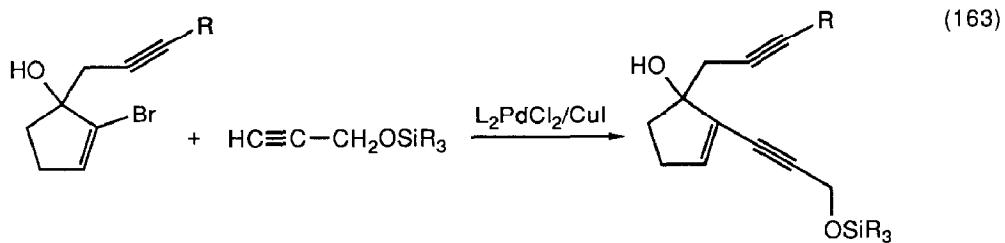
1,2-Dibromomethyl benzene cycloadded to olefins in the presence of nickel(0) complexes (equation 158) [181]. TCNE cycloadded to metal dienyl complexes (equation 159) [182].

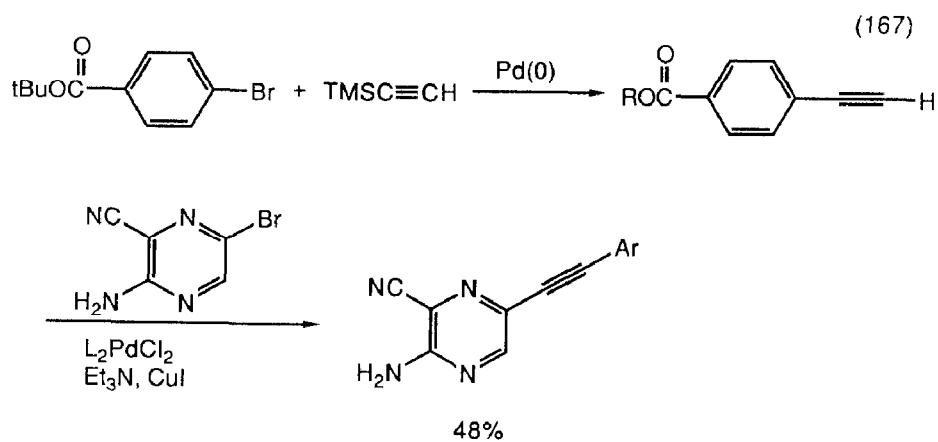
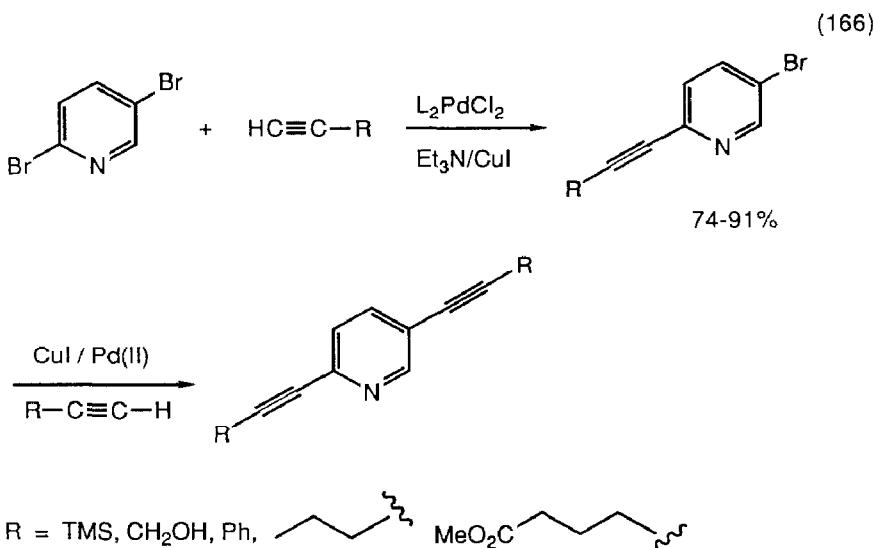


## 6. Alkylation of Alkynes

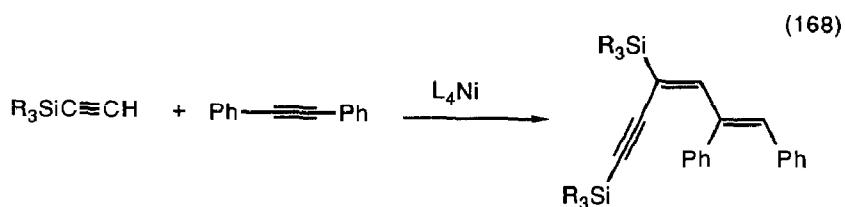
The palladium(0)/copper(I) catalyzed alkylation of alkynes with halides has continued to be developed for the homologation of enynes (equation 160) [183]. Vinyl triflates (equation 161) [184], allenic halides (equation 162) [185], and vinylic halides (equation 163) [186] all underwent the reaction efficiently. Heteroaromatic halides such as pyrimidines (equation 164) [187], bromopyridines (equation 165) [188], (equation 166) [189] and bromopyrazines (equation 167) [190] all were efficiently alkynylated. Nickel, copper, and palladium complexes catalyzed the cross coupling reactions of magnesium diacetylides with organic halides [191].



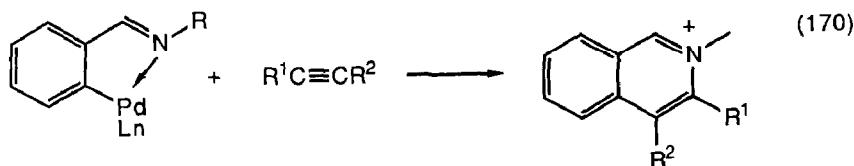
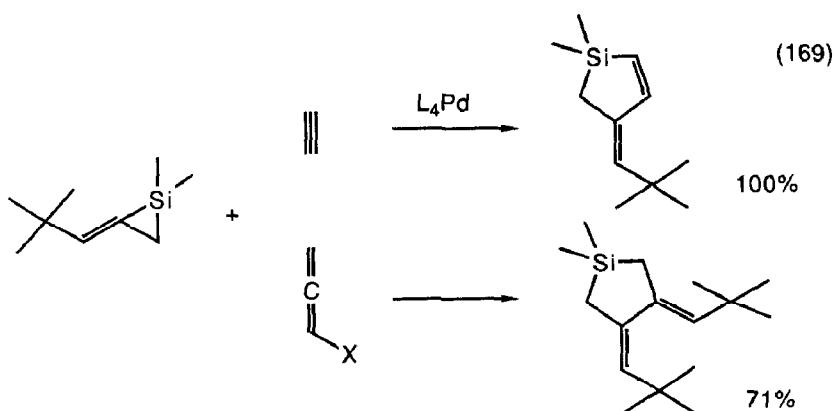
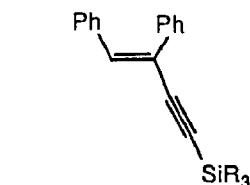




Nickel(0) complexes catalyzed the addition of trimethylsilylacetylene to diphenyl acetylene (equation 168) [191]. Palladium(0) catalyzed the addition of vinyl cyclopropylsilanes to alkynes (equation 169) [192]. Alkynes inserted into *o*-palladated aryl imines (equation 170) [193].

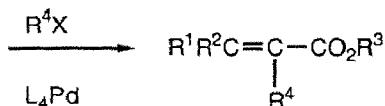
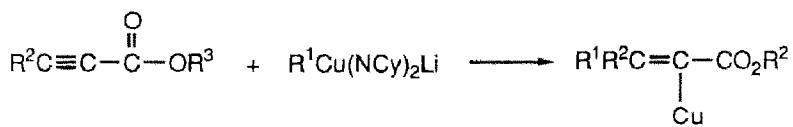


+

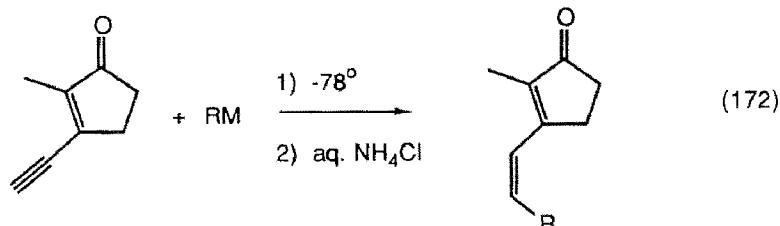
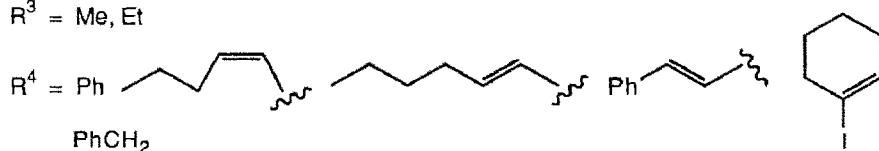
 $R^1 = Ph, pMeOPh, Bz, Me, tBu, pMePh$  $R^2 = Et, MeO_2C, Ph$ *fair yields*

"Carbocupration - a powerful tool for insect sex pheromones synthesis" is the title of a review (70 references) [194]. Acetylenic esters underwent facile carbocupration by organocopper species (equation 171) [195] as did ynenones (equation 172) [196] and simple alkynes (equation 173) [197]. Alkynes were silylated and stannylylated by appropriate organolithium reagents in the presence of manganese(II) chloride (equation 174) [198]. Titanocene dichloride catalyzed the hydroalumination of alkynes by lithium aluminum hydride [199]. Zirconocene dichloride/trimethyl aluminum cyclized  $\omega$ -bromo alkynes (equation 175) [200]. Vinylnickel complexes were prepared by the insertion of alkynes into alkynickel complexes [201]. Nucleophilic addition reactions with cationic iron  $\pi$ -alkyne and related complexes has been reviewed [202], and has been used to synthesize functionalized enones (equation 176) [203].

(171)

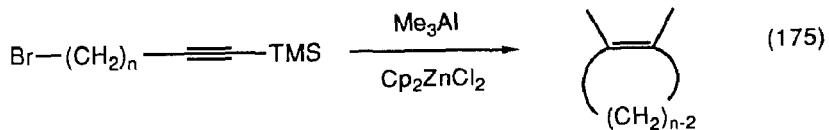
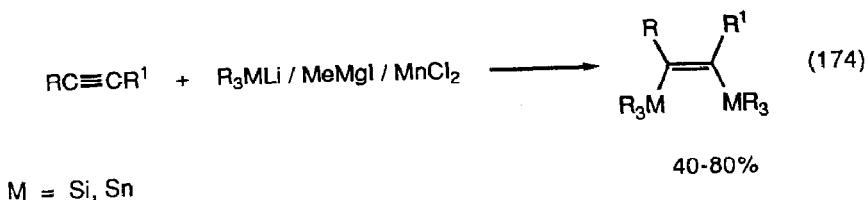
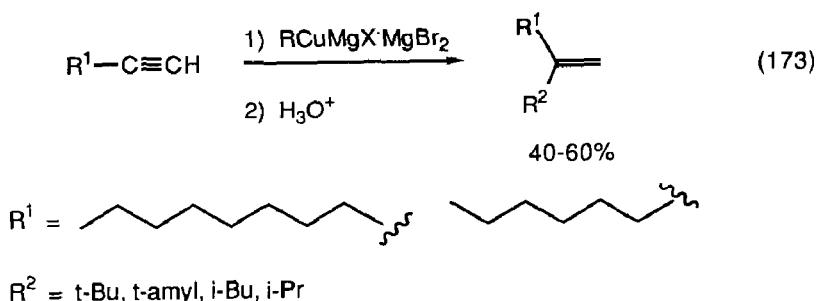


40-70%

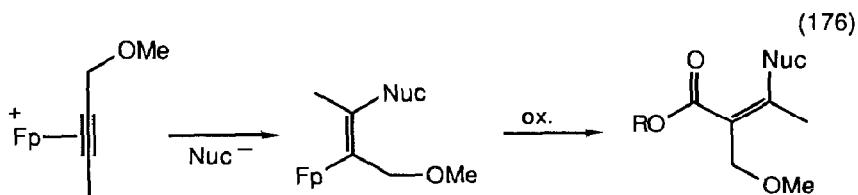
 $\text{R}^1 = \text{Me, Bu, Ph}$  $\text{R}^2 = \text{H, Me, Ph}$  $\text{R}^3 = \text{Me, Et}$ 

70-95%

 $\text{RM} = \text{Me}_2\text{CuLi}_2, \text{MeCuCNLi}, \text{Me}_2\text{CuCNLi}_2, \text{Et}_2\text{CuCNMgBr}_2, \text{iPrBuPh}$



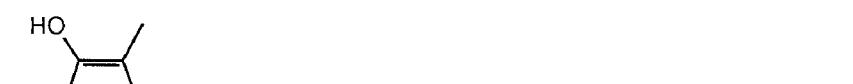
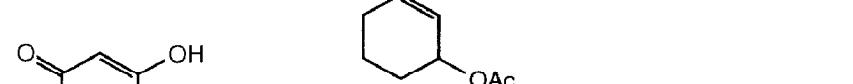
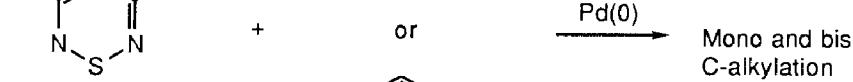
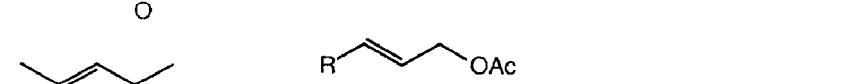
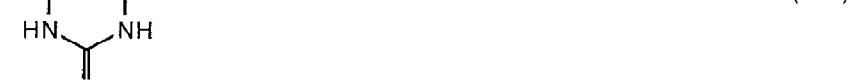
$n = 3, 5$



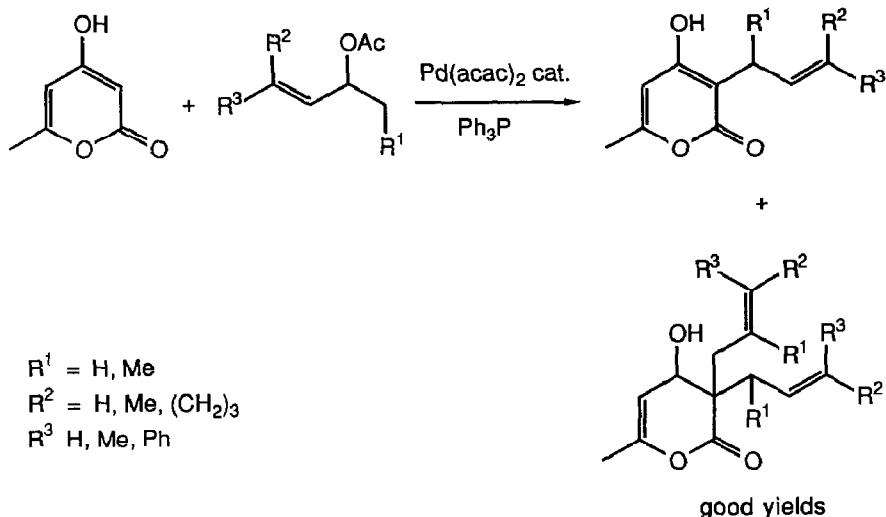
$\text{Nuc} = \text{Me}_2\text{CuCNLi}_2, \text{Ph}_2\text{CuCNLi}_2$

### 7. Alkylation of Allyl, Propargyl and Allenyl Systems

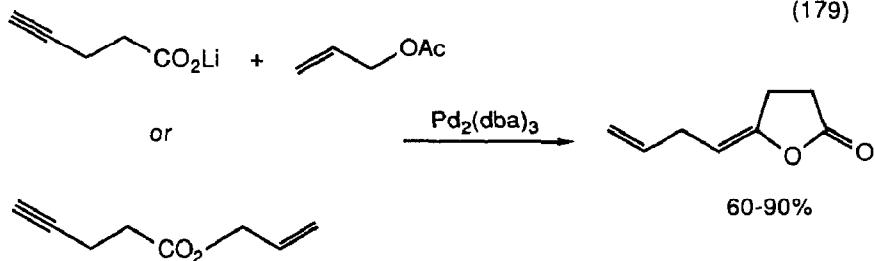
Metal-catalyzed alkylation of allylic acetates continues to be heavily exploited for synthesis. A variety of stabilized carbanions underwent mono and *bis* alkylation by allylic acetates in the presence of palladium(0) catalysts (equation 177) [204], (equation 178) [205] [206]. Iron nitrosyl carbonyl complexes catalyzed the alkylation of allylic acetates by malonates in a process claimed not to involve common  $\pi$ -allyl intermediates [207]. The lithium salt of 4-pentynoic acid was converted to a lactone when alkylated by allylic acetates in the presence of palladium(0) catalysts (equation 179) [208]. Allylic acetates containing trifluoromethyl groups also were efficiently alkylated by stabilized carbanions in the presence of palladium(0) catalysts (equation 180) (equation 181) [209]. Allylic acetate side chains on chromium arene complexes were efficiently alkylated by stabilized carbanions in the presence of palladium(0) catalysts (equation 182) [210].



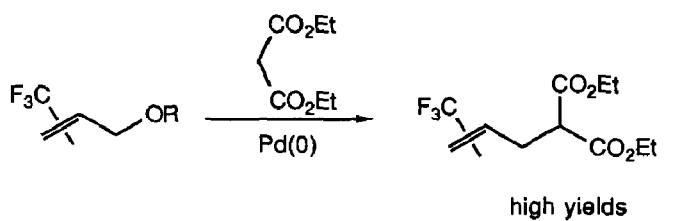
(178)

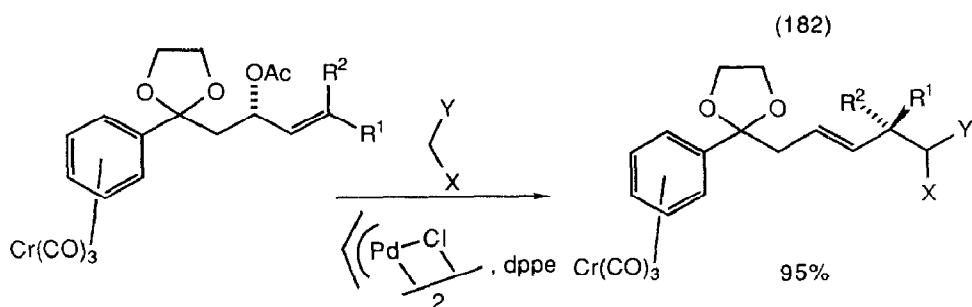
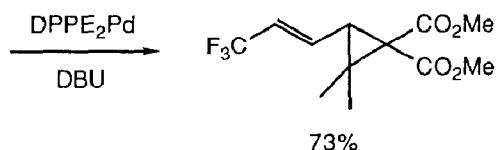
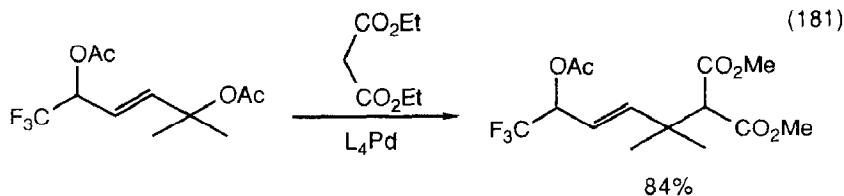


(179)



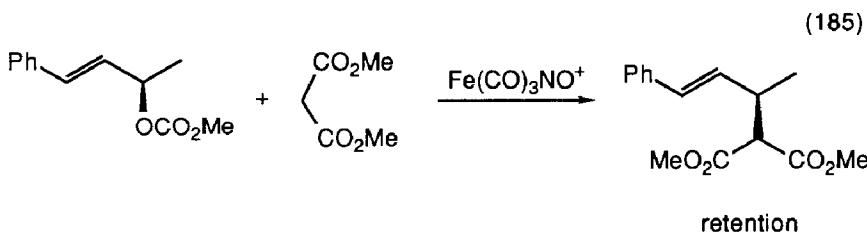
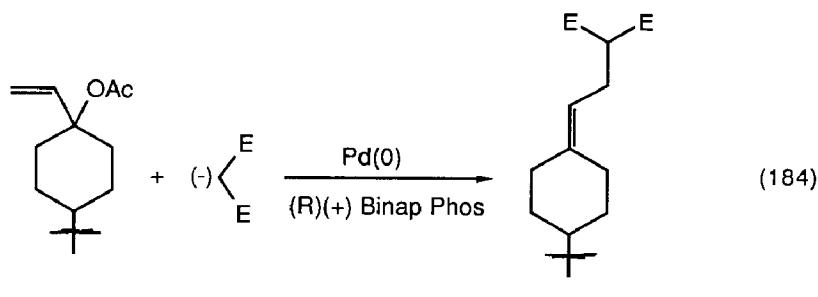
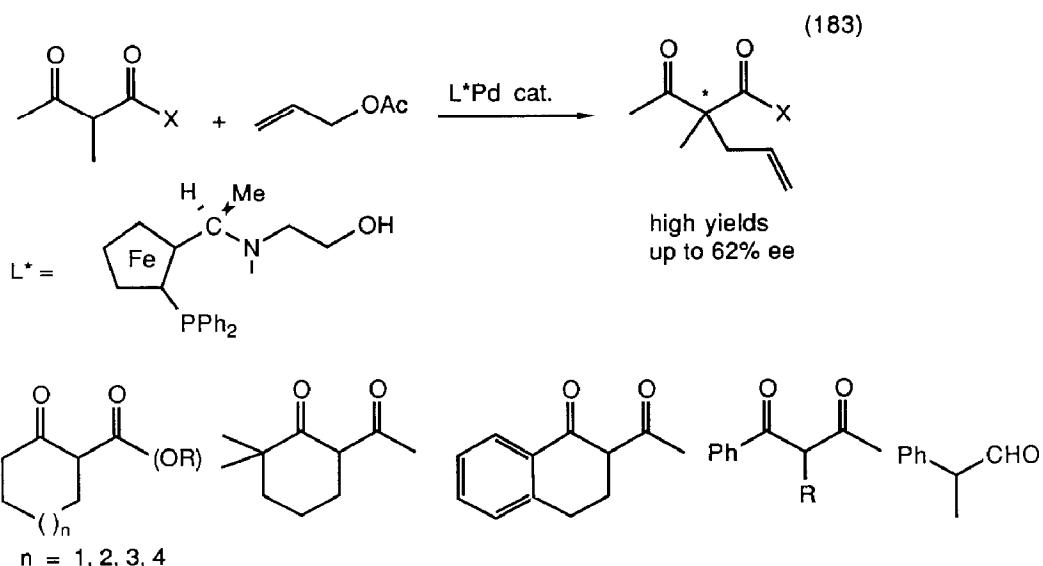
(180)

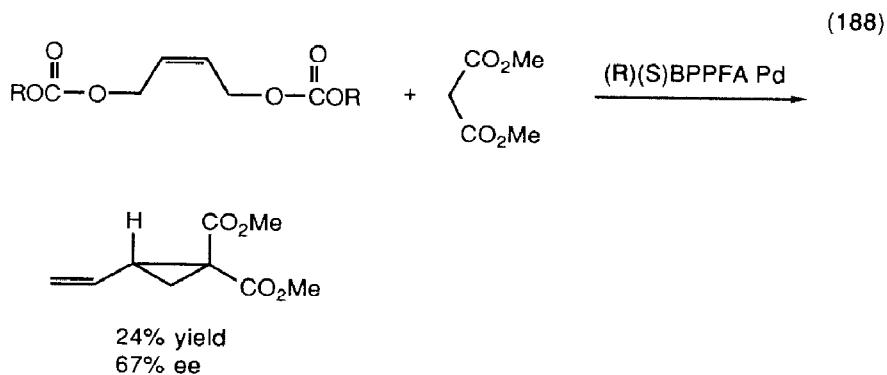
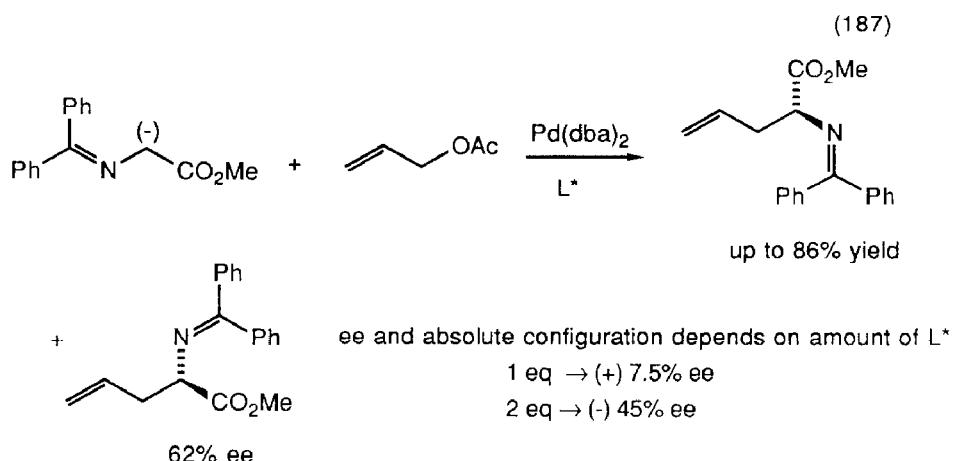
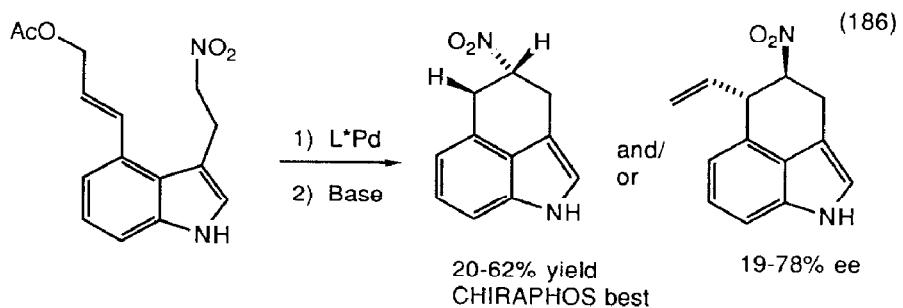


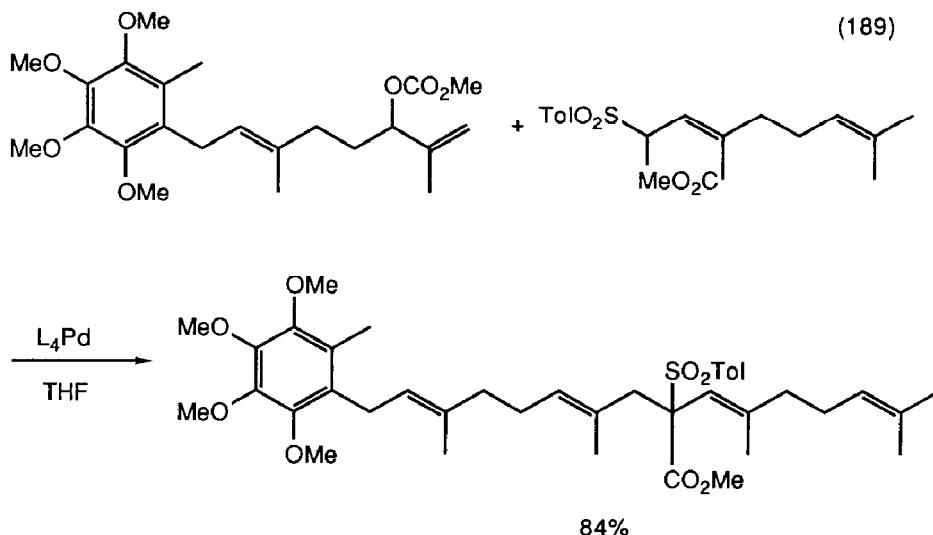


(S,S)  $R^1 = \text{Me}; R^2 = \text{H}$   
 $R^1 = \text{H}; R^2 = \text{Me}$

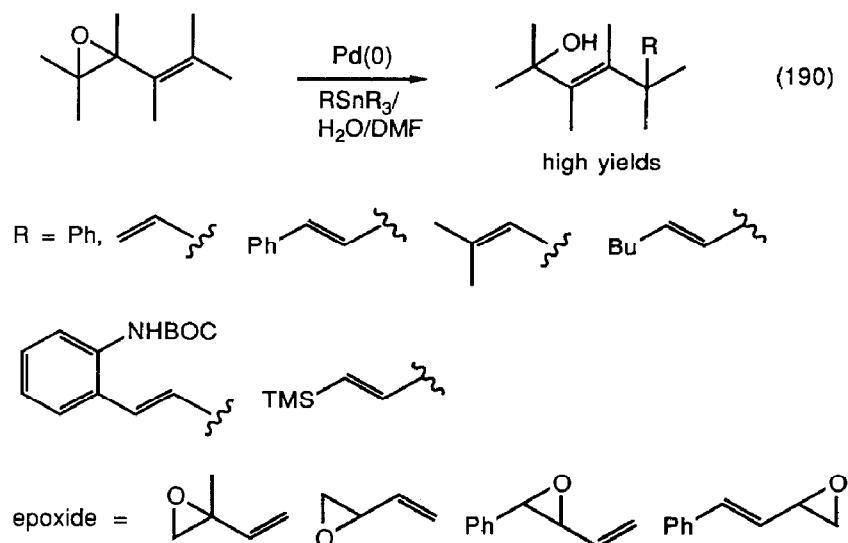
Use of chiral catalysts induced asymmetry in fair to excellent yields in the metal-catalyzed allylic alkylation process (equation 183) [211], (equation 184) [212], (equation 185) [213]. This was used in the synthesis of natural product precursors such as ergot alkaloids (equation 186) [214],  $\alpha$ -amino acids (equation 187) [215] and chrysanthemic acids (equation 188) [216]. Palladium catalyzed allylation of allylic acetates was shown to proceed with net retention, as the result of two inversions, while the same conversion catalyzed by  $\text{Mo}(\text{CO})_3(\text{MeCN})_3$  went with inversion, as a result of an oxidative addition with retention, followed by a nucleophilic attack with inversion [217]. Polyisoprenoid quinones were made by palladium(0) catalyzed allylation (equation 189) [218].

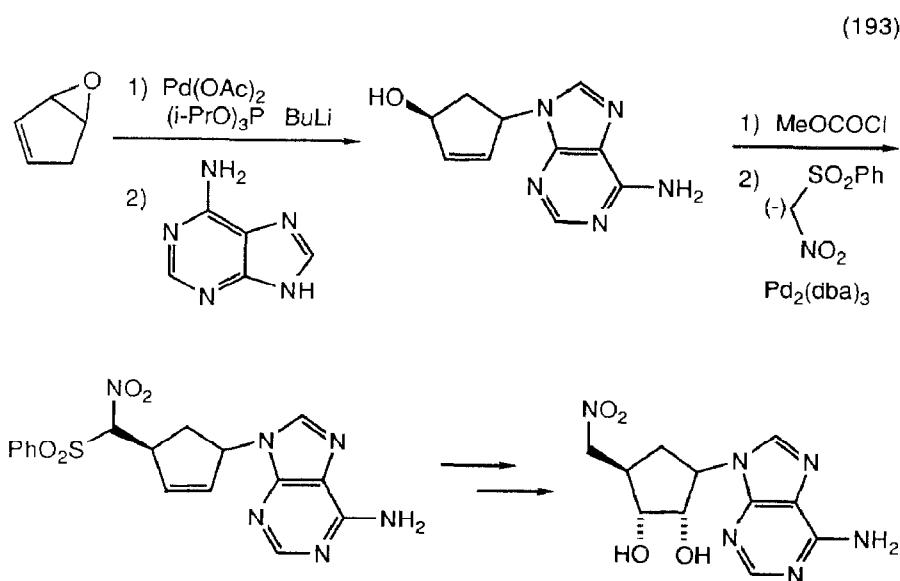
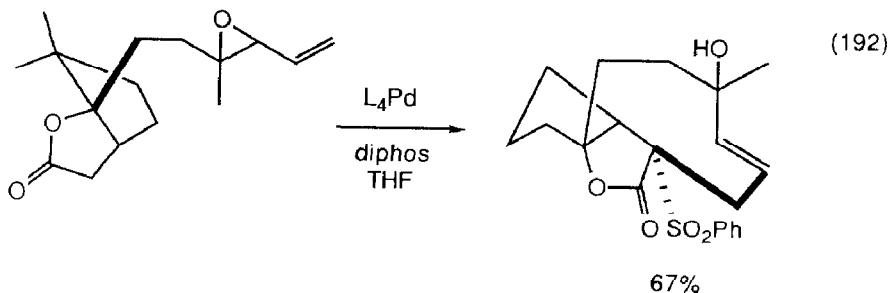
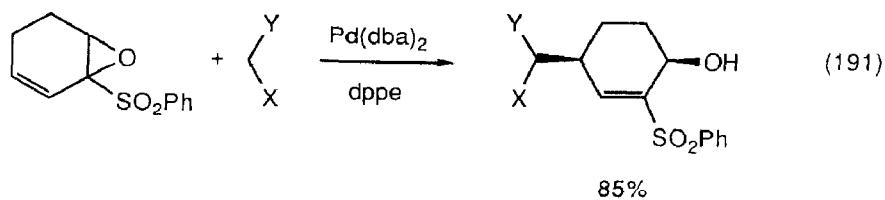


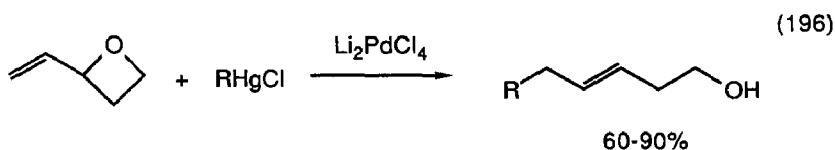
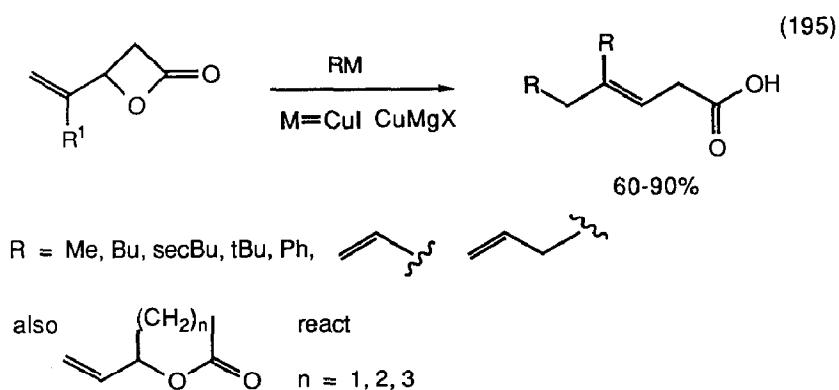
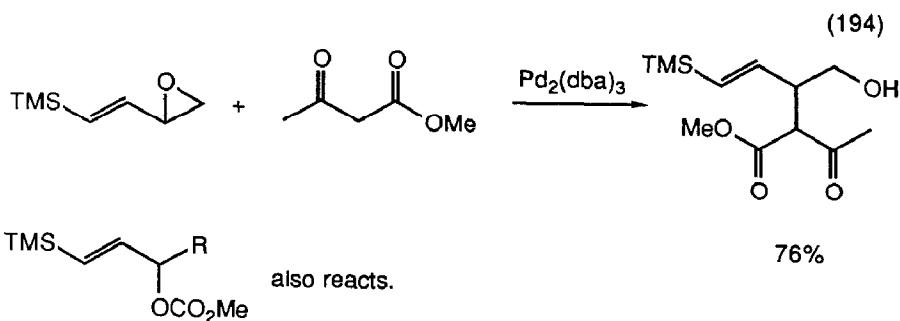




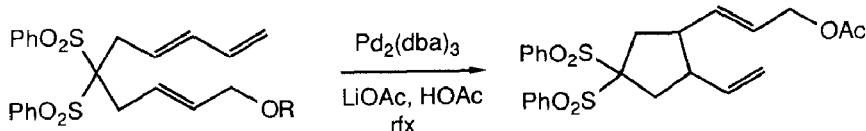
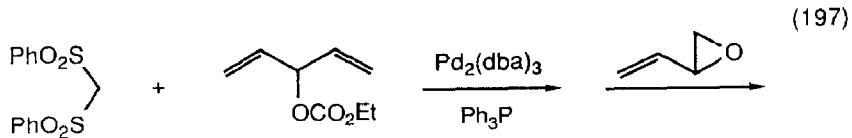
Palladium(0) complexes catalyzed a number of alkylation of allylic epoxides (equation 190) [219], (equation 191) [220], (equation 192) [221], (equation 193) [222], (equation 194) [223], and allyl- $\beta$ -lactones (equation 195) [224], allyl oxetanes (equation 196) [225], and allyl carbonates (equation 197) [226].



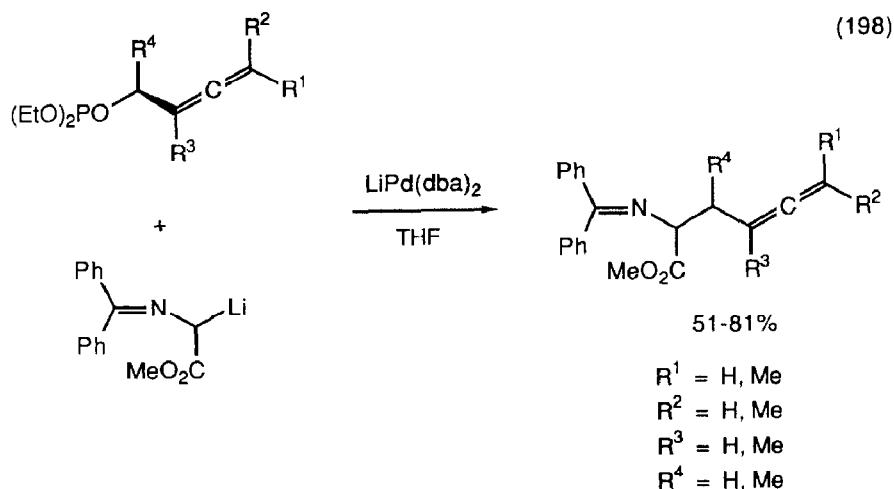


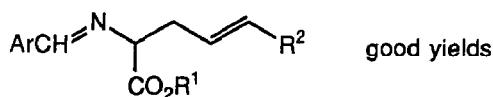
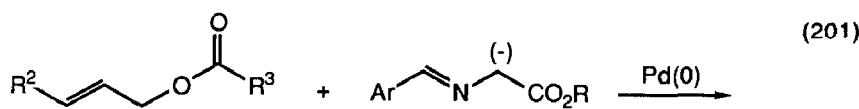
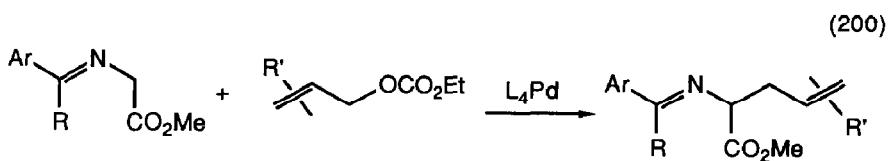
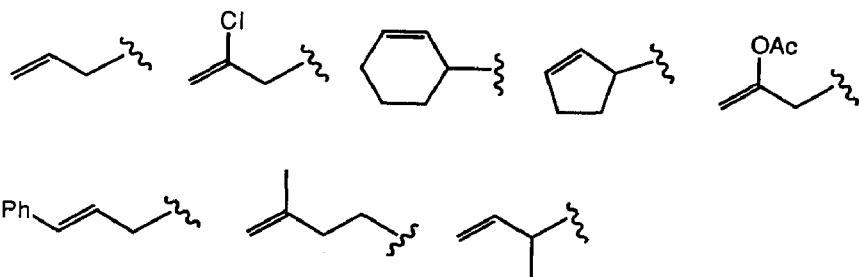
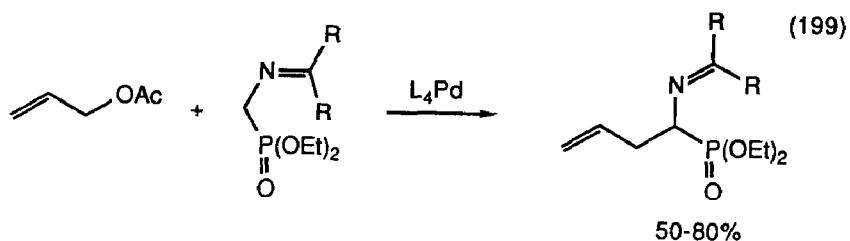


R = Ph, mMeOPh, mNO<sub>2</sub>Ph, mMeO<sub>2</sub>CPh

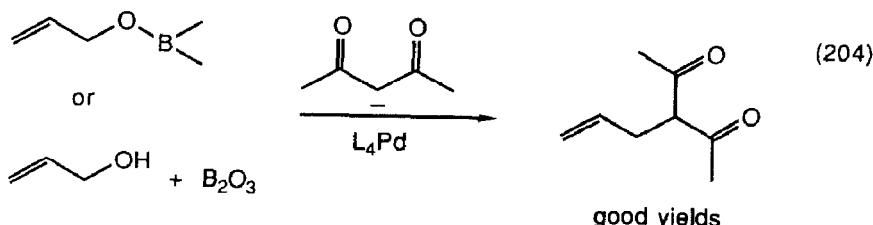
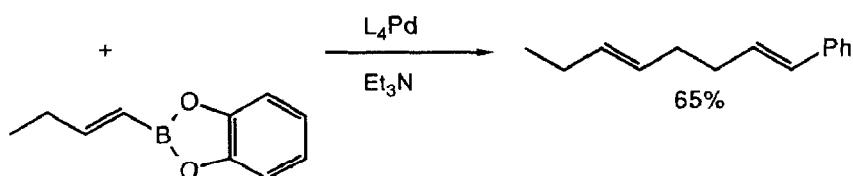
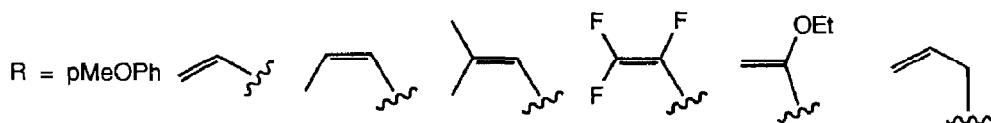
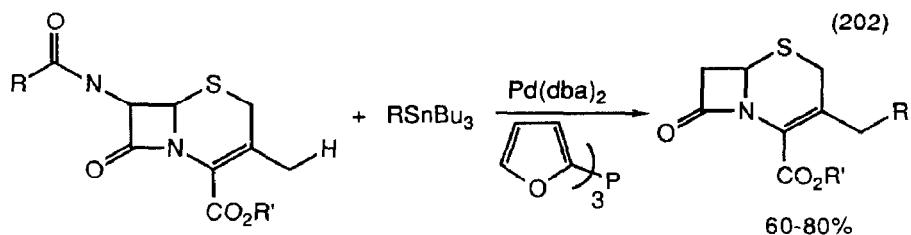


$\alpha$ -Amino acid precursors were synthesized by alkylating allylic phosphonates (equation 198) [227], acetates (equation 199) [228], and carbonates (equation 200) [229], (equation 201) [230] in the presence of palladium(0) catalysts. Metallated ketimines alkylated allyl esters in the presence of palladium catalysts [231].

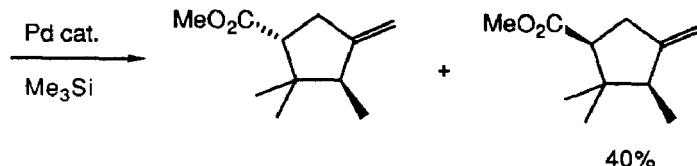
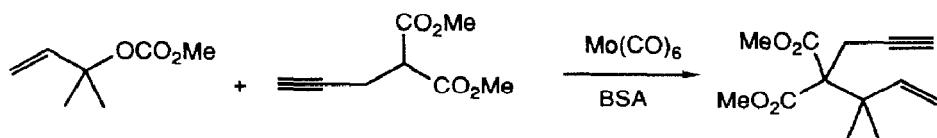


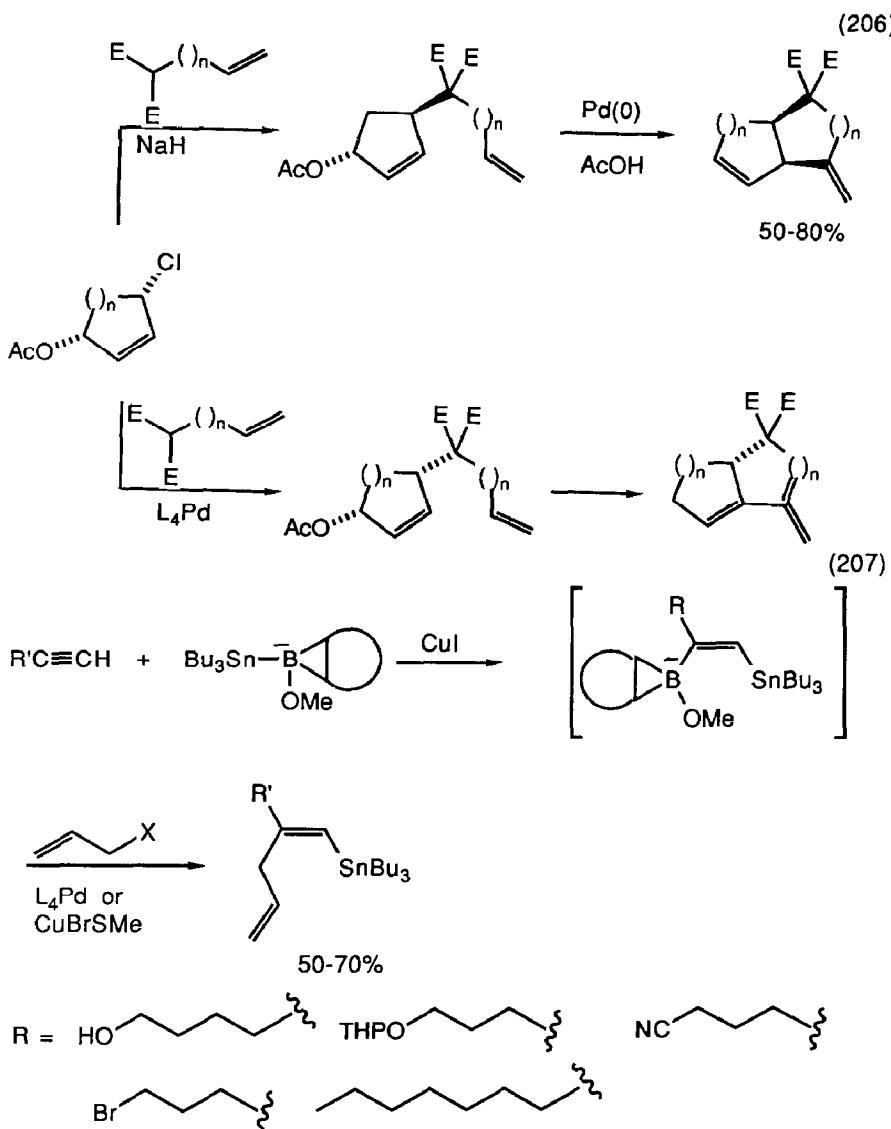


Palladium(0) complexes catalyzed the alkylation of allylic halides by tin reagents in cephalosporins (equation 202) [232]. Alkenylborates coupled to allylic phenoxides in the presence of palladium(0) catalysts (equation 203) [233], (equation 204) [234]. Cyclic (equation 205) [235] and polycyclic systems (equation 206) [236] were synthesized utilizing palladium(0) catalysts. Copper(I) was used to catalyze the alkylation of allylic halides by vinyl borates (equation 207) [237].

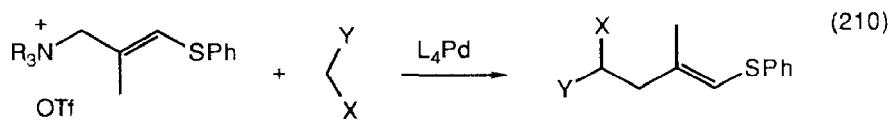
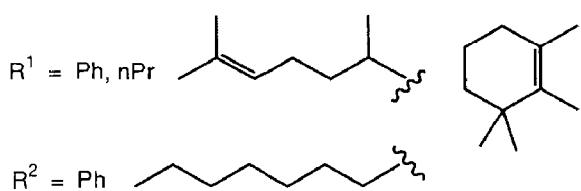
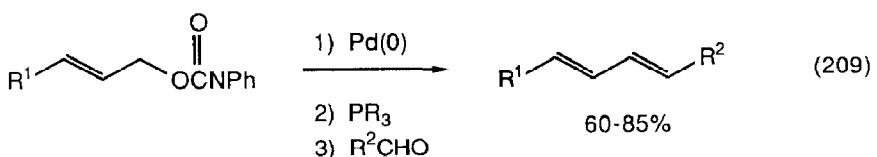
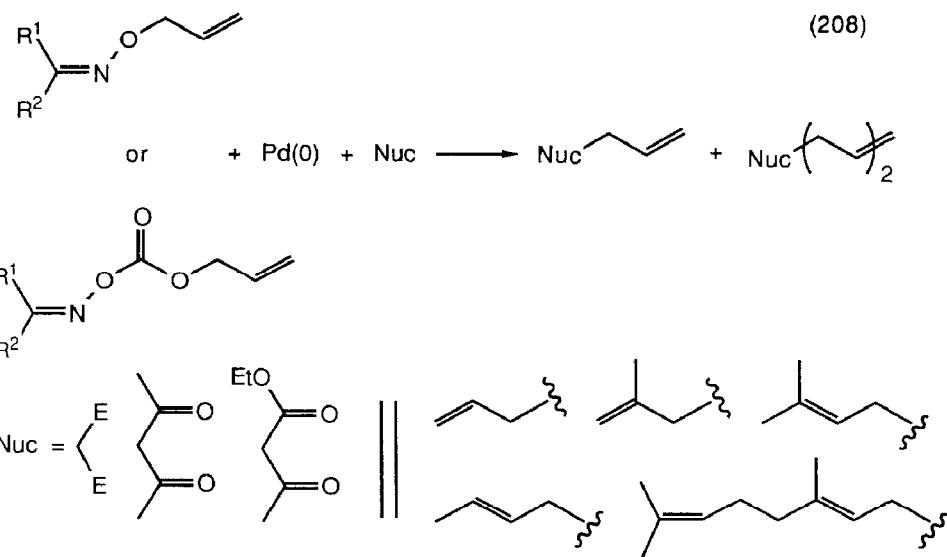


(205)

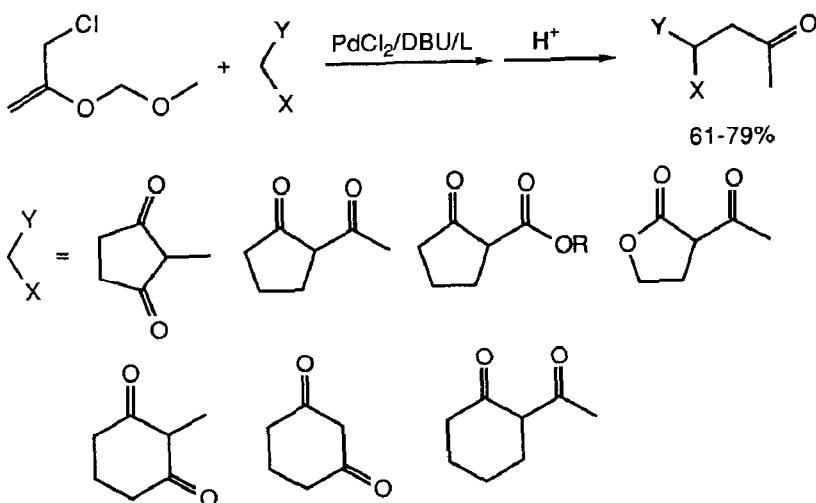




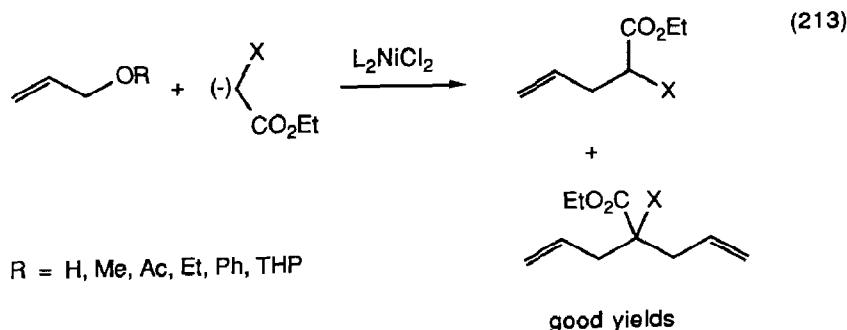
Allylic alcohols were alkylated by stabilized carbanions in the presence of palladium(0) and arsenic(III) oxide [238]. O-Allyl oximes and carbonates were alkylated in the presence of palladium(0) catalysts (equation 208) [239]. Allylic carbamates coupled to aldehydes to form dienes using palladium(0) catalysts (equation 209) [240]. N-Allyl quaternary ammonium salts were also alkylated using palladium catalysts (equation 210) [241] as were allylic chlorides (equation 211) [242]. Palladium catalyzed the arylation of allylic alcohols by iodobenzene (equation 212) [243].



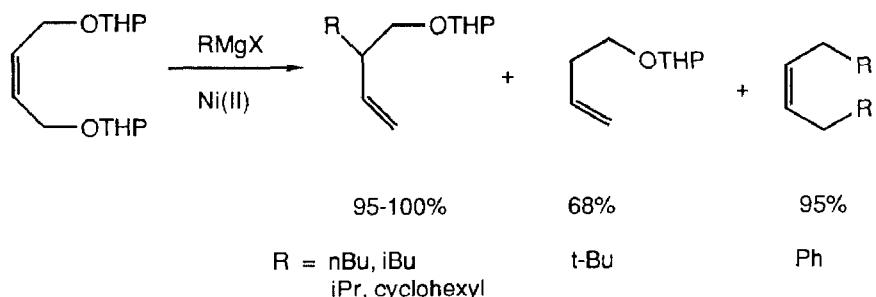
(211)



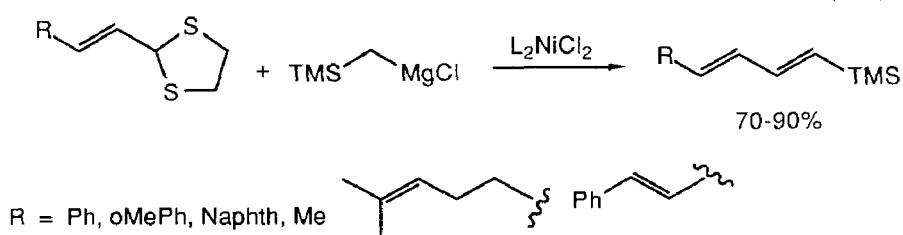
Nickel(II) phosphines catalyzed the alkylation of a wide range of allylic substrates by stabilized carbanions (equation 213) [244]. Nickel(II) salts catalyzed the reaction between Grignard reagents and allyl THP ethers (equation 214) [245] and dithianes of unsaturated aldehydes (equation 215) [246]. Chiral  $\pi$ -allylnickel halides alkylated a variety of organic halides (equation 216) [247].



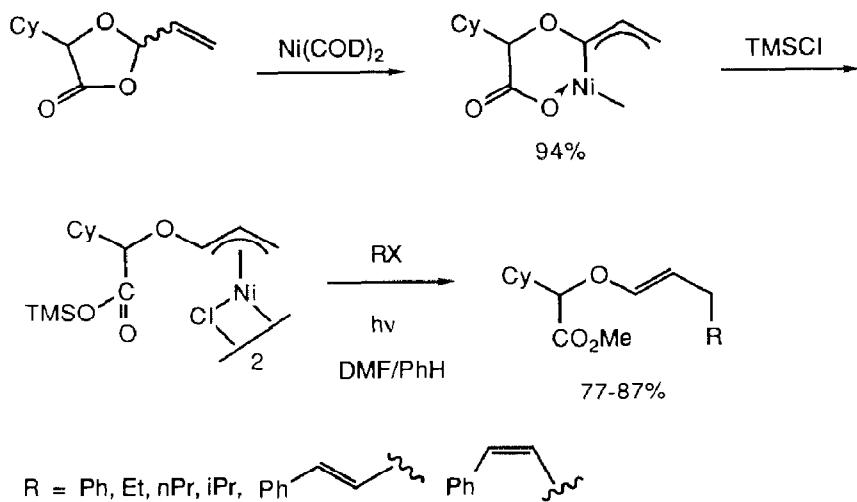
(214)



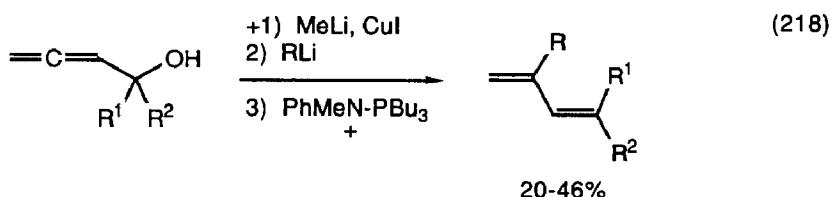
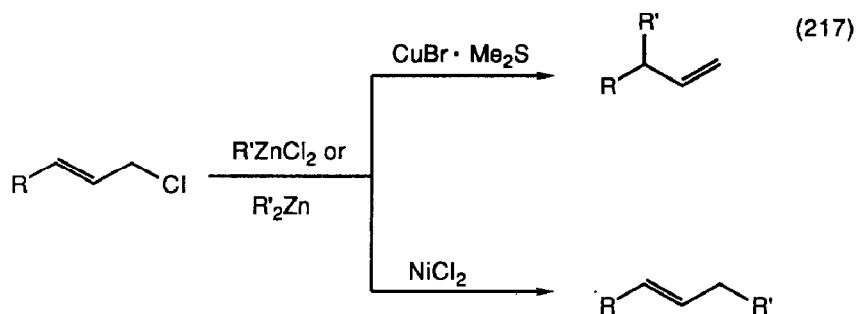
(215)



(216)



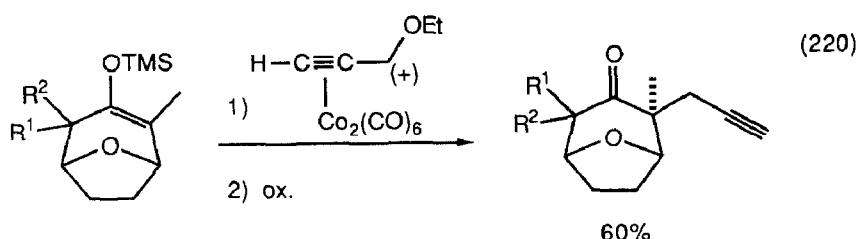
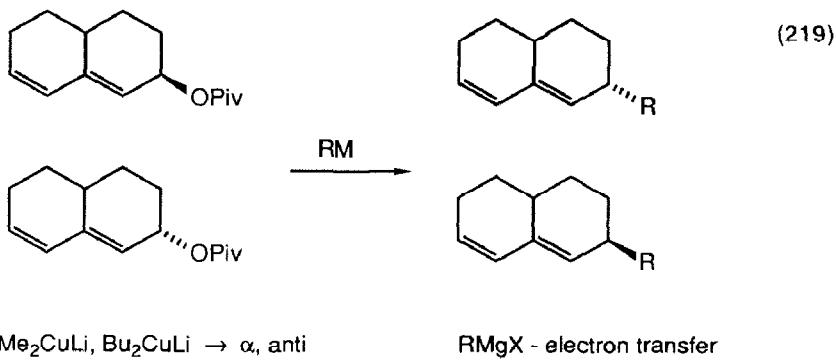
Alkylzinc halides alkylated allylic halides by  $S_N2'$  processes in the presence of copper(I) bromide, and by  $S_N2$  processes in the presence of nickel(II) salts (equation 217) [248]. Allenic alcohols were alkylated by copper reagents to give dienes (equation 218) [249]. The stereochemistry of the alkylation of allylic ethers by copper reagents has been studied (equation 219) [250]. Cobalt stabilized propargyl cations were alkylated by trimethylsilylenol ethers (equation 220) [251].



$R^1 = H$

$R^2 = H, Ph$

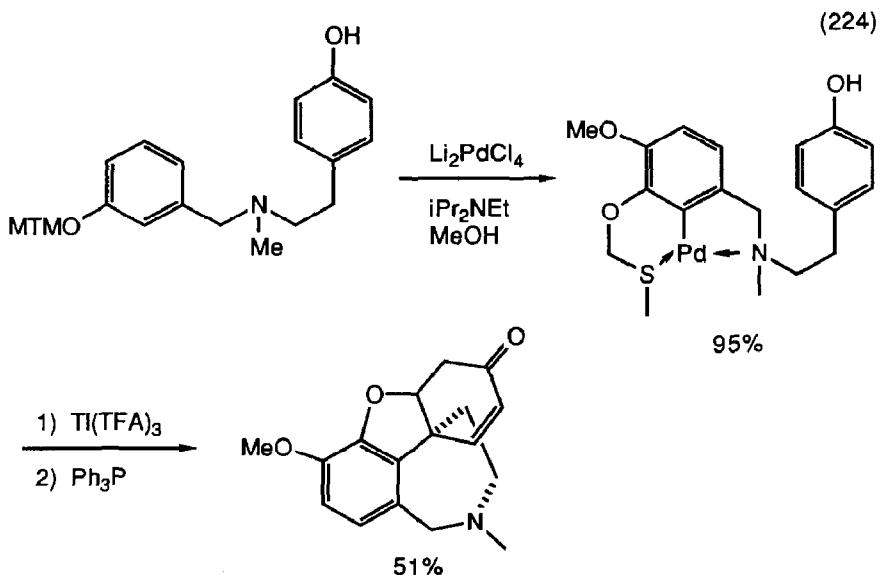
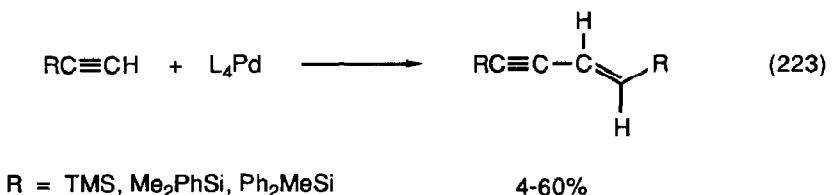
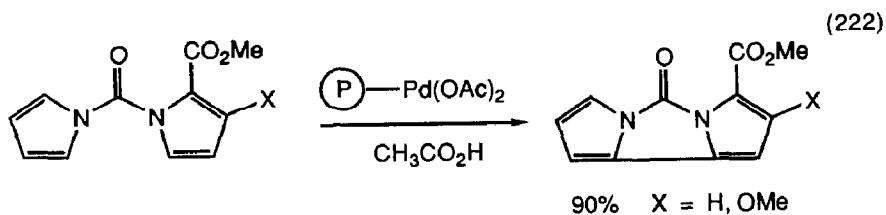
$R = Me, Ph$



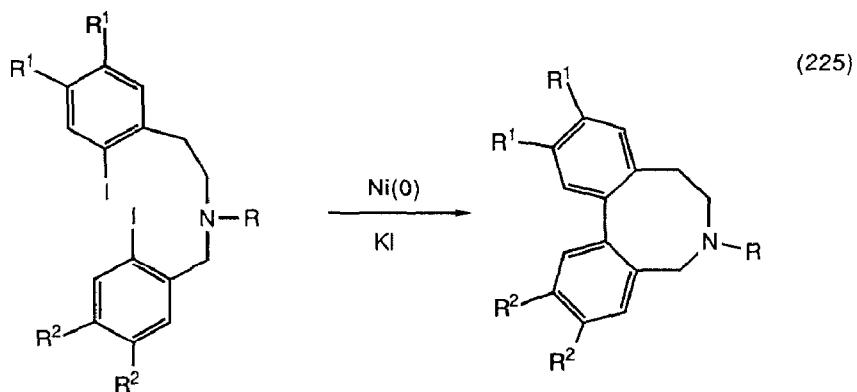
### 8. Coupling Reactions

Palladium complexes catalyzed the homocoupling of a number of organometallic reagents including vinylstannanes to 1,3-dienes [252], aryl bismuth and antimony complexes to biaryls [253] and aryltellurium trichlorides to biaryls [254]. Unsymmetrical dialkyltellurium(II) species also coupled (equation 221) [255]. Polymer supported palladium(II) effected aryl coupling (equation 222) [256] while palladium(0) catalyzed the dimerization of terminal alkynes (equation 223) [257]. Oxidation of a complex o-palladated aromatic species resulted in coupling to give norwidine (equation 224) [258].

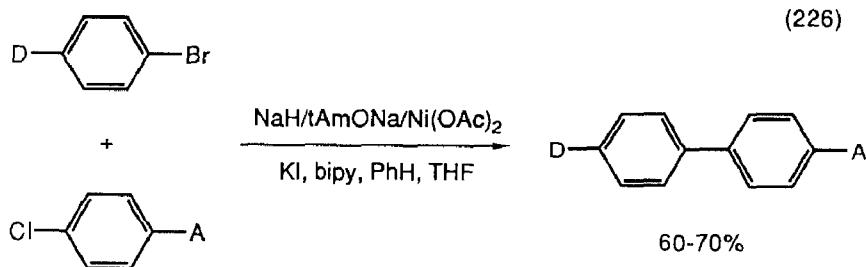




Nickel(0) catalyzed the coupling of aryl halides in the preparation of  $\alpha$ -androgenic blocking agents (equation 225) [259]. Unsymmetrical biaryls were prepared by nickel complex coupling of aryl halides (equation 226) [260]. Chiral biaryls were synthesized using copper powder coupling of aryl halides (equation 227) [261]. Aryl chlorides coupled when treated with activated copper metal (equation 228) [262].

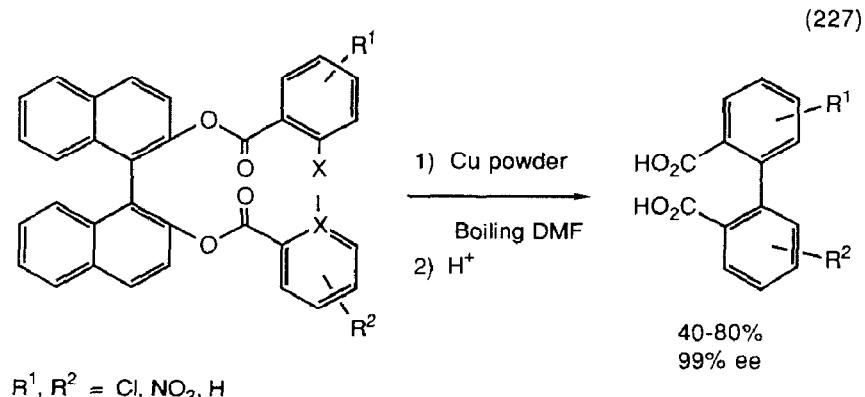


$R^1, R^2 = H, OMe$

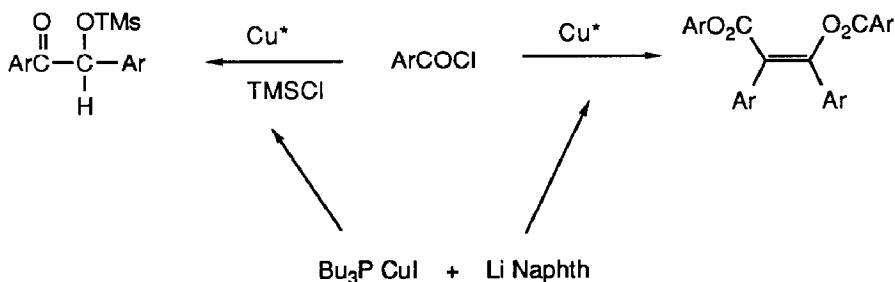


D = MeO, OH, SMe, Me<sub>2</sub>N

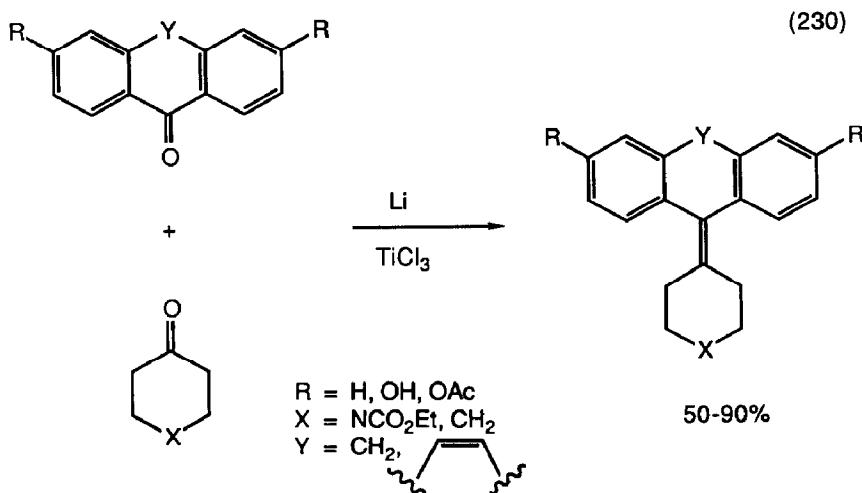
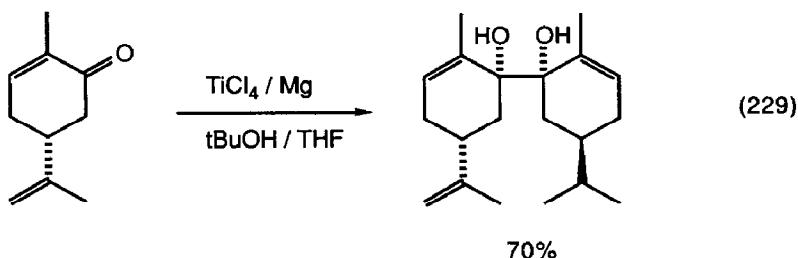
A = CF<sub>3</sub>, F, CH(OMe)<sub>2</sub>, CMe(OCH<sub>2</sub>)<sub>2</sub>CN

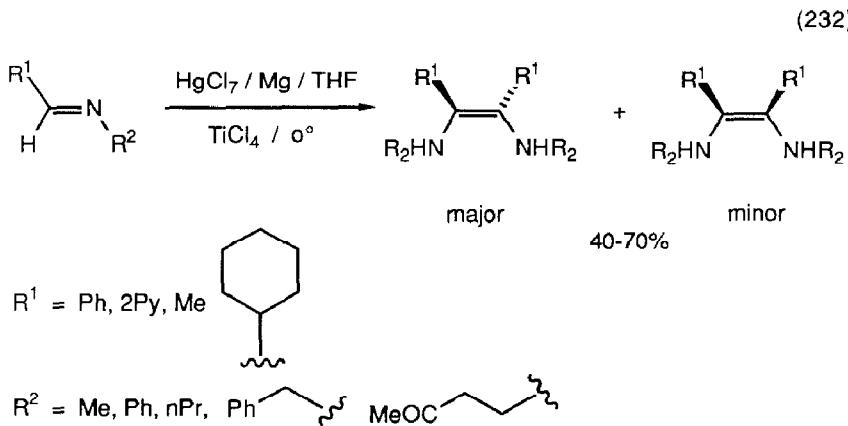
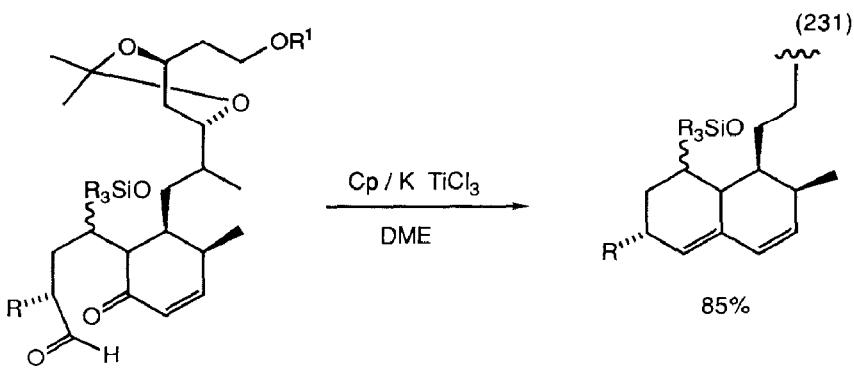


(228)

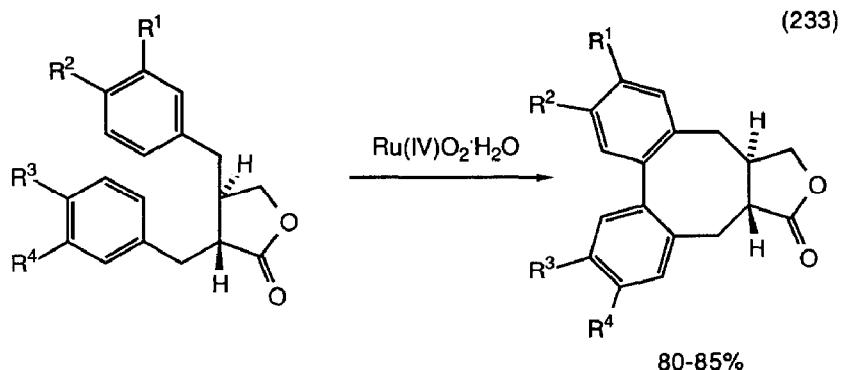


Carbonyl coupling reactions using transition metals, lanthanides and actinides have been reviewed (104 references) [263]. Low valent titanium coupled ketones to give diols (equation 229) [264] and alkenes (equation 230) [265], (equation 231) [266], and coupled imines to give diamines (equation 232) [267]. Reductive coupling using zero valent titanium has been reviewed (13 references) [268].



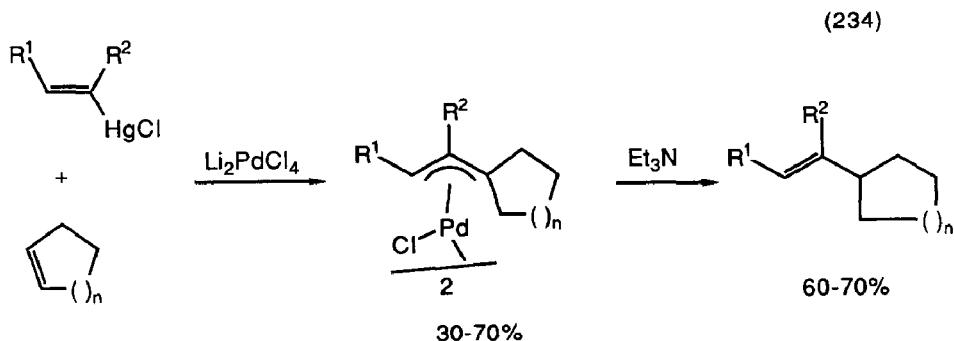


Ruthenium(IV) oxidatively coupled aromatics (equation 233) [269]. The oxidative coupling of arenes was catalyzed by osmium(IV), iridium(IV) and rhodium(III) chloride under photolytic conditions [270]. Molybdenum pentachloride oxidatively coupled benzyltrimethylsilane with aromatic compounds [271].

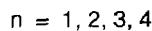
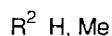
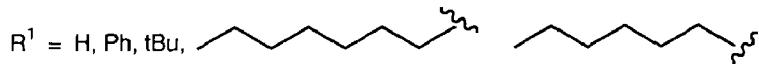


### 9. Alkylation of $\pi$ -Allyl Complexes

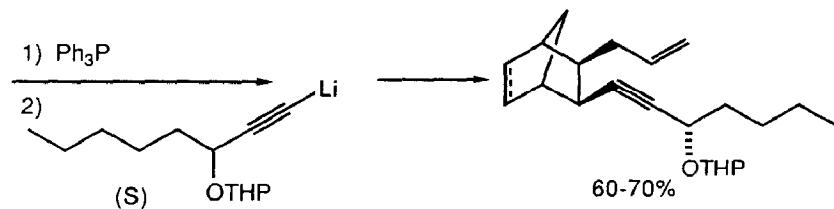
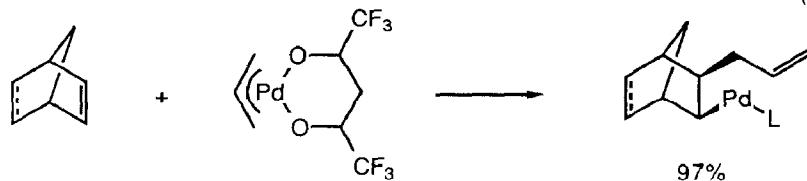
The mechanism of formation of  $\pi$ -allylpalladium complexes from olefins was the topic of a dissertation [272], wherein it was shown that intramolecular abstraction of a proton by chloride was a key step [273]. Vinyl mercuric halides added to olefins to give  $\pi$ -allylpalladium chloride complexes in the presence of palladium(II) chloride (equation 234) [274]. Bicyclic analogs of prostaglandins were synthesized from olefin insertion into  $\pi$ -allylpalladium chloride complexes (equation 235) [275].  $\pi$ -Allylpalladium complexes were generated by decarboxylation of allyl  $\beta$ -keto carboxylates [276]. Photolytic decomposition of  $\pi$ -allylpalladium halides produced 1,5-dienes or conjugated ketones, depending on conditions (equation 236) [277]. The stereochemistry of nucleophilic attack on chiral  $\pi$ -allylpalladium complexes was (again) shown to go with inversion [278]. Phenylmercuric chloride arylated  $\pi$ -allylpalladium chloride complexes to give allyl benzene [279]. TCNE oxidatively coupled the allyl ligands of *bis*- $\pi$ -allylpalladium to produce biallyl [280].  $\pi$ -Allyliron complexes underwent alkylation by stabilized carbanions (equation 237) [281].



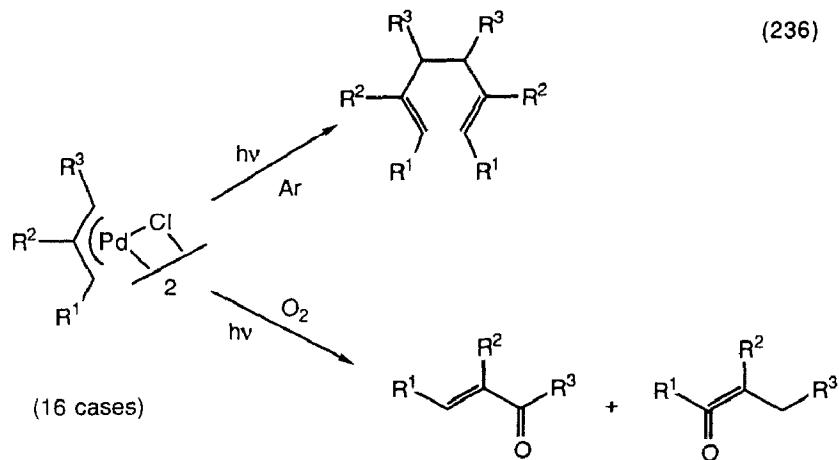
(equation 234 continued)

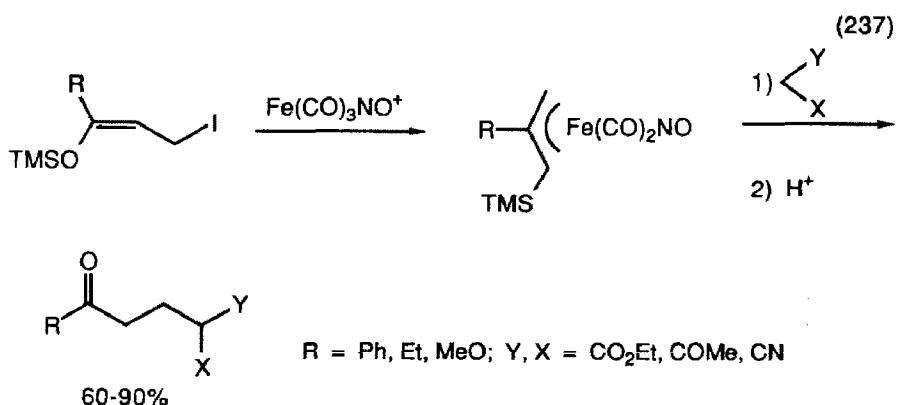


(235)



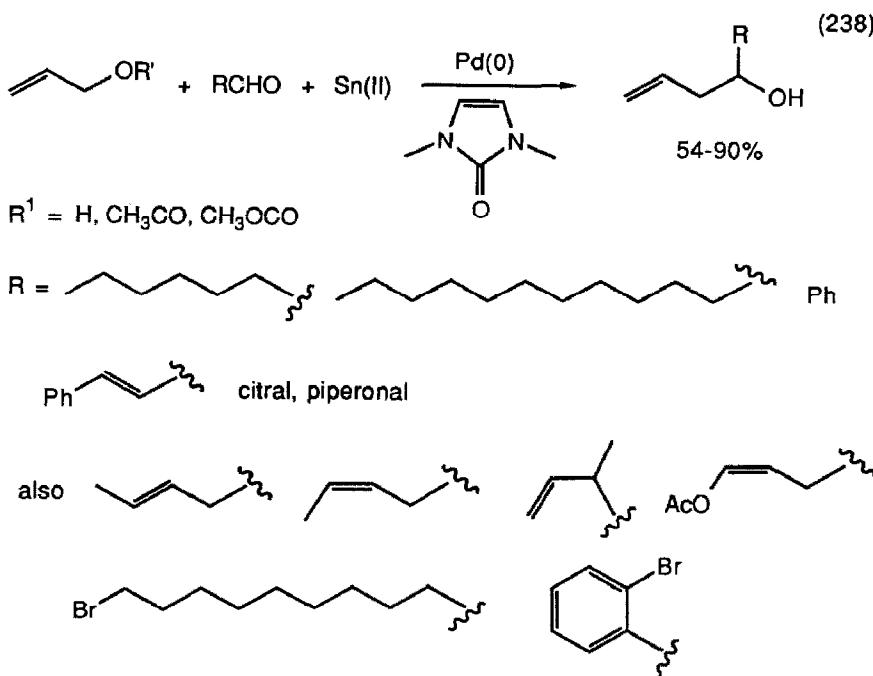
(236)

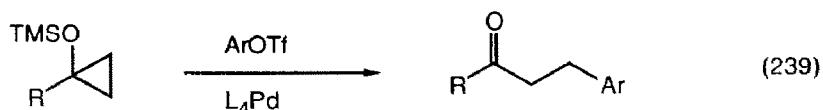




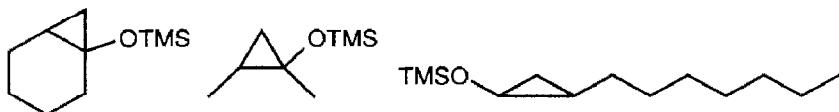
#### 10. Alkylation of Carbonyl Compounds

Palladium(0)/tin(II) systems catalyzed the alkylation of aldehydes by allylic acetates (equation 238) [282], allylic carbonates [283], and allylic alcohols [284]. Palladium(0) catalyzed the alkylation of homoenolates by aryl triflates (equation 239) [285].



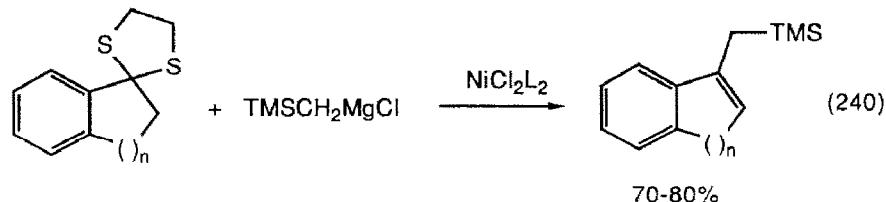


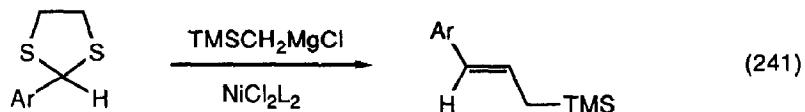
Ar = 1-Naphth, p-NO<sub>2</sub>Ph, p-MeOPh



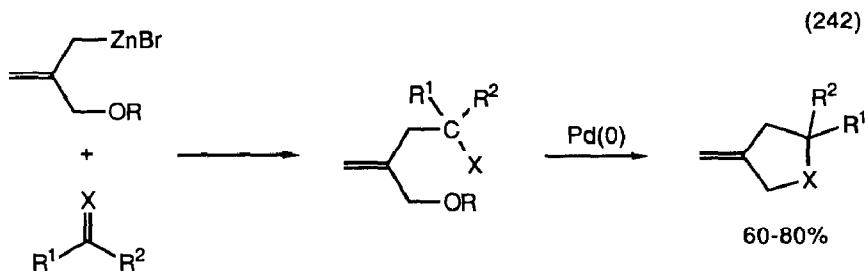
also work.

Nickel(II) chloride/phosphine complexes catalyzed the alkylation of dithianes by Grignard reagents (equation 240) [286], (equation 241) [287]. Ketones were alkylated by alkyl zinc reagents, then closed to furans or pyrrolidines by palladium(0) catalysts (equation 242) [288]. Aldehydes were alkylated by functionalized organocupper species in the presence of BF<sub>3</sub>·OEt<sub>2</sub> (equation 243) [289]. Formamides were converted to imines by reaction with organocuprates [290]. The effects of added trimethylsilyl chloride on the reaction of higher order cuprates with  $\alpha$ -phenylacetaldehyde was studied [291].

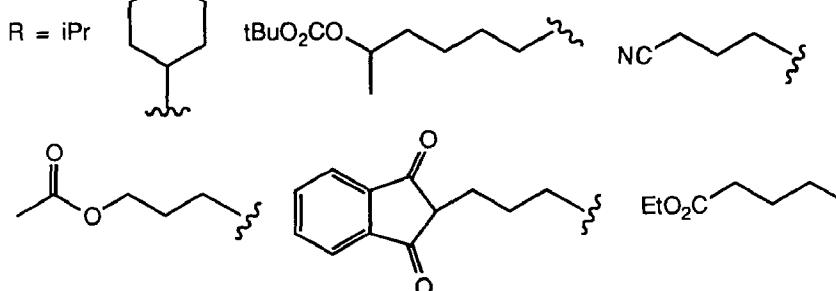
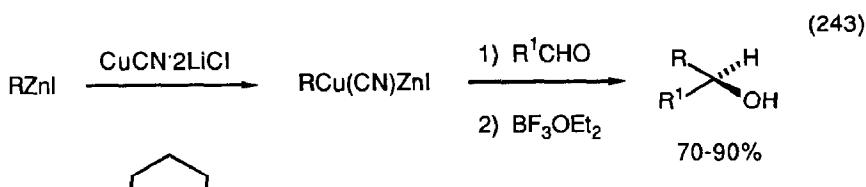
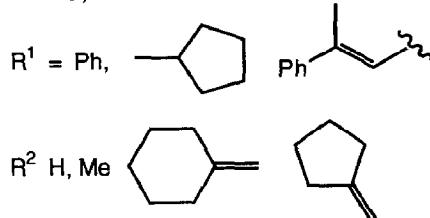




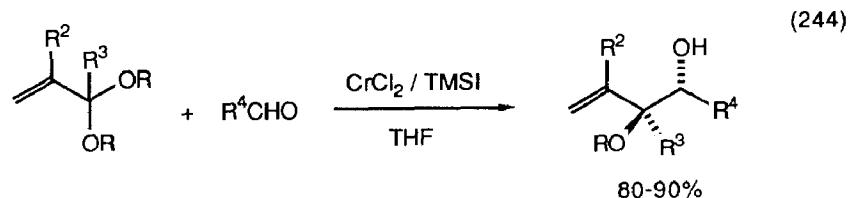
$\text{Ar} = \text{Ph}, \text{oMePh}, \text{pMeOPh}, 3,5\text{-Me}_2\text{Ph}, 3,4\text{-}\begin{array}{c} \text{O} \\ \diagup \\ \text{O} \end{array}\text{Ph}$



$\text{X} = \text{O}, \text{NR}^3$

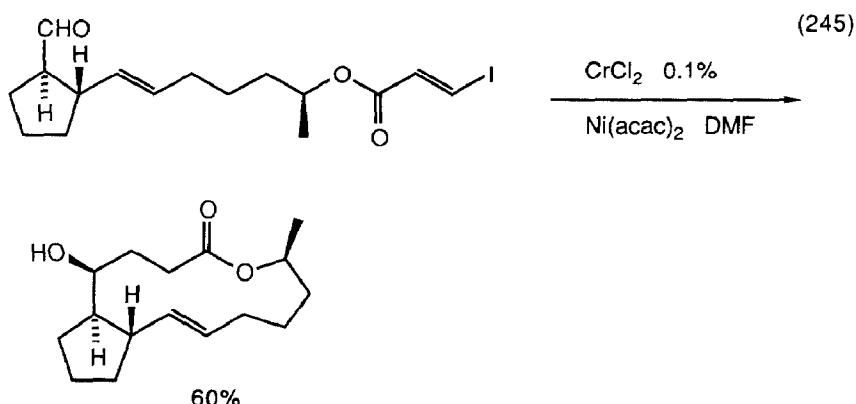
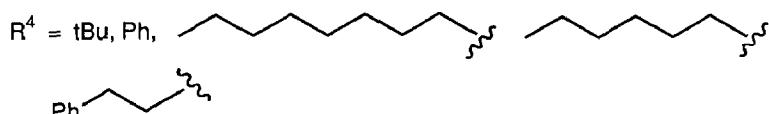


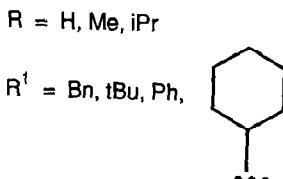
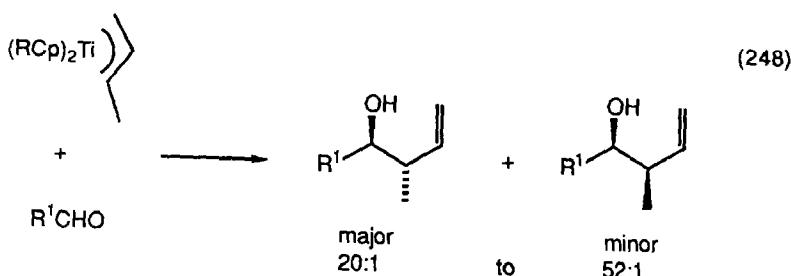
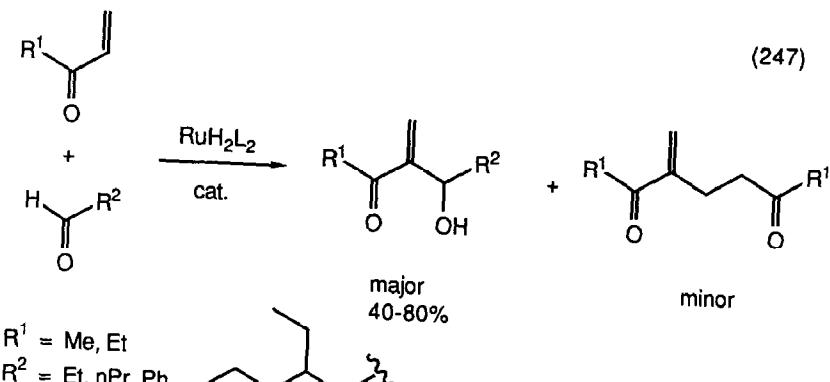
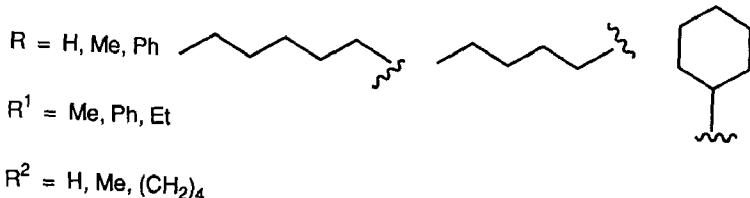
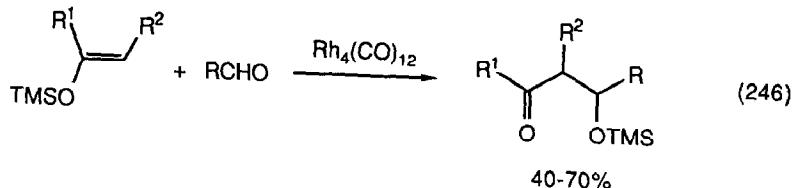
Chromium(II) chloride reductively alkylated aldehydes with acetals (equation 244) [292] and, intramolecularly, with vinyl halides (equation 245) [293]. Rh<sub>4</sub>(CO)<sub>12</sub> catalyzed the alkylation of aldehydes and acetals by trimethylsilylenol ethers (equation 246) [294,295]. Ruthenium(II) hydrides catalyzed the alkylation of conjugated enones by aldehydes (equation 247) [296]. Allyltitanium species alkylated carbonyl compounds (equation 248) [297], (equation 249) [298], (equation 250) [299].

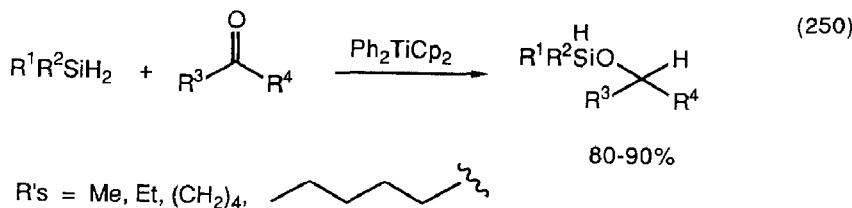
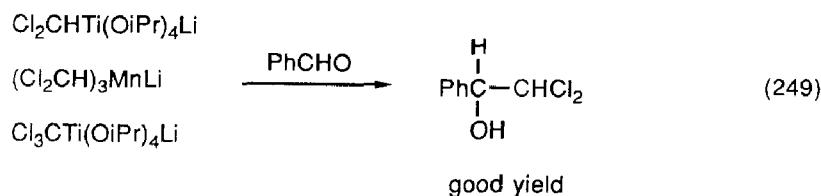


$\text{R} = \text{Me, Bn}$

$\text{R}^2, \text{R}^3 = \text{H, Me}$

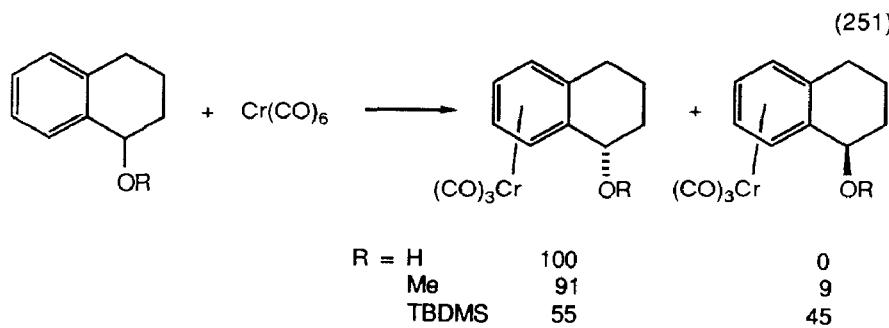




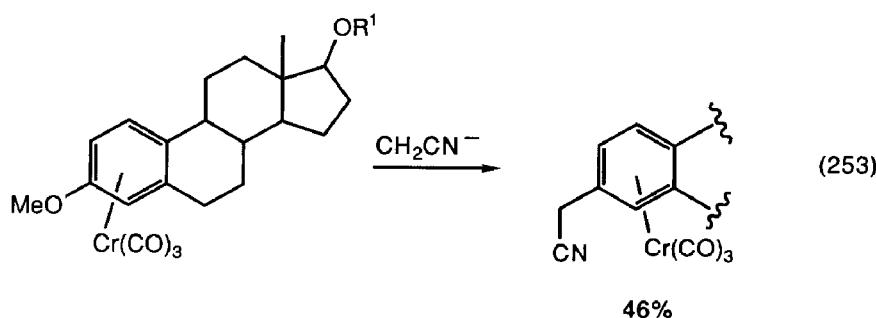
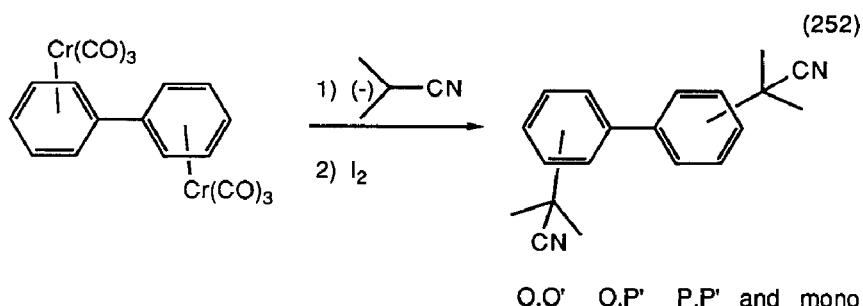


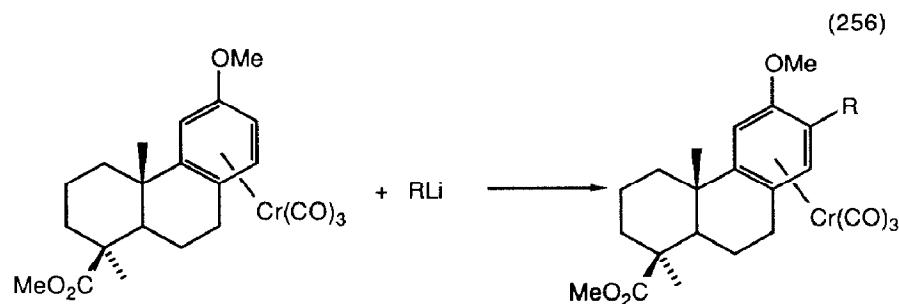
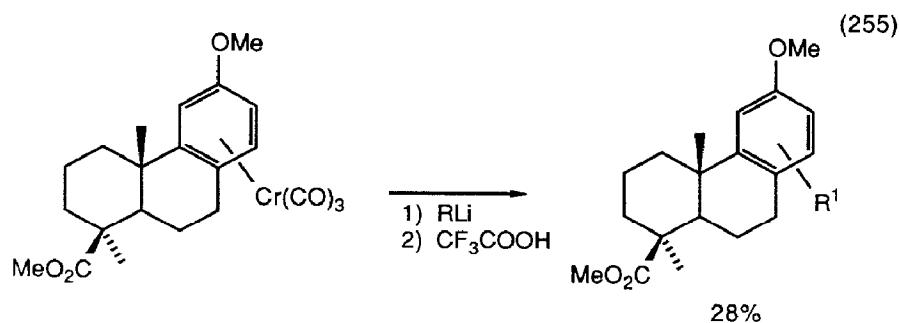
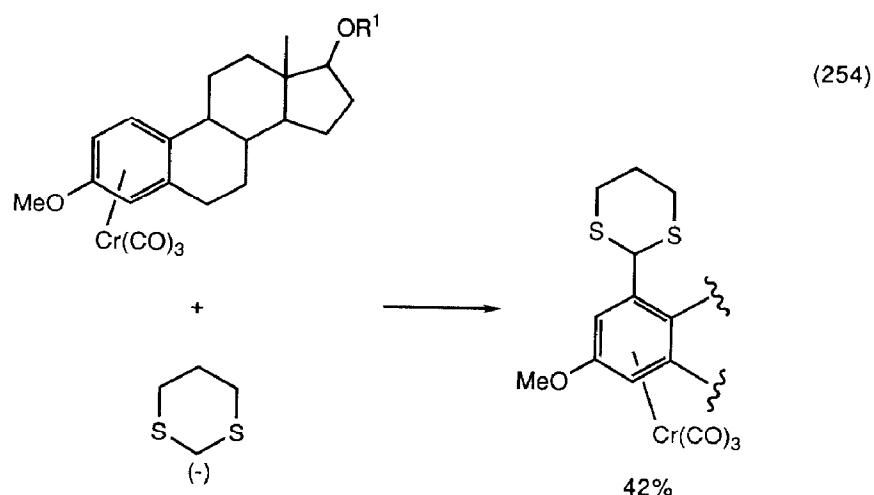
### 11. Alkylation of Aromatic Compounds

The following reviews on this subject have appeared: "Arenechromium tricarbonyls in organic synthesis" (167 references) [300,301]; "Aromatic nucleophilic substitution reactions in arenes activated by  $\pi$ -complex formation with transition metals" (69 references) [302]; and "Nucleophilic addition and substitution reactions on arenes complexed with cyclopentadienyl iron" (dissertation) [303]. Chromium hexacarbonyl complexes tetrahydronaphthalenes preferentially from the face of the oxygen (equation 251) [304].

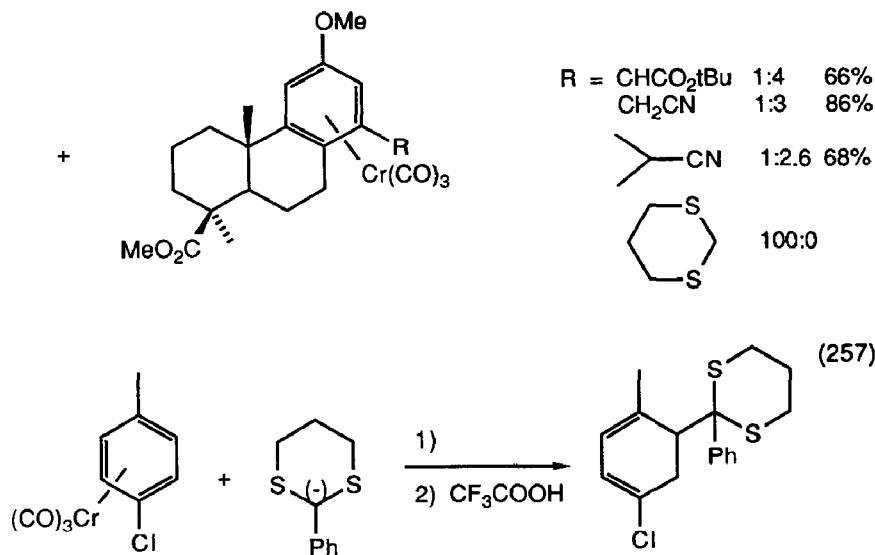


Bis-chromium complexes of diphenyl underwent *bisalkylation* to give mixture of isomers (equation 252) [305]. Acetonitrile anion displaced an aromatic methoxy group on the chromium complexed ring of an aromatic steroid (equation 253) [306], while dithiane alkylated the ring meta to it (equation 254) [307]. Alkylation of chromium complexed tricyclic species has been developed (equation 255) [308], (equation 256) [309]. Dithianes added to chromium complexed *p*-chlorotoluene to give the substituted cyclohexadiene after protonation (equation 257) [310]. Chromium complexes indolines, tetrahydroquinolines and dimethylanilines were reversibly alkylated, and the reaction could be optimized to give the desired regioisomer as the major product by adjusting reaction conditions [311].

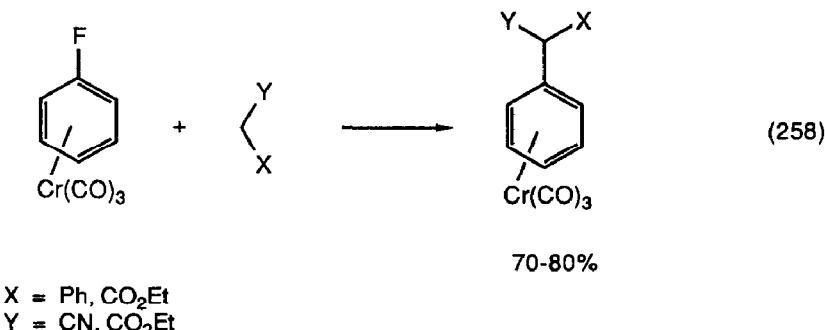


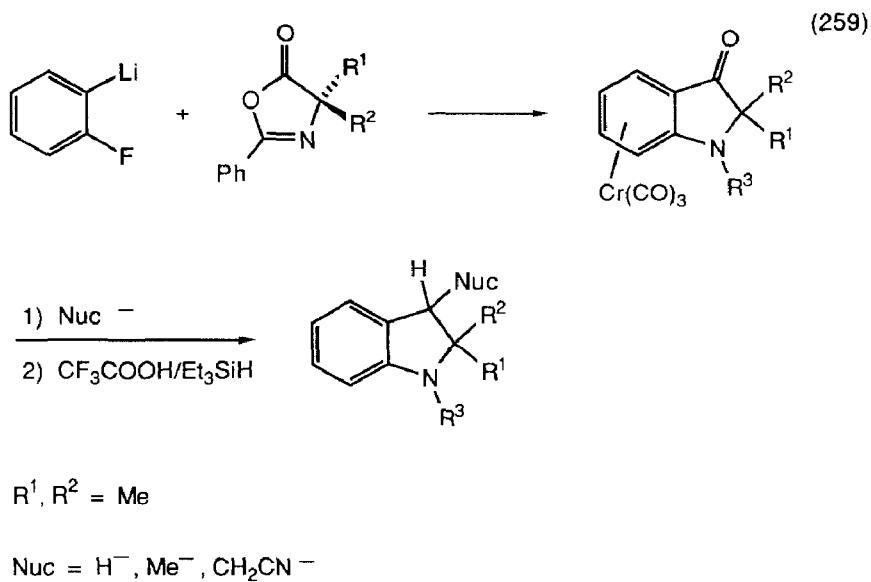


(equation 256 continued)

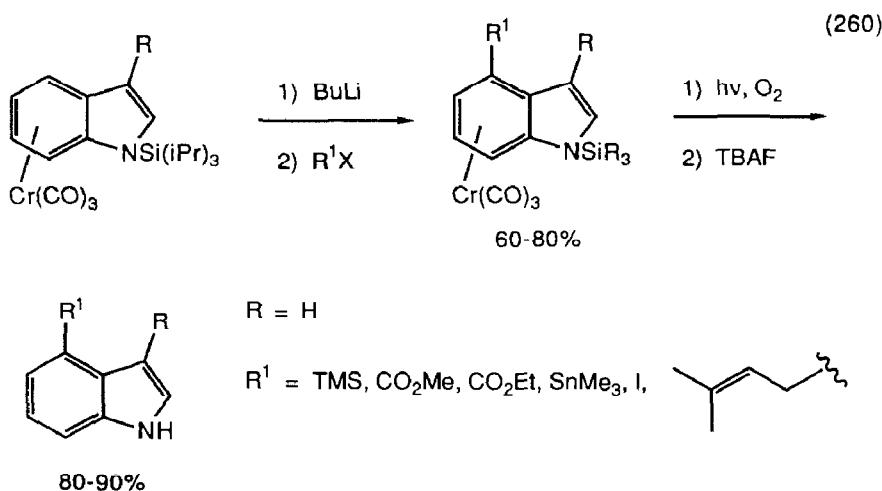


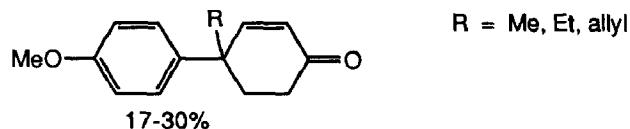
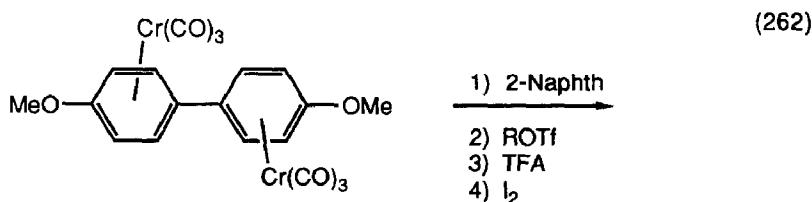
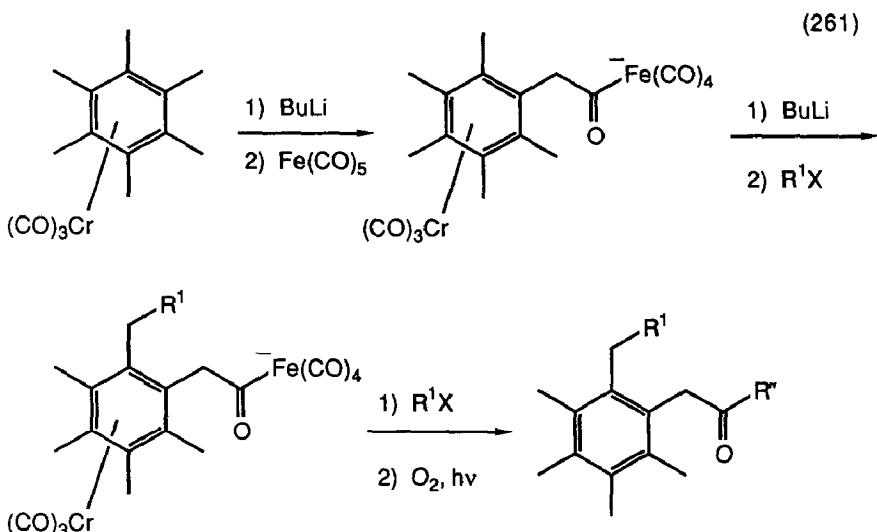
Chromium complexed fluorobenzene was alkylated with displacement of fluoride by stabilized carbanions (equation 258) [312]. Lithiated chromium tricarbonyl complexes of fluorobenzene were used to synthesize indolines (equation 259) [313]. Fluoroarenes were converted to aromatic ethers by nucleophilic substitution of fluoride by alkoxide on benzene rhodium(III) dicationic complexes [314].



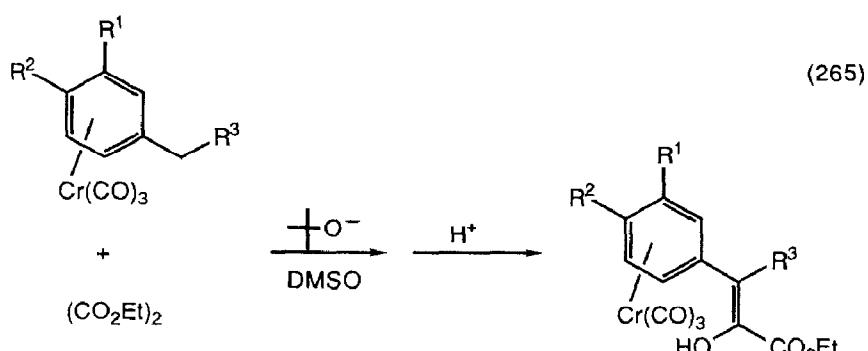
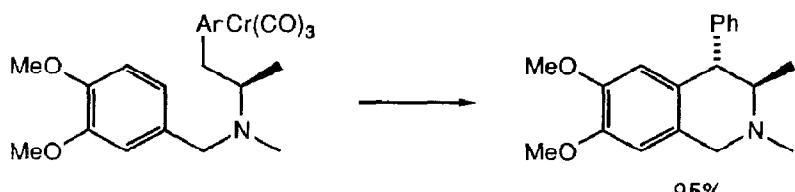
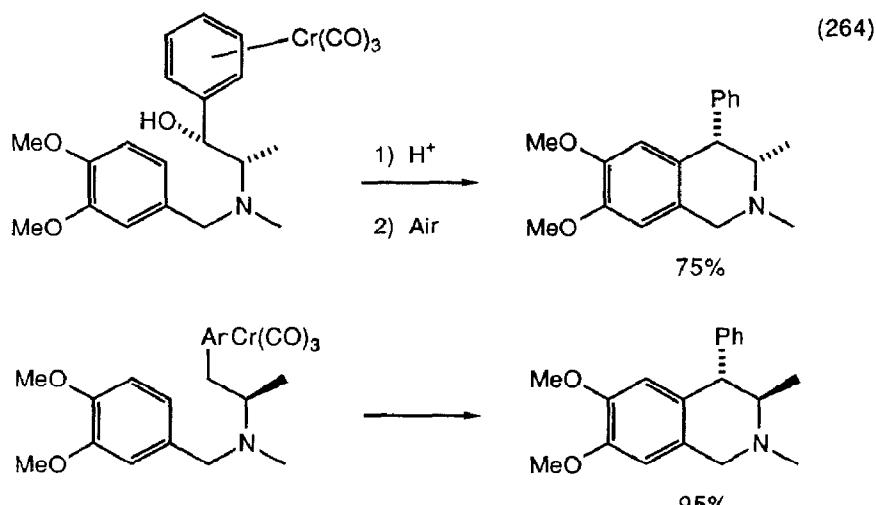
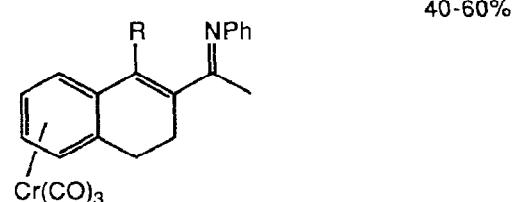
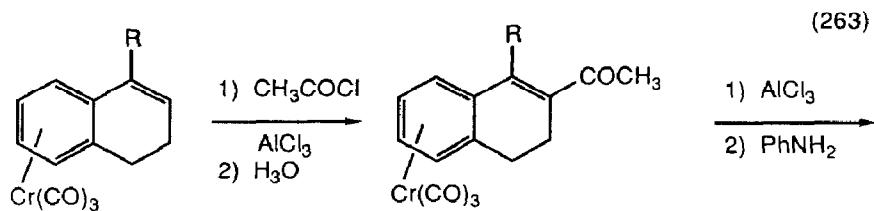


Chromium complexed indole was lithiated and functionalized in the 4-position (equation 260) [315]. Chromium-complexed hexamethyl benzene was benzylically lithiated and acylated using iron carbonyl chemistry (equation 261) [316]. The *bis*-chromium tricarbonyl complex of 4,4'-dimethoxybiphenyl was reduced and alkylated (equation 262) [317].



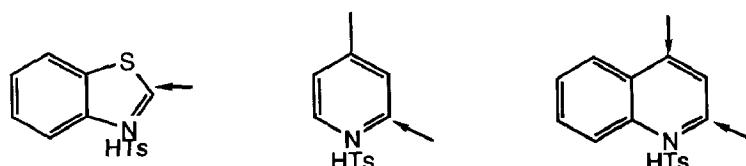
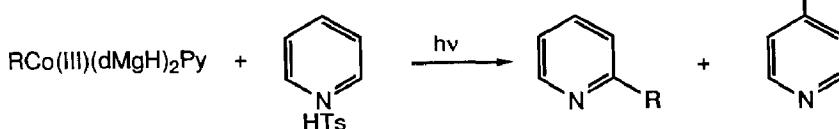
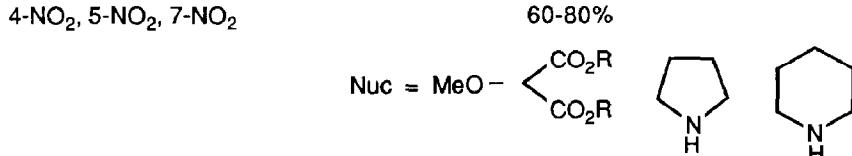
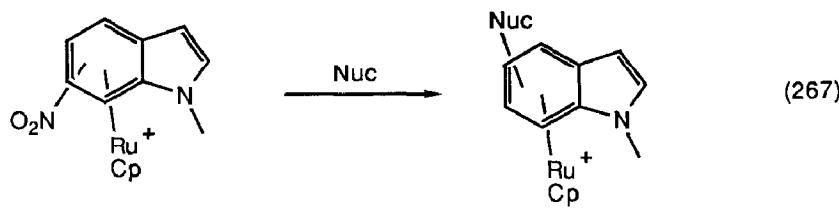
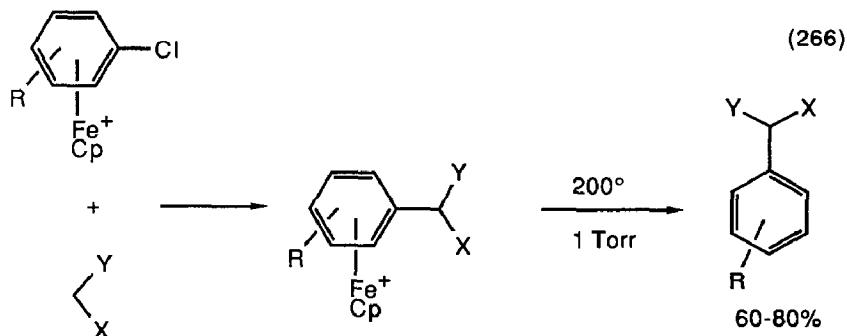


Chromium complexed tetralin underwent Friedel-Crafts acylation at the  $\beta$ -position (equation 263) [318]. Chromium tricarbonyl stabilized benzyl cations and permitted a cationic cyclization with a high degree of stereocontrol (equation 264) [319]. Arenechromium tricarbonyl complexes underwent facile alkylation at the benzylic position under base-catalyzed conditions (equation 265) [320].



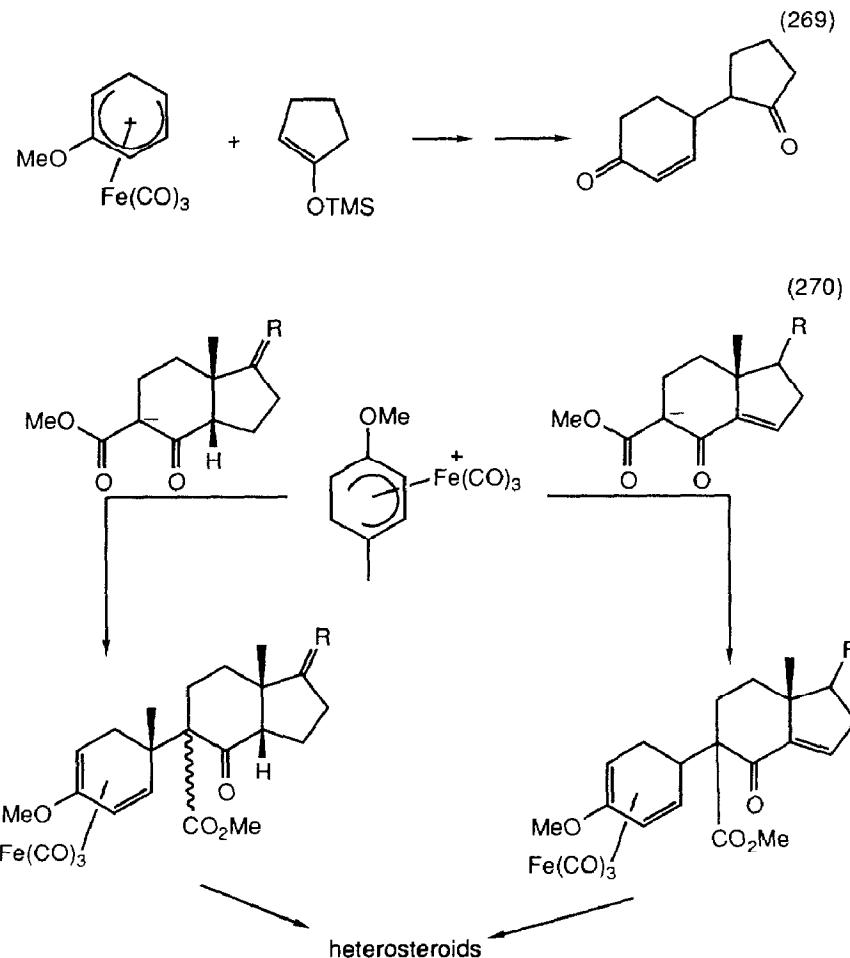
$\text{R}^1 = \text{H, Me}; \text{R}^2 = \text{H, tBu, OMe}; \text{R}^3 = \text{H, Me, Et}$

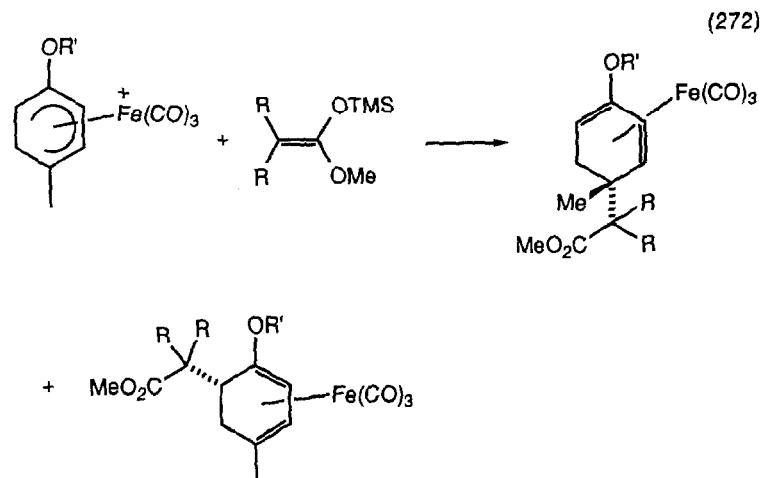
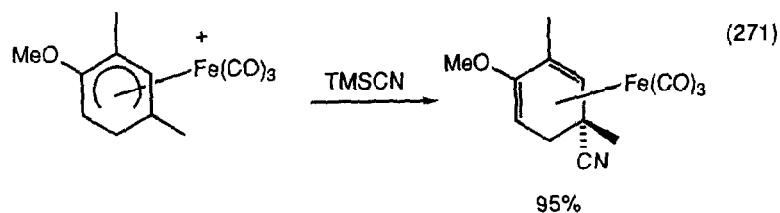
Chlorobenzene complexed to iron underwent displacement of chloride by stabilized anions (equation 266) [321]. Cationic ruthenium complexes of nitro indoles underwent nucleophilic displacement of the nitro groups (equation 267) [322] as did corresponding complexes of chloroindoles [323]. Tosic acid salts of pyridines and quinolines were alkylated by alkylcobalt DMG complexes under photolytic conditions (equation 268) [324].



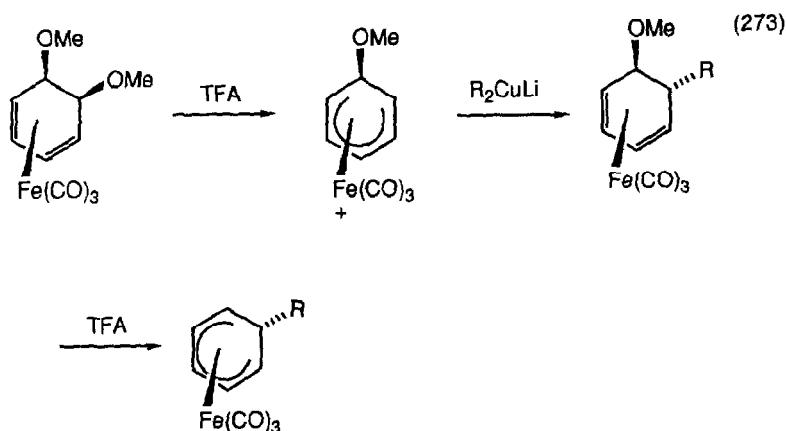
## 12. Alkylation of Dienyl and Diene Complexes

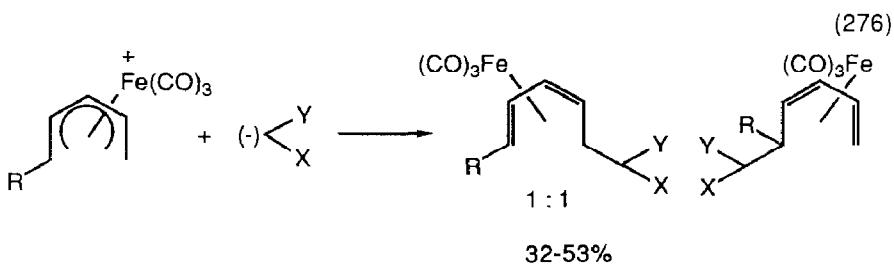
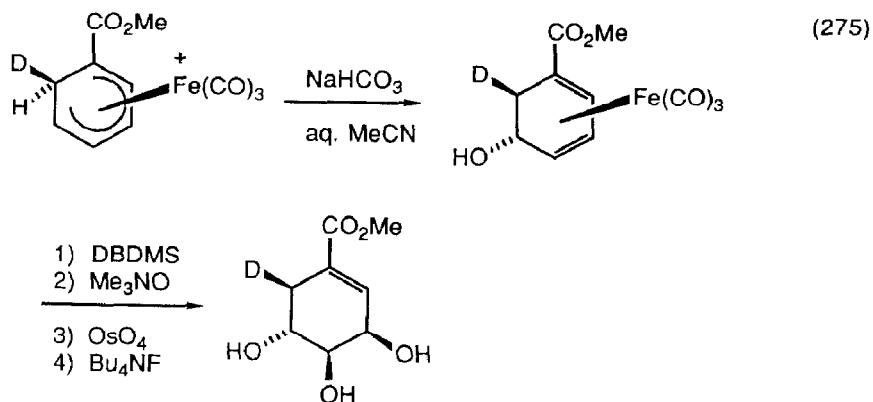
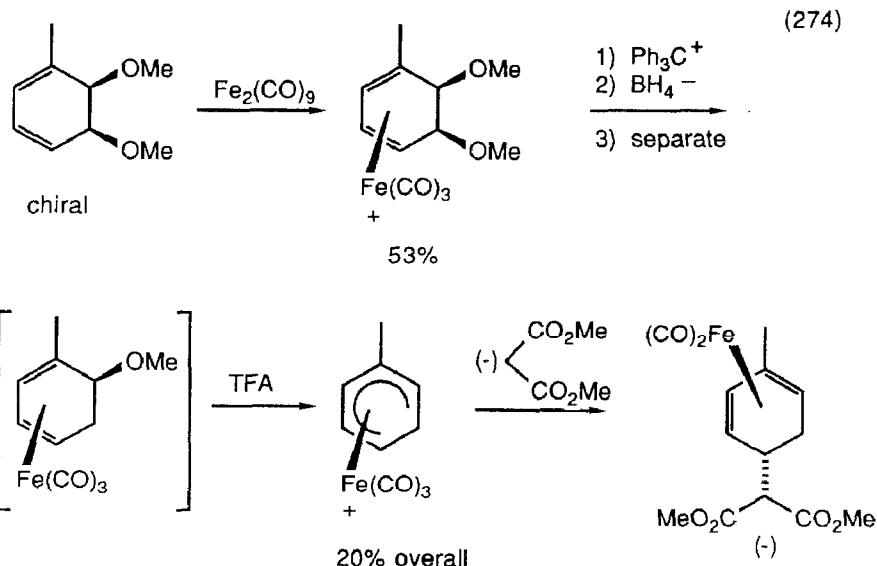
Alkylation of cationic iron dienyl complexes continues to be used to synthesize complex materials (equation 269) [325], (equation 270) [326], (equation 271) [327], (equation 272) [328]. Because nucleophilic attack always occurs from the face opposite the metal, the use of resolved chiral complexes results in enantiospecific processes (equation 273) [329], (equation 274) [330], (equation 275) [331]. Acyclic dienyl complexes of iron have also been alkylated (equation 276) [332]. Cationic molybdenum diene complexes were alkylated by a variety of carbanions to produce  $\pi$ -allylmolybdenum complexes (equation 277) [333]. Cationic cobalt diene complexes were cycloalkylated by *bistrimethylsilyl* enolates (equation 278) [334].

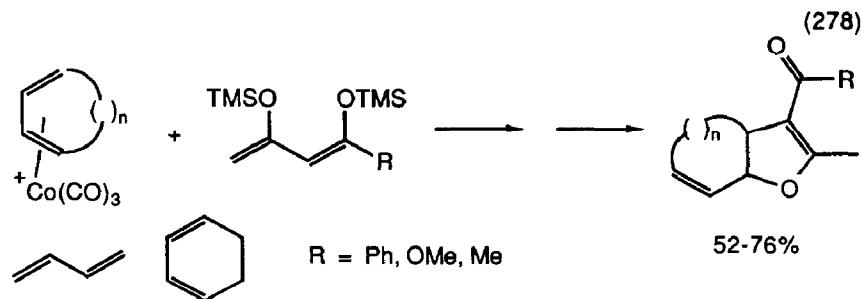
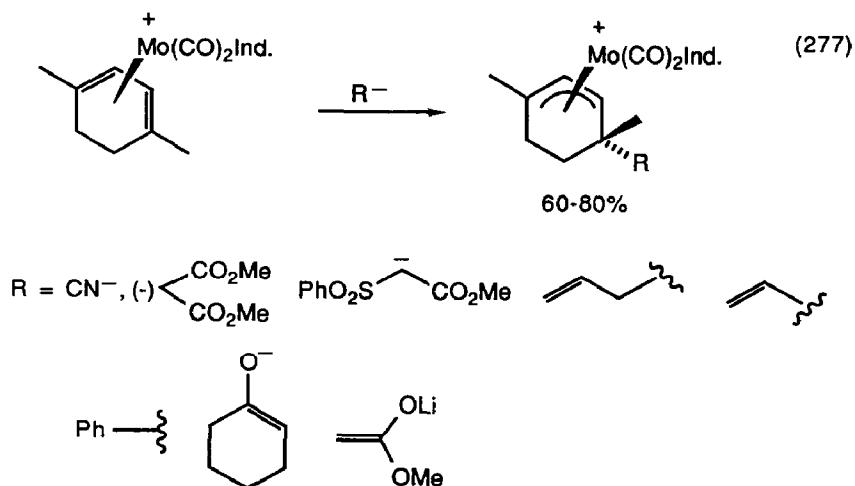




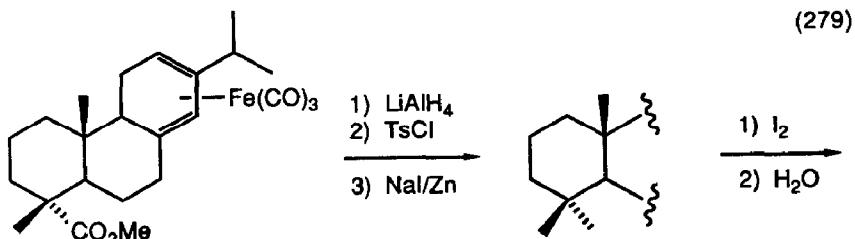
R, R' = Me<sub>2</sub>, Et-Ph, Ph-H, (CH<sub>2</sub>)<sub>4</sub>, (CH<sub>2</sub>)<sub>5</sub>, 60-90%; 10:1 to 100:1



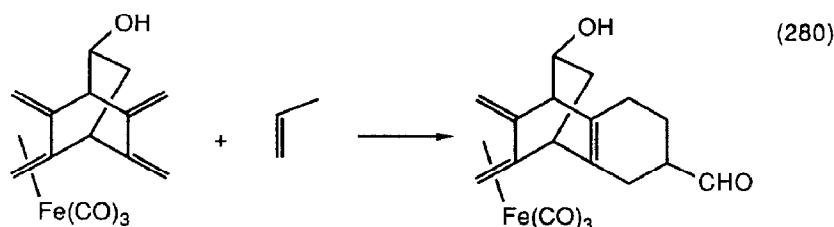
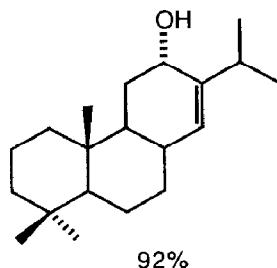




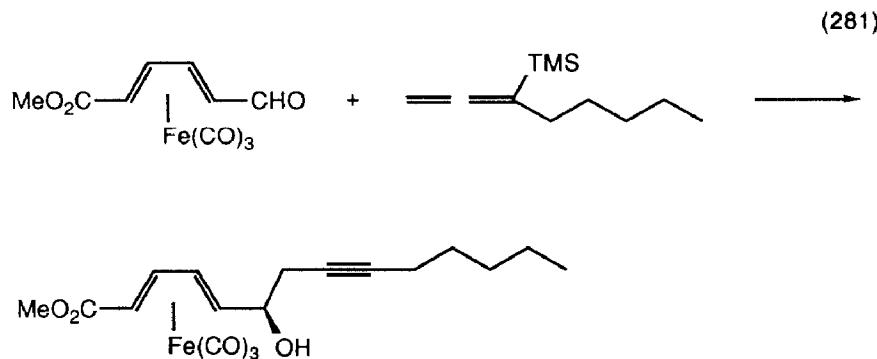
Dienes were protected from reduction (equation 279) [335] and from Diels-Alder reaction (equation 280) [336] by complexation to the iron tricarbonyl fragment.

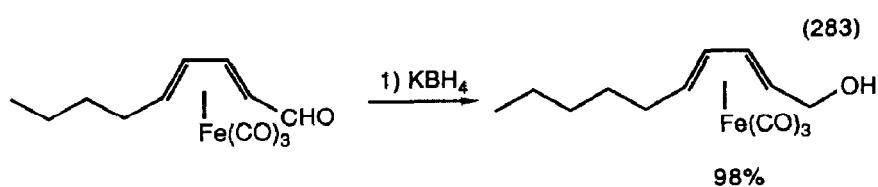
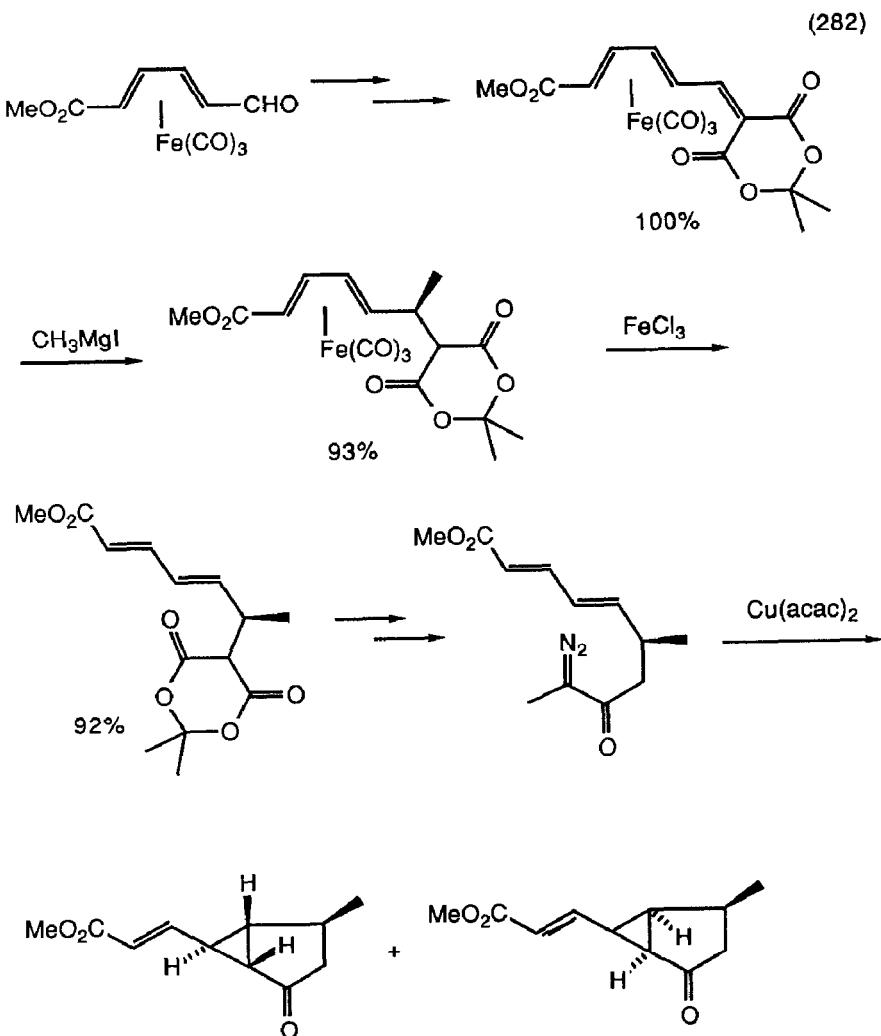


(equation 279 continued)

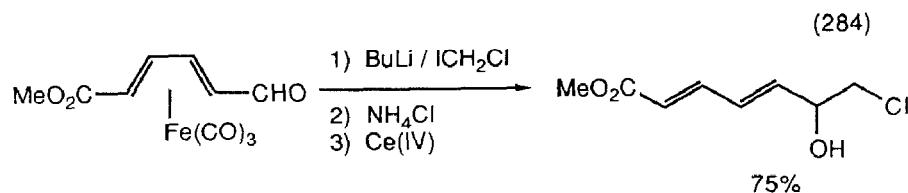
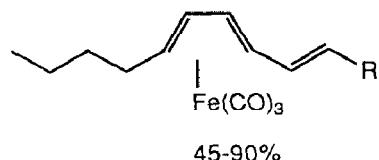
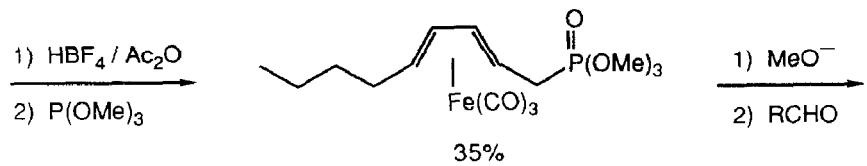


Diene aldehydes were protected from polymerization and other reactions while carrying out reactions at the aldehyde by coordinating the diene to iron tricarbonyl (equation 281) [337], (equation 282) [338], (equation 283) [339], (equation 284) [340].

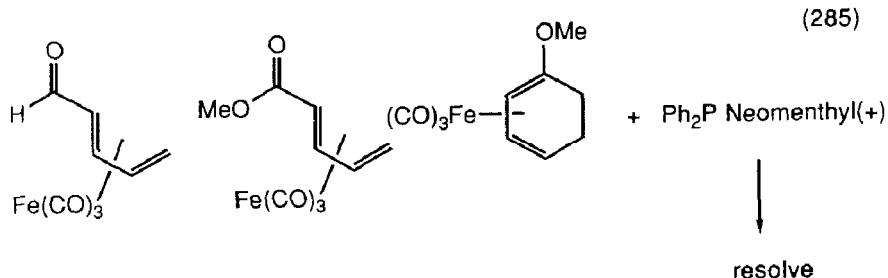


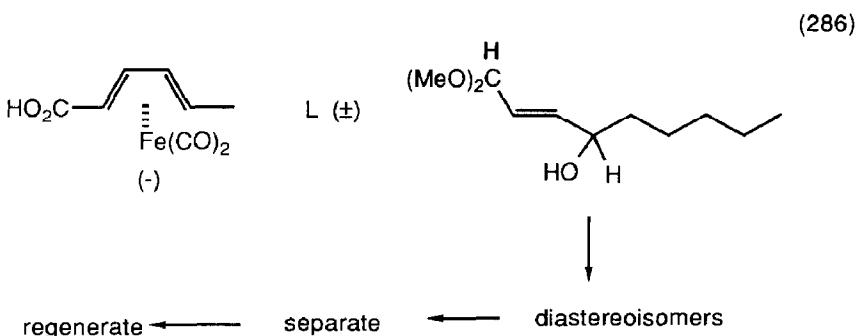


(equation 283 continued)



Iron diene complexes have been resolved by replacing CO with a chiral phosphine to generate diastereoisomers (equation 285) [341]. Allylic alcohols have been resolved using chiral iron diene complexes as the resolving agent (equation 286) [342].

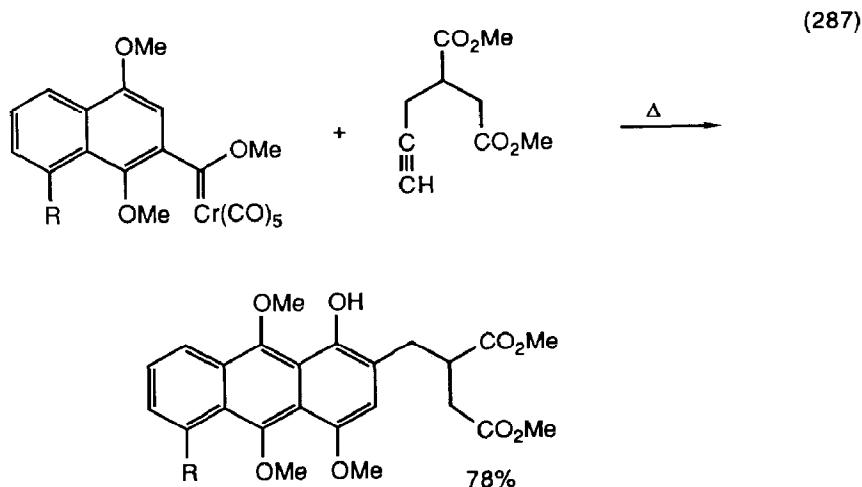


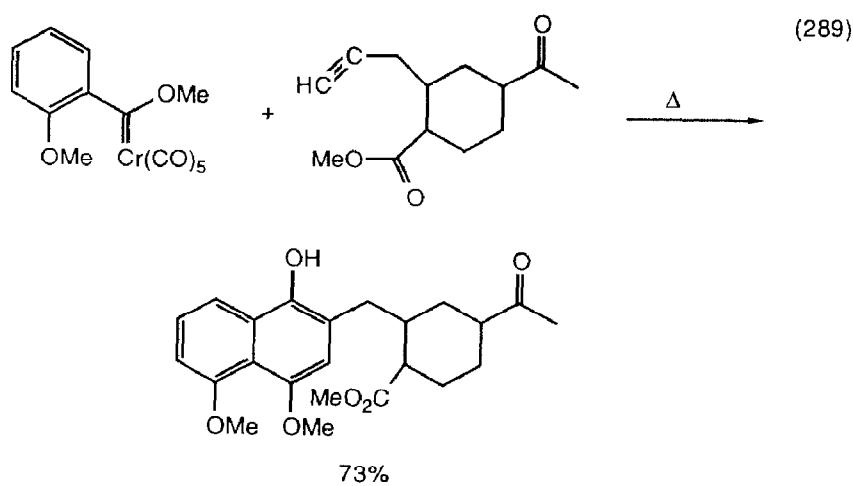
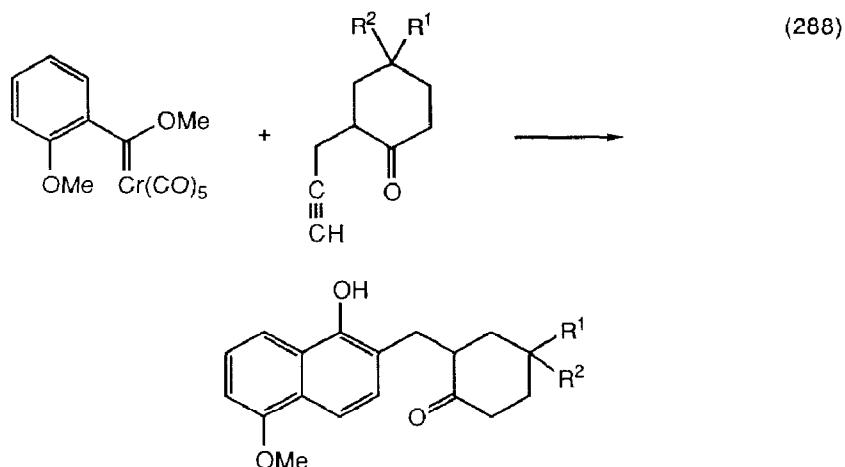


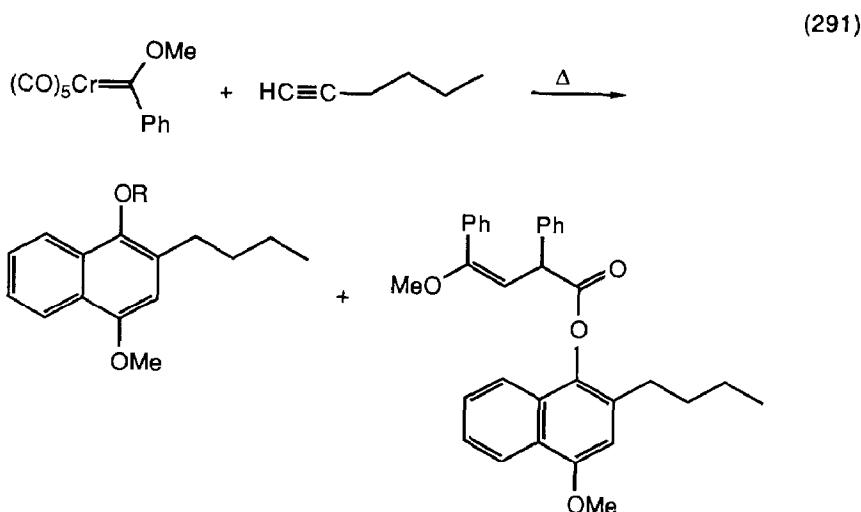
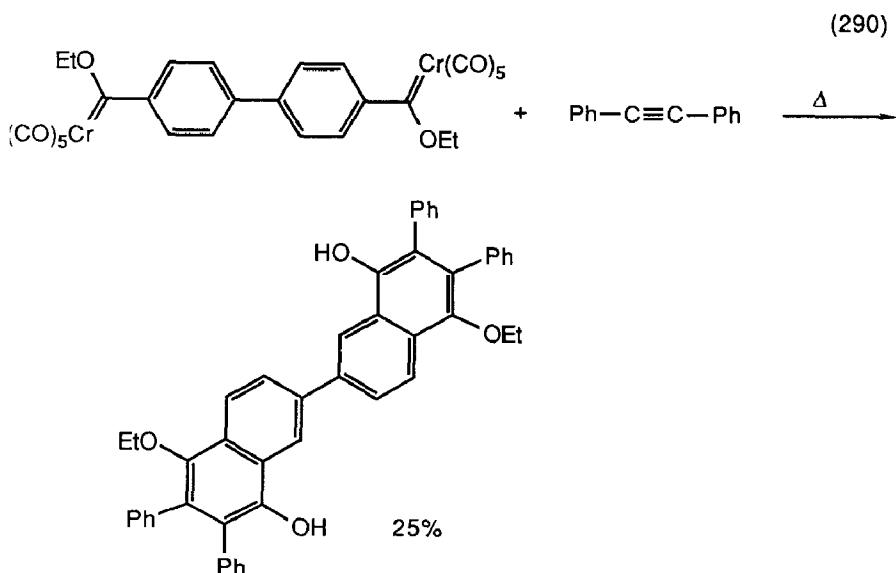
### 13. Metal-Carbene Reactions

Metal carbenes in cycloaddition reactions has been reviewed (40 references) [343]. Chromium carbene complexes in the synthesis of organic compounds was the topic of a dissertation [344] as was carbene-chromium complexes in the synthesis of nogalarol [345]. Recent aspects of transition metal catalyzed reactions of carbenes in the realm of biologically active substances have been reviewed (118 references) [346]. The text of lectures on coupling reactions of Fischer-type carbene complexes of iron [347], and regio- and stereoselectivity via transition metal carbene complexes [348] has appeared. An efficient synthesis of aminocarbene complexes of chromium from amides has appeared [349]. Synthetic routes to metal-alkylidene complexes have been reviewed (70 references) [350].

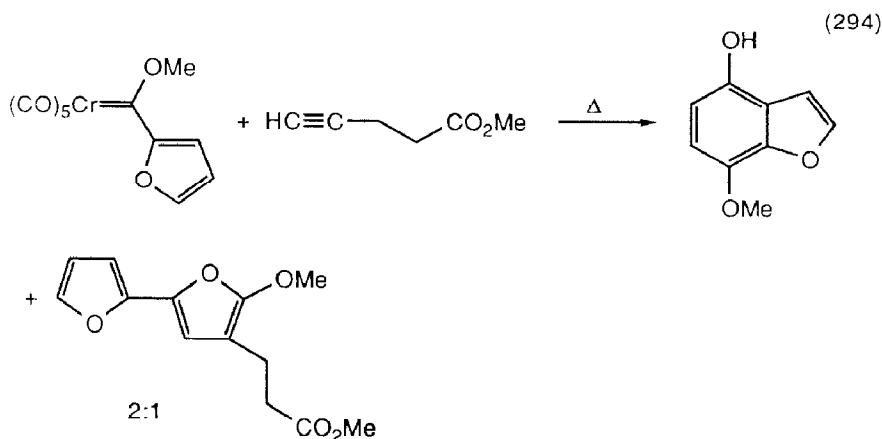
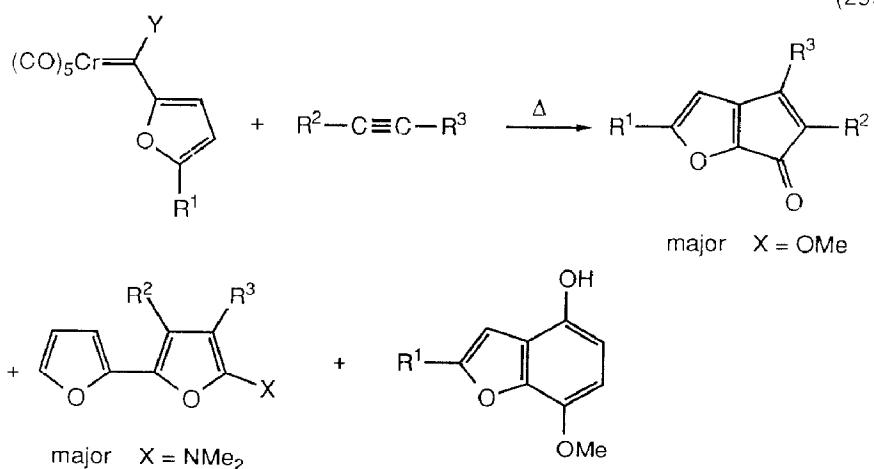
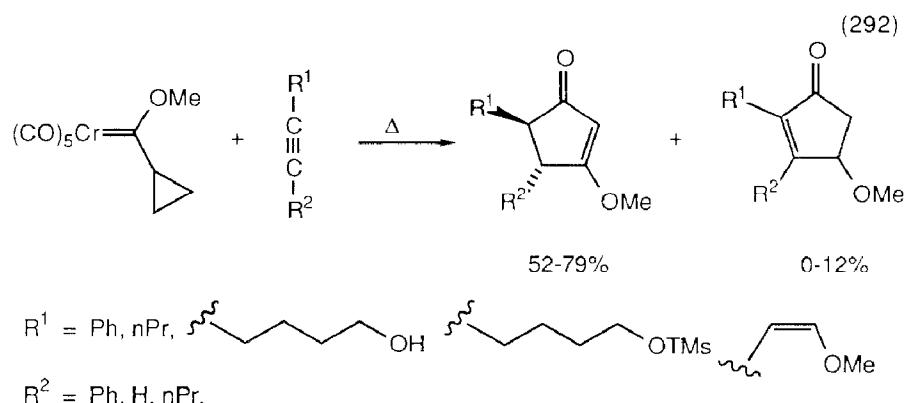
The thermal reactions of Group(VI) carbenes with alkynes continue to be exploited to synthesize polycyclic aromatic compounds (equation 287) (equation 288) [351], (equation 289) [352], (equation 290) [353], (equation 291) [354].

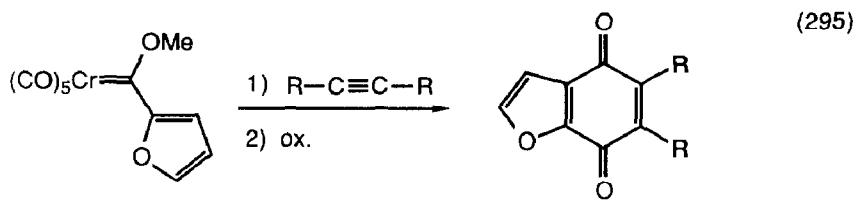




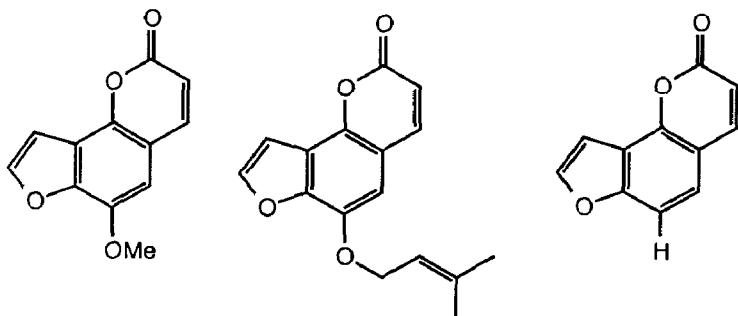


Cyclopropyl carbene complexes underwent reaction with alkynes to give cyclopentenones (equation 292) [355]. Furyl carbenes gave complex products depending on the heteroatom (equation 293) [356], (equation 294) [357]. They were used to synthesize a number of natural products (equation 295) [358].

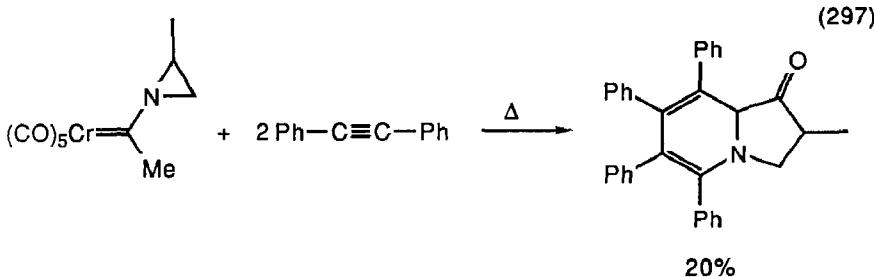
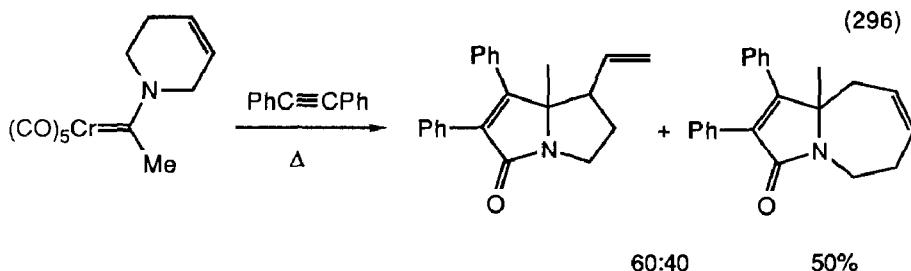


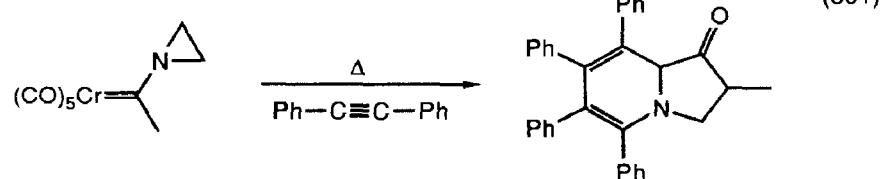
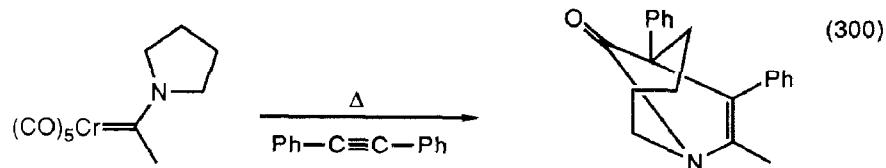
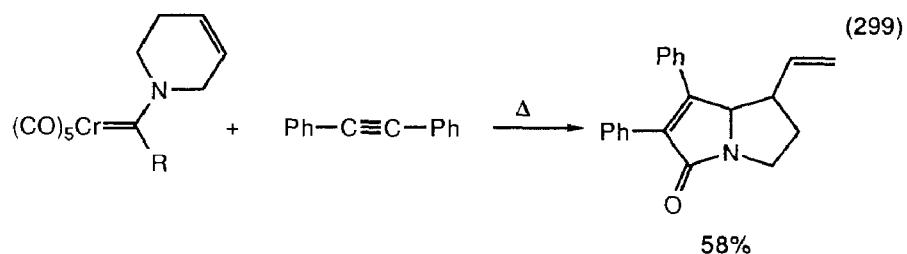
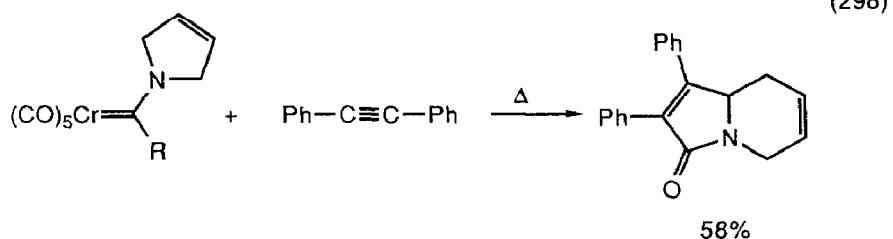


used to synthesize:



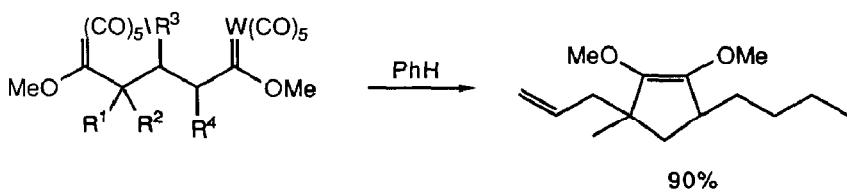
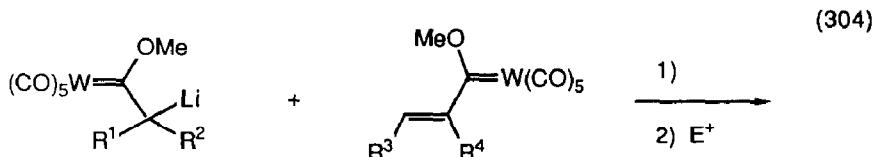
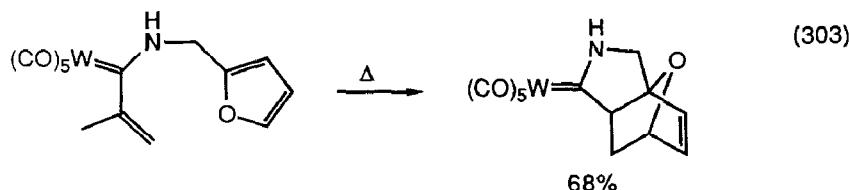
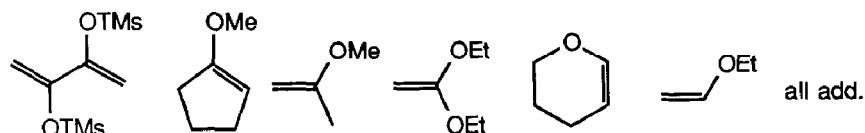
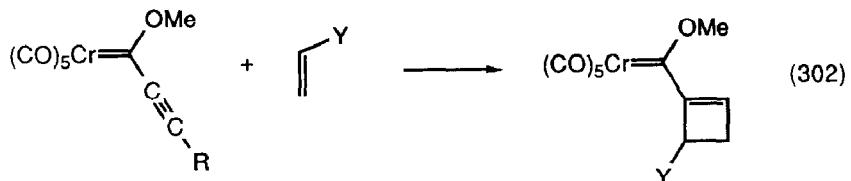
Aminocarbene complexes underwent profound rearrangements in their reactions with alkynes (equation 296) [359], (equation 297) [360], (equations 298-301) [361].

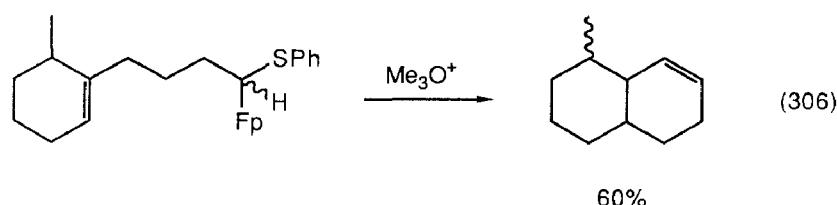
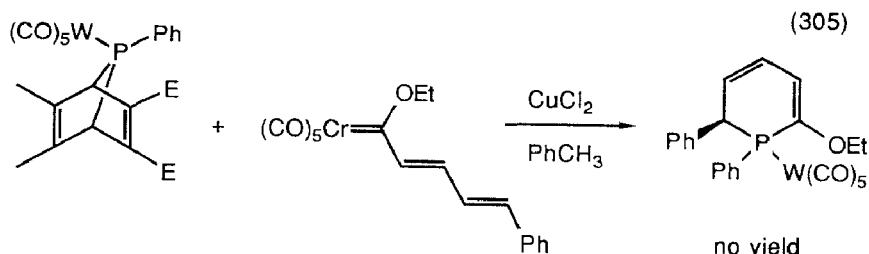




Acetylenic carbenes underwent 2+2 cycloaddition reactions at the alkyne with electron-rich olefins (equation 302) [362], while unsaturated tungsten carbenes underwent intramolecular 4+2 cycloaddition (equation 303) [363]. Cyclic bis enol ethers were prepared by the Michael addition of a carbene anion to an  $\alpha,\beta$ -unsaturated carbene to give an  $\alpha,\omega$ -bis carbene, which was coupled by thermal decomposition (equation 304) [364]. Two other unusual carbene reactions are shown in equation 305 [365], and equation 306 [366].

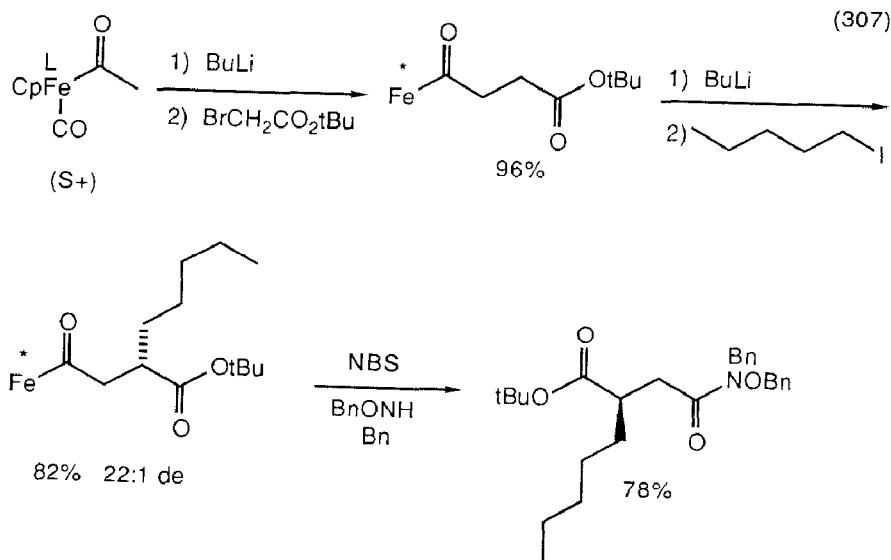
Diastereoselective reactions of chiral acetylides at iron carbene complexes have been studied [367].



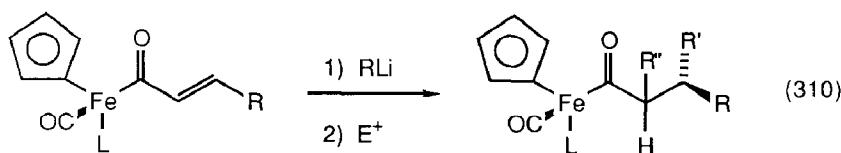
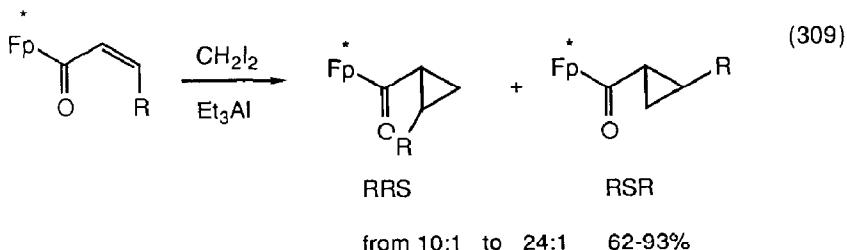
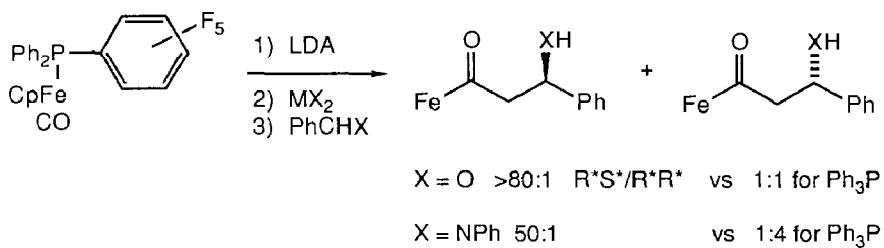


#### 14. Alkylation of Metal Enolates

Asymmetric synthesis via the iron chiral auxiliary  $\eta^5\text{-C}_5\text{H}_5\text{Fe}(\text{CO})\text{PPh}_3$  has been reviewed (34 references) [368], as has confirmational analysis of transition metal  $\eta^1\text{-acyl}$  complexes (61 references) [369]. The contents of a lecture on formation of carbon-carbon bonds using organotransition metal enolates have appeared [370]. Chiral acyliron enolates continue to be used in asymmetric synthesis (equation 307) [371], (equation 308) [372], (equation 309) [373], (equation 310) [374], (equation 311) [375].

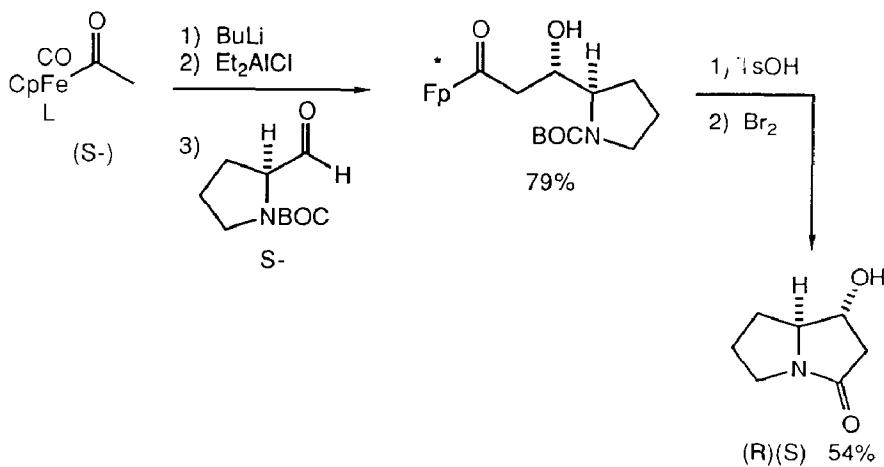


(308)



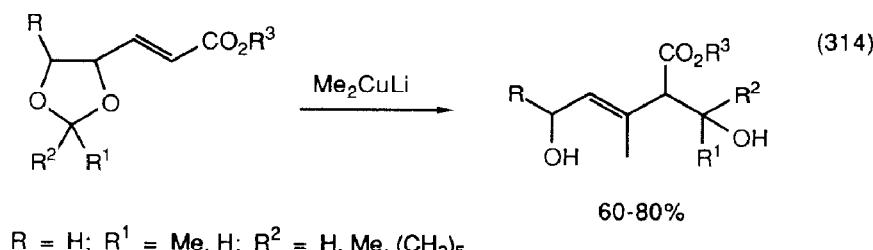
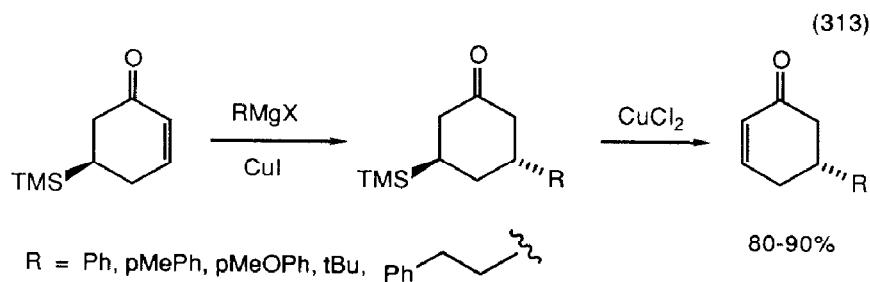
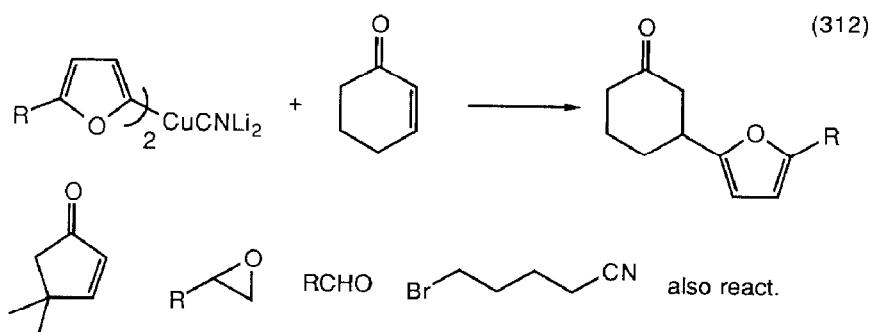
Studied effect of L, R, R', E, R" on yield + de.

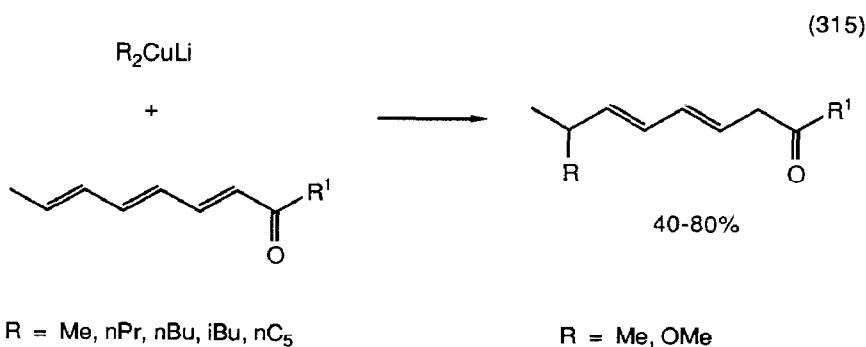
(311)



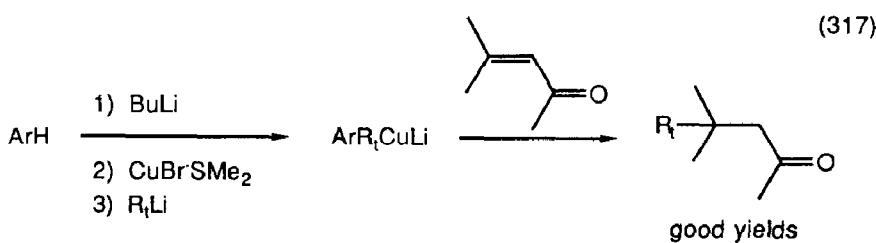
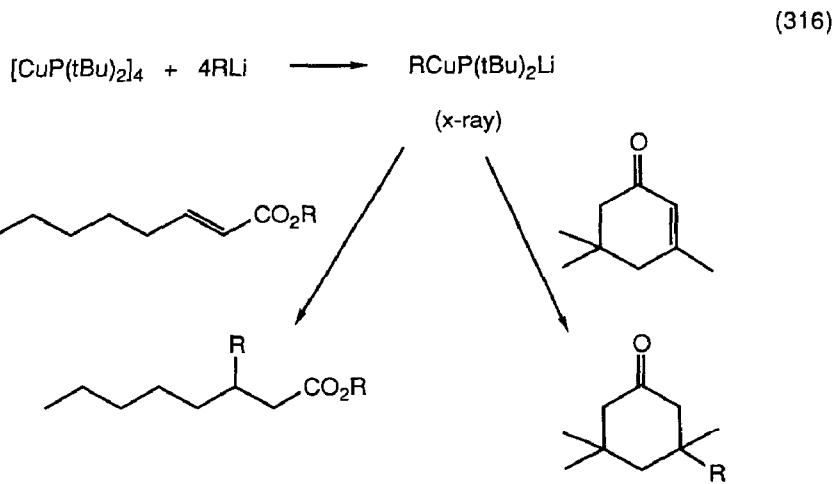
### B. Conjugate Addition.

Organocuprate additions to  $\alpha,\beta$ -unsaturated compounds have been reviewed (32 references) [376]. The contents of a lecture dealing with intramolecularly assisted addition of lithium dimethylcuprate to  $\alpha,\beta$ -unsaturated esters have appeared [377].  $\alpha$ -Furylcuprates added efficiently to conjugated enones (equation 312) [378]. Copper catalyzed addition of  $\beta'$ -trimethylsilyl- $\alpha,\beta$ -unsaturated enones produced alkylated enones (equation 313) [379]. Methyl cuprates added with rearrangement to  $\alpha,\beta$ -unsaturated ester-diol ketals (equation 314) [380], and added 1,8 to trienones (equation 315) [381].

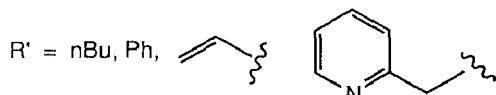
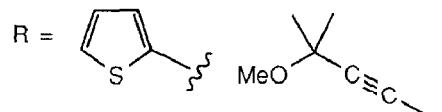
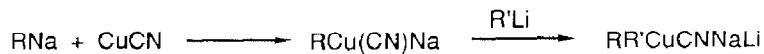
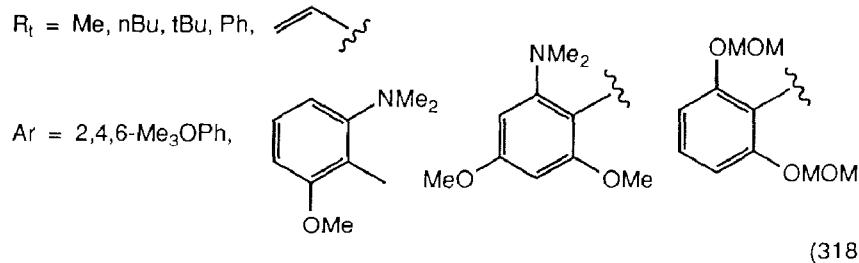




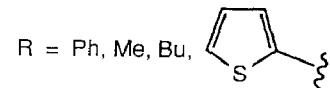
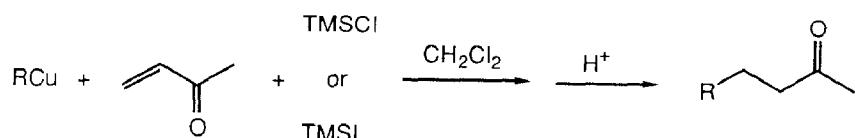
New types of organocopper species continue to be developed. Copper phosphide reagents were stable at reflux in THF for hours, yet underwent 1,4-addition reactions efficiently (equation 316) [382]. Oxygenated arenes were developed as nontransferrable R groups for organocuprates (equation 317) [383]. The reagent  $RR'CuCNNaLi$ , was less reactive to 1,4-addition and more reactive to 1,2-addition to aldehydes (equation 318) [384]. Organocopper reagents  $RCu$  added efficiently in a 1,4 manner to conjugated enones in the presence of trimethylsilyl chloride or iodide (equation 319) [385].



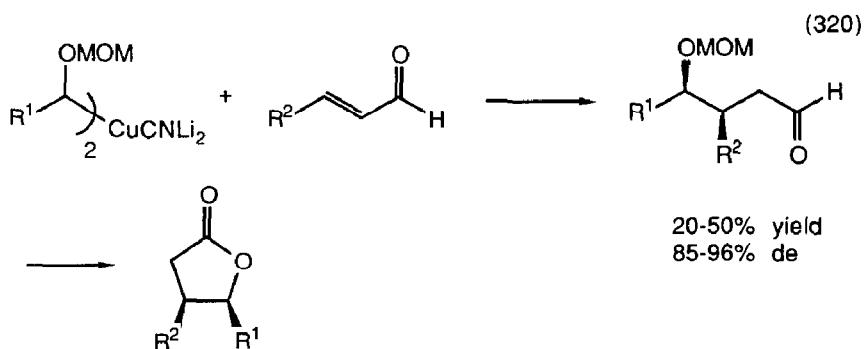
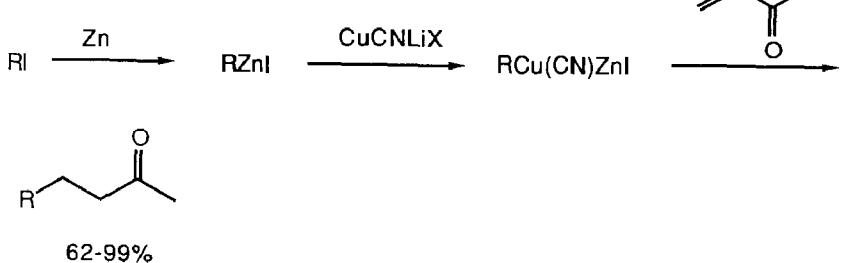
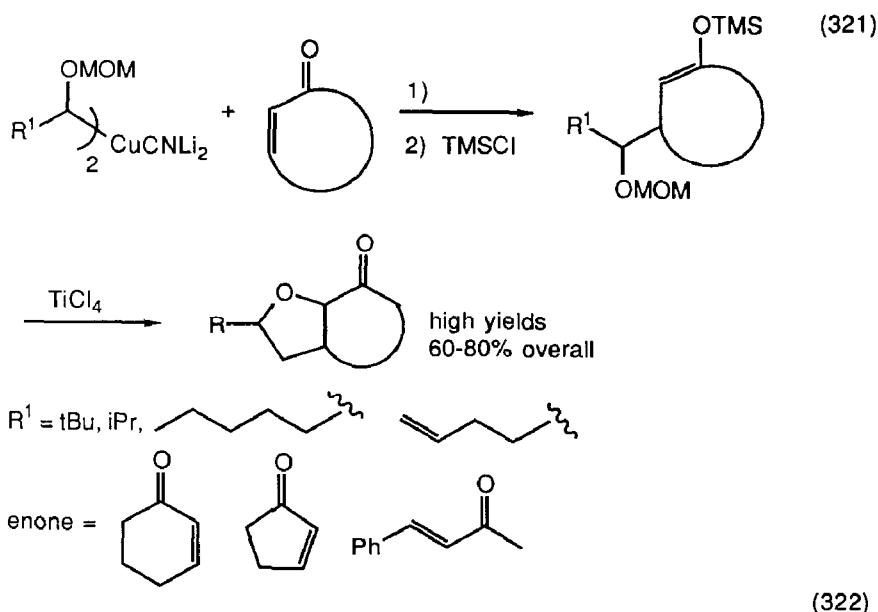
(equation 317 continued)



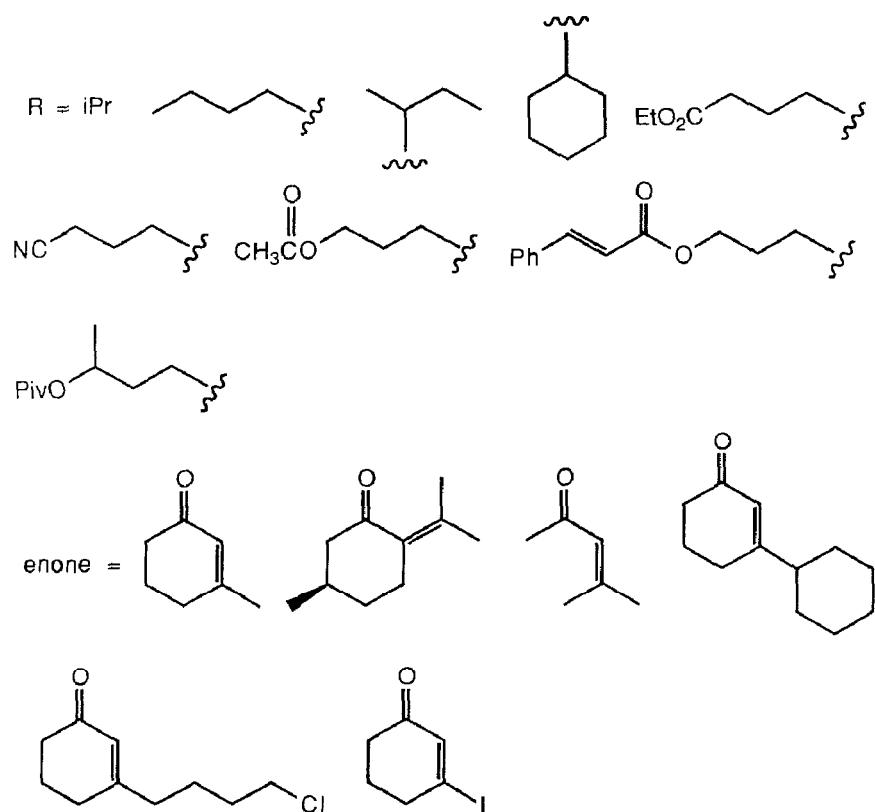
(319)



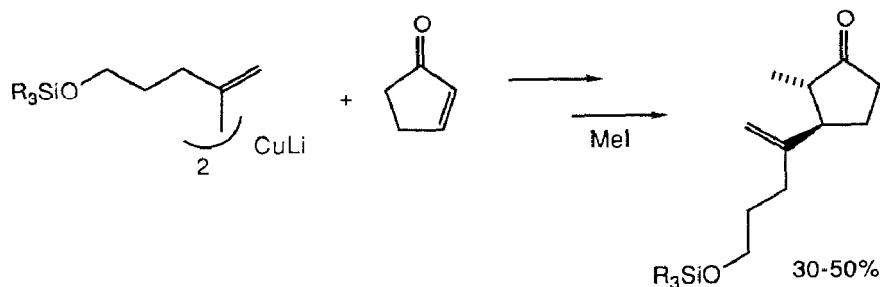
A number of functionalized organocupper reagents have been developed. These include  $\alpha$ -methoxymethyl reagents (equation 320) [386], (equation 321) [387], chloro, keto, and cyano containing reagents (equation 322) [388][389], trimethylsilyl reagents (equation 323) [390], (equation 324) [391], trimethylstannyl reagents (equation 325) [392], and trimethylsilylimine reagents (equation 326) [393]. Perfluoromethyl-acetylenic ketones underwent 1,4-alkylation by cyanocuprates (equation 327) [394].

 $\text{R}^1 = \text{iPr, nBu}$  $\text{R}^2 = \text{nBu, Ph, } \text{---CH}_2\text{CH}_2\text{---}$ 

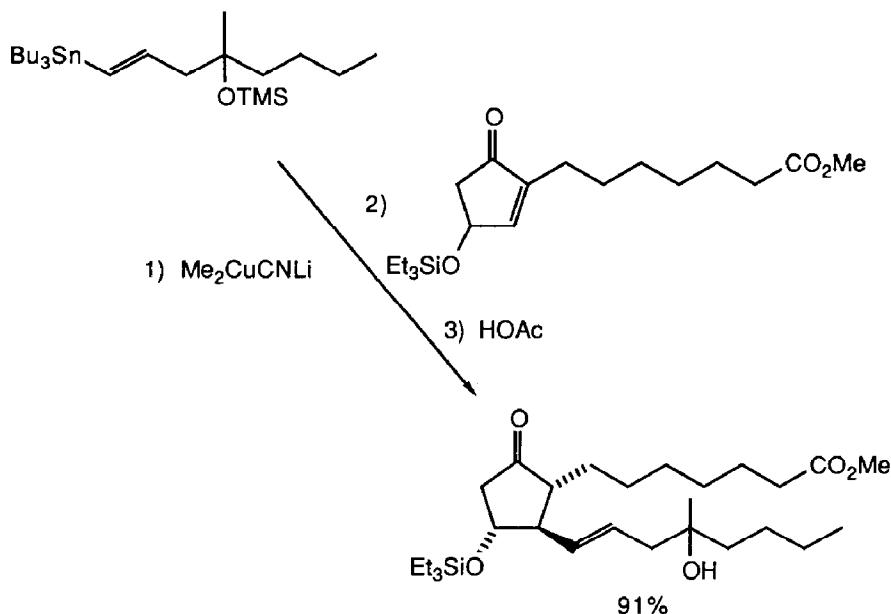
(equation 322 continued)



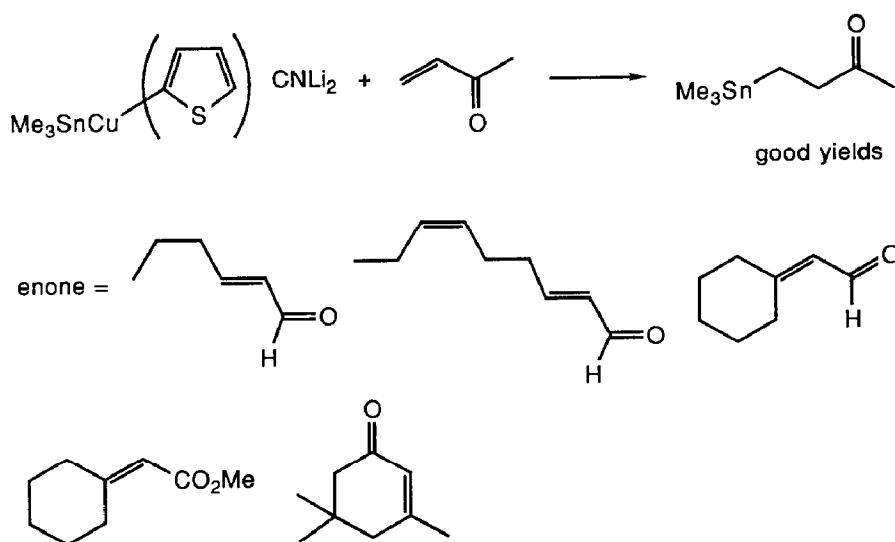
(323)



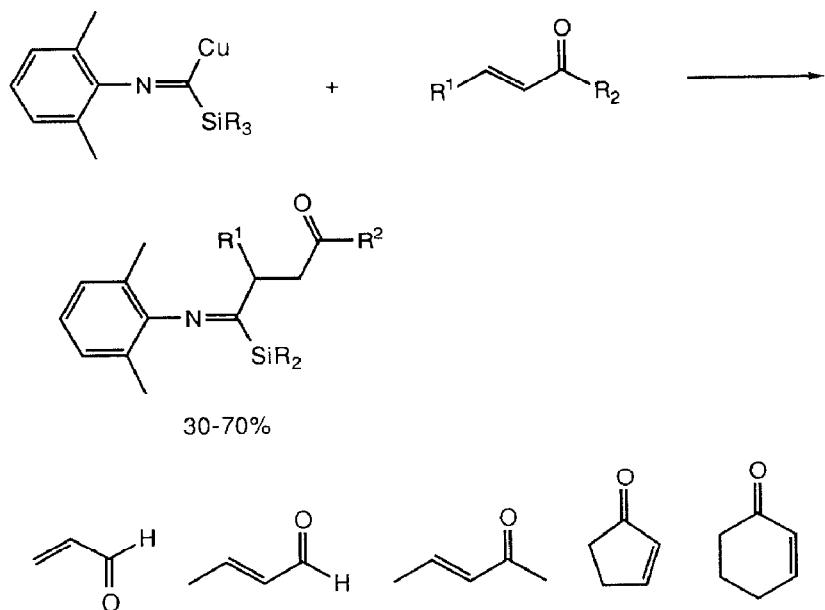
(324)



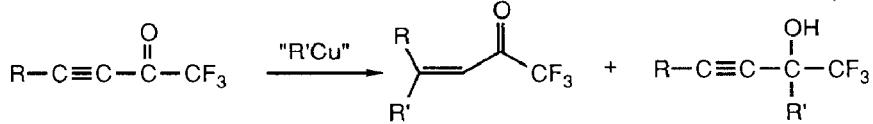
(325)



(326)

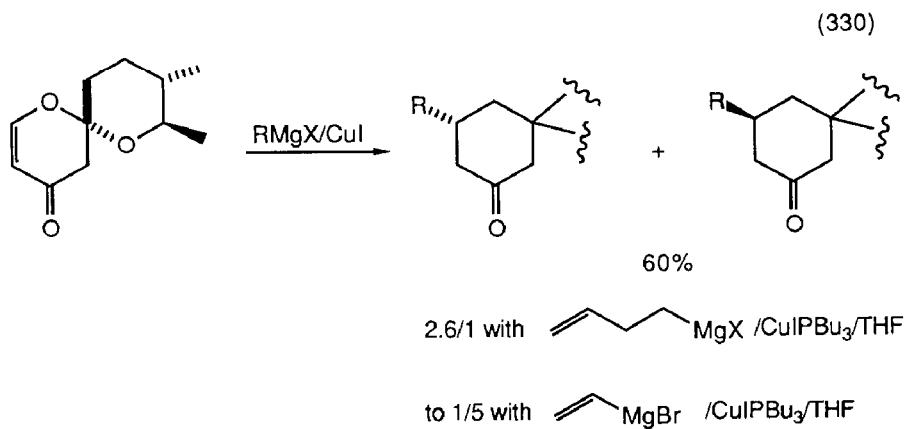
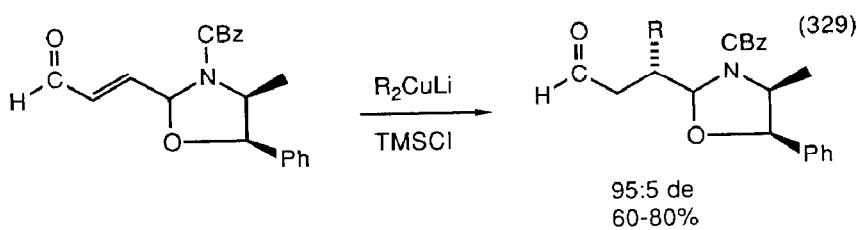
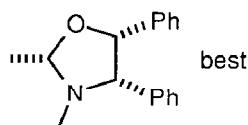
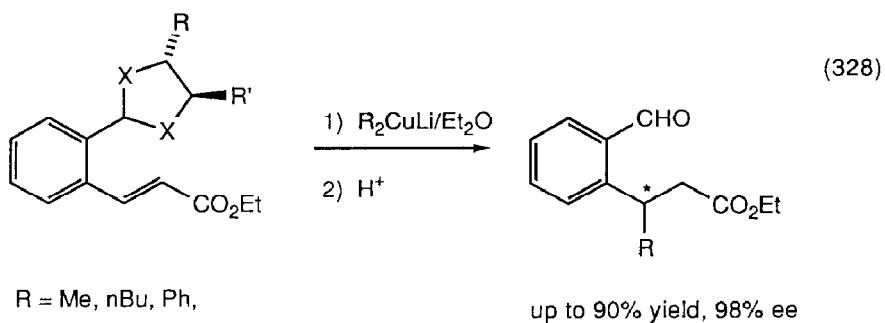


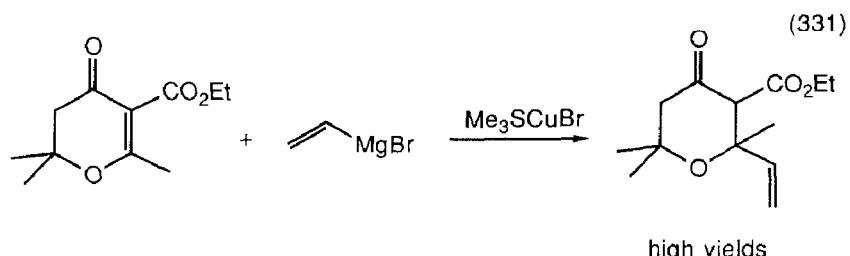
(327)



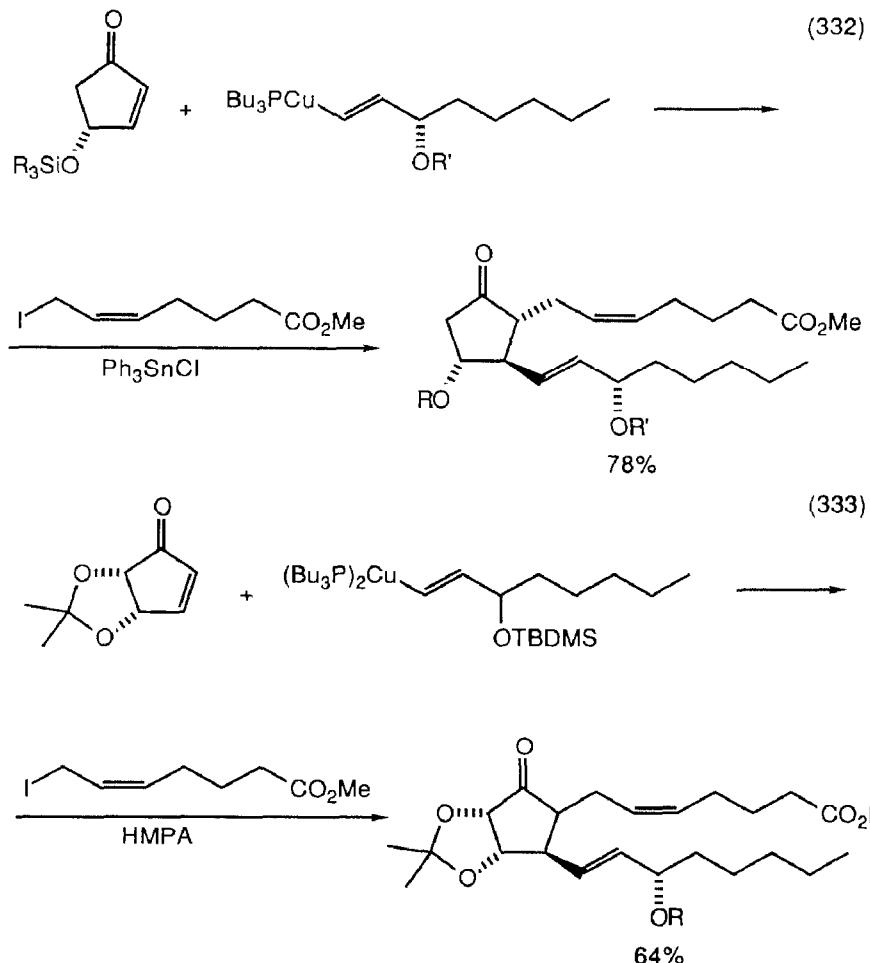
Me<sub>2</sub>CuCNLi<sub>2</sub> tBuCuCNLi add 100%, 1,4 in 65-80% yield

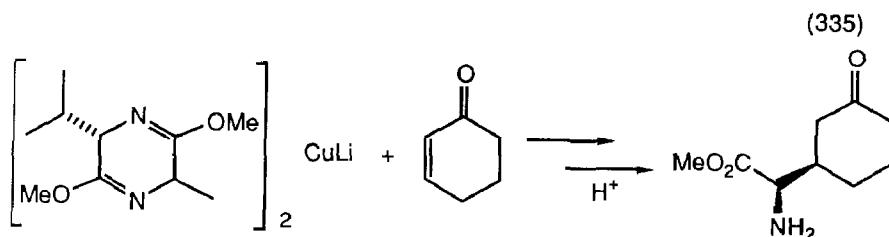
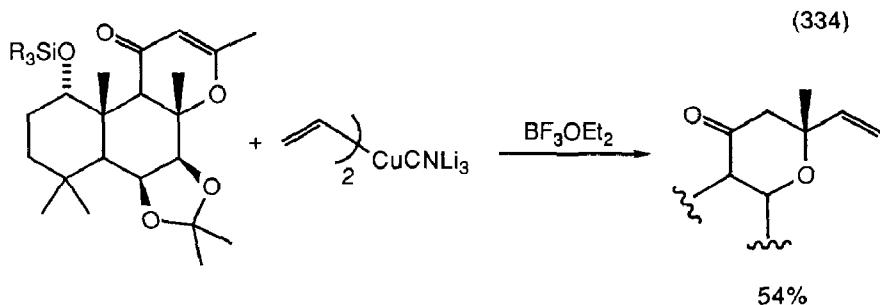
Chiral auxiliaries were used to efficiently induce asymmetry into conjugate additions of enones (equation 328) [395], (equation 329) [396], (equation 330) [397].  $\gamma$ -Pyrones underwent both 1,2- and 1,4-alkylations by cuprates (equation 331) [398].



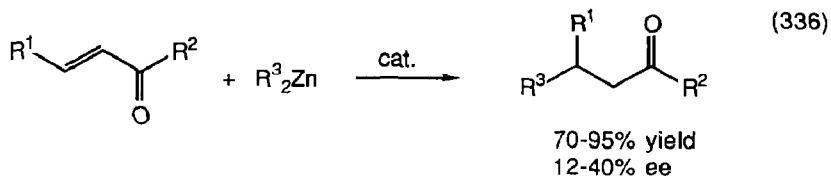


Copper assisted 1,4-additions have been used in the synthesis of prostaglandins (equation 332) [399], (equation 333) [400], ( $\pm$ )-forskolin (equation 334) [401] and unnatural  $\alpha$ -amino acids (equation 335) [402].

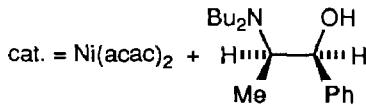




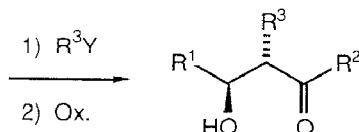
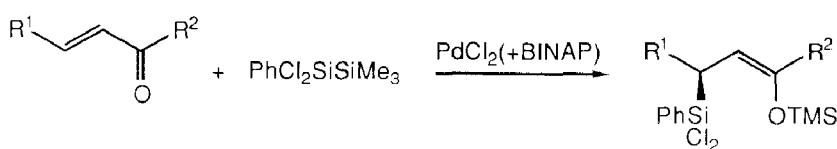
Chiral nickel complexes catalyzed the asymmetric 1,4-alkylation of enones by organozinc reagents (equation 336) [403], while chiral palladium complexes catalyzed the 1,4-silylation of enones (equation 337) [404]. Other odd conjugate additions are seen in equation 338 [405], equation 339 [406], equation 340 [407], and equation 341 [408].



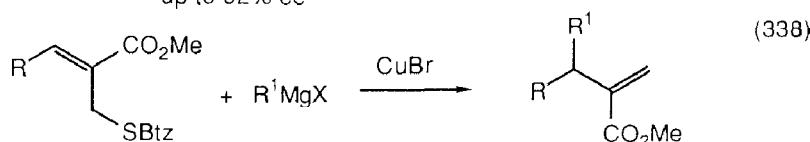
$R^1 = Ph, Me; R^2 = Ph, Me; R^3 = Me, Et$



(337)



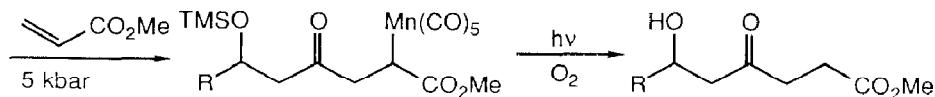
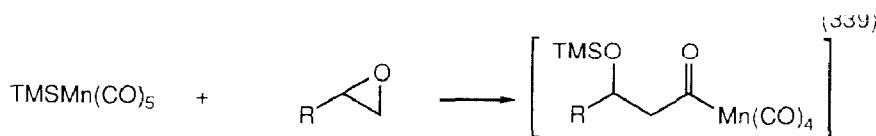
up to 92% ee



85-95%

$\text{R} = \text{Me, iPr, CO}_2\text{Et, } \text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_2\text{S}^\bullet$

$\text{R}' = \text{nBu, TMSCH}_2, \text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_2\text{S}^\bullet$

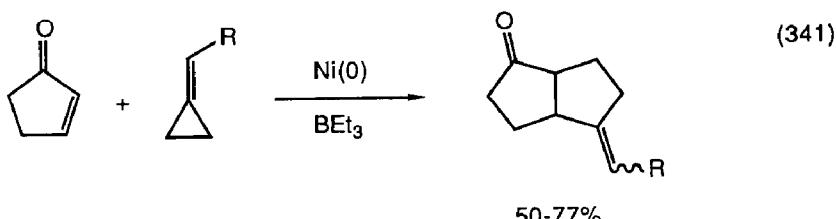
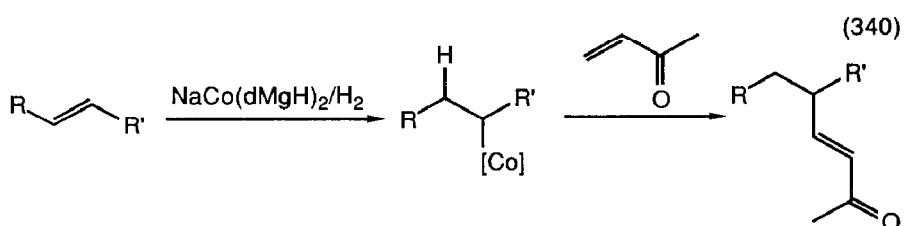


good yields

$\text{R} = \text{Me, BzO } \text{S}^\bullet$



$\equiv\text{C}-\text{Ph} \quad \equiv\text{C}-\text{CO}_2\text{Me}$

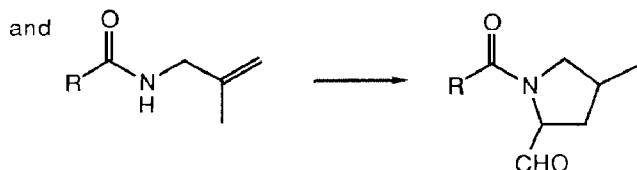
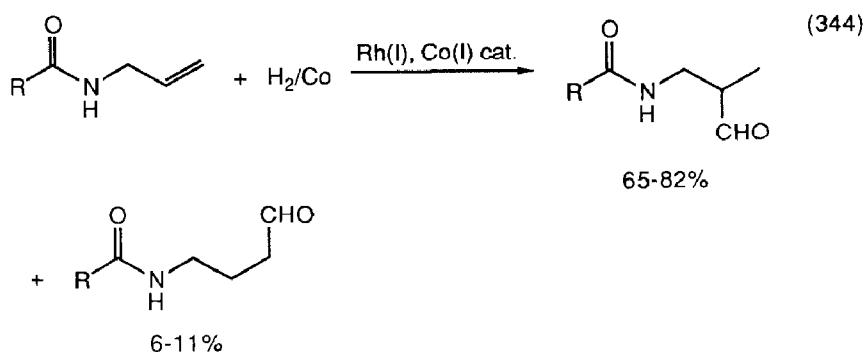
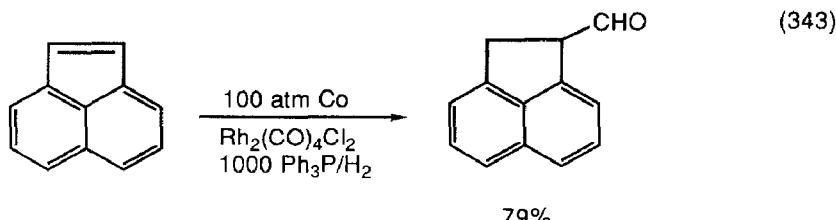
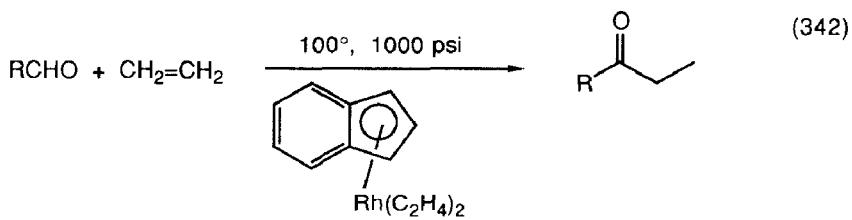


### C Acylation Reactions (excluding Hydroformylation)

The following reviews dealing with carbonylation have appeared: "Metal catalyzed carbonylation and oxidation-reduction reactions" (26 references) [409]; "Metal Carbonyls" (152 references) [410]; "Carbonylation"; (55 references) [411]; "Transition metals in organic synthesis: hydroformylation, reduction and oxidation. Annual survey covering the year 1987" (673 references) [412]; "Metal carbonyl catalysts for the synthesis of organic compounds from CO and molecular H<sub>2</sub>" (165 references) [413]; "New aspects of carbonylation catalyzed by transition metal complexes" (52 references) [414]; "Substituted carbonylation of organic compounds catalyzed by palladium complexes" (141 references) [415]; "Organic reactions catalyzed by electrogenerated organometallic species. Activation of hydrocarbons and carbon monoxide and related reactions" (10 references) [416]; "Carbonylation reaction under atmospheric pressure with copper carbonyl catalysts. Modified Koch reaction" (25 references) [417]; "Carbonylation with octacarbonylcobalt and pentacarbonyliron in phase-transfer catalysis" (48 references) [418]; "Silylcobalt carbonyl and new catalytic reactions" (33 references) [419].

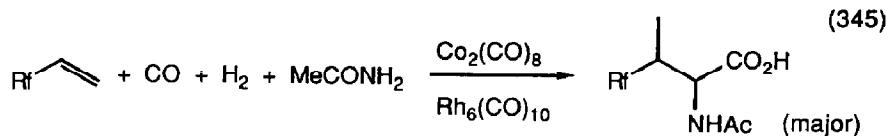
#### 1. Carbonylation of Alkenes and Alkynes

Indene rhodium(I) ethylene complexes catalyzed the addition of aldehydes to ethylene to give ethyl ketones (equation 342) [420]. Fluorene was efficiently acylated by carbon monoxide/hydrogen and rhodium(I) catalysts (equation 343) [421]. N-Allyl amides were hydroformylated with chelation control of the regiochemistry (equation 344) [422].

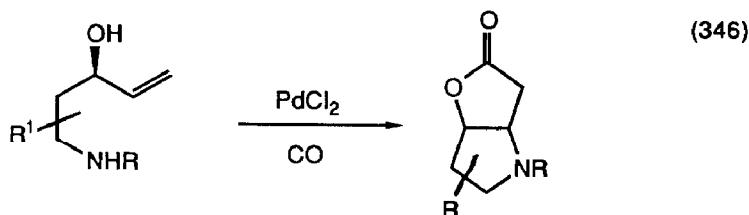
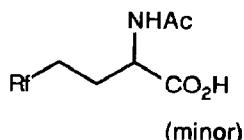


Terminal alkenes ranging from ethylene through octadecene were efficiently hydrocarboxylated by palladium catalysts, and carbon monoxide [423]. The acyl complex intermediate in this process was isolated [424]. Olefins were converted to esters by addition of methyl formate in the presence of  $\text{RuH}_2\text{L}_4$  [425].

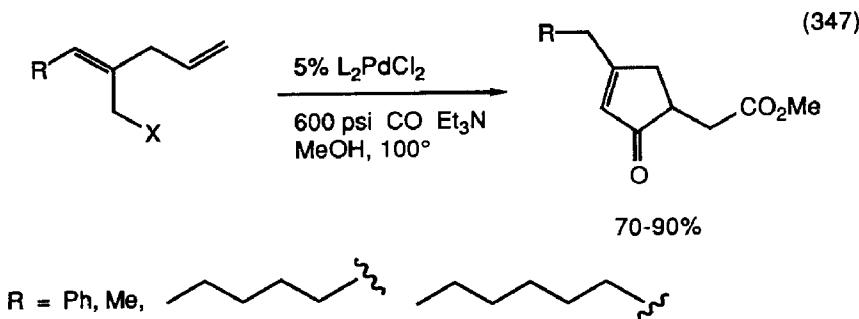
Cobalt and rhodium complexes catalyzed the amidocarbonylation of olefins (equation 345) [426], a topic that has been reviewed (43 references) [427]. Palladium(II) salts catalyzed the aminoacylation (equation 346) [428] and the *bis* carbonylation (equation 347) [429] of olefinic substrates, as well as the acetoxy carbonylation (equation 348) [430].



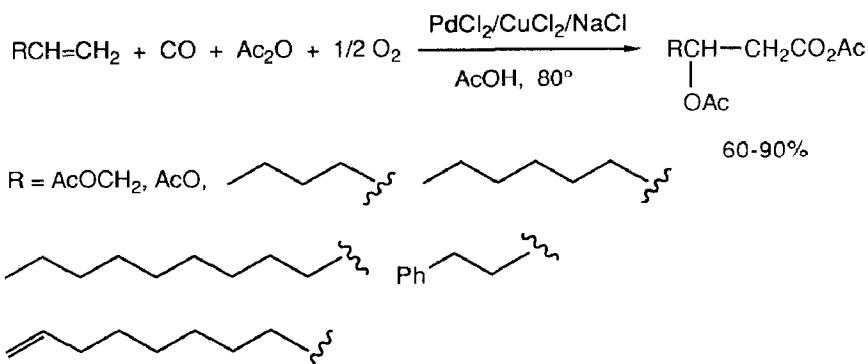
+



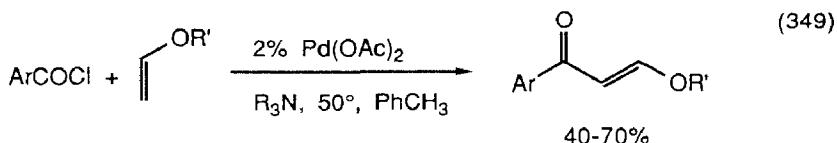
(effects of R, solvent, conditions)



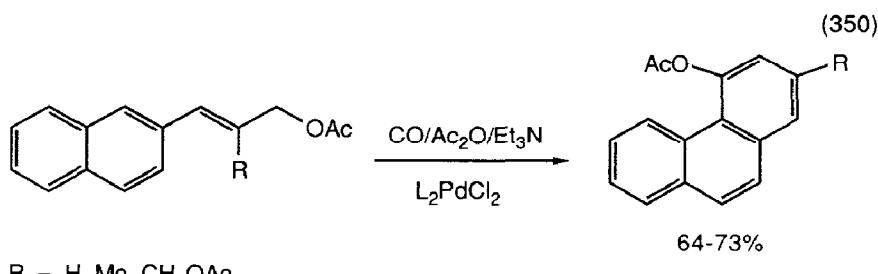
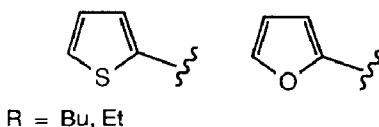
(348)

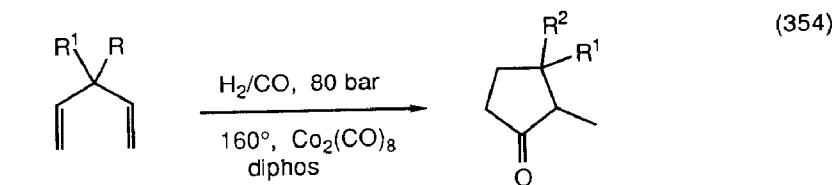
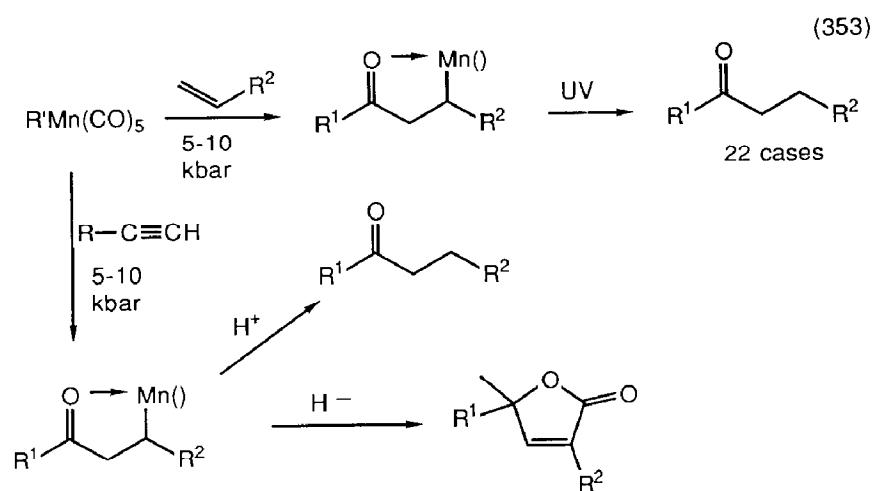
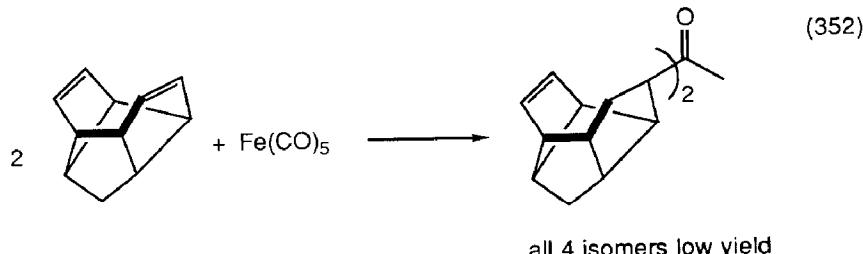
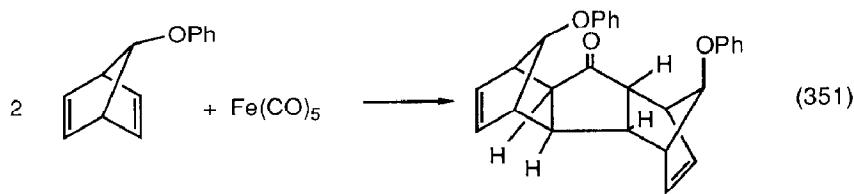


Palladium complexes catalyzed the acylation of olefins by acid chlorides (equation 349) [431] and the cyclocarbonylation of aryl compounds with allylic side chains (equation 350) [432]. Iron carbonyl formed dimer ketones from strained olefins (equation 351) [433], (equation 352) [434]. Alkylmanganese carbonyls acylated olefins and alkynes under high pressure (equation 353) [435]. Cobalt and rhodium(I) complexes catalyzed the cyclocarbonylation of dienes (equation 354) [436].



$\text{Ar} = \text{Ph, mMeOPh, pClPh, pBrPh, pNO}_2\text{Ph, 3,4,5-Me}_3\text{O}_3\text{Ph, 2-Naphth,}$



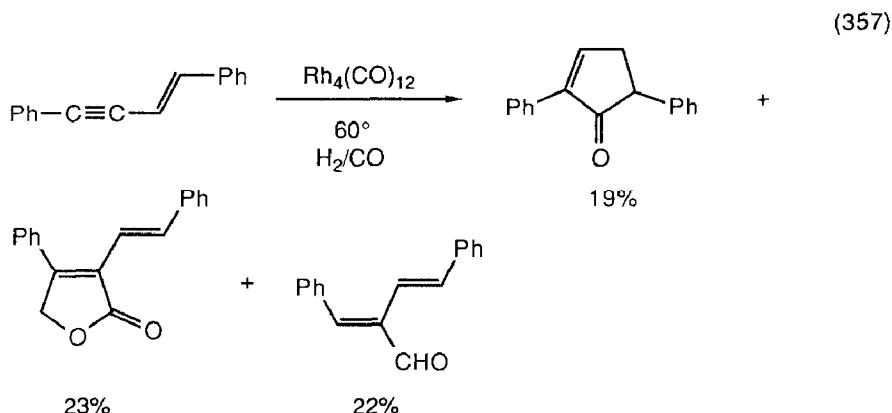
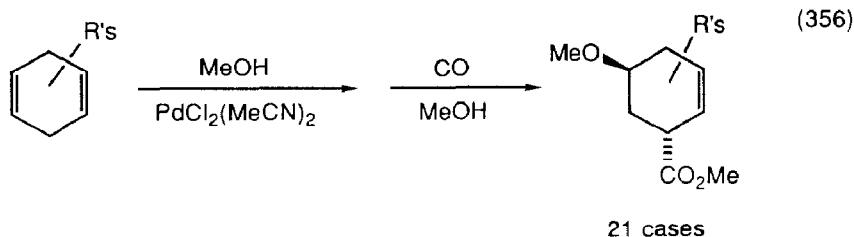
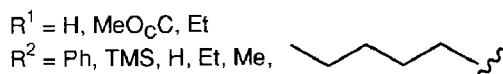
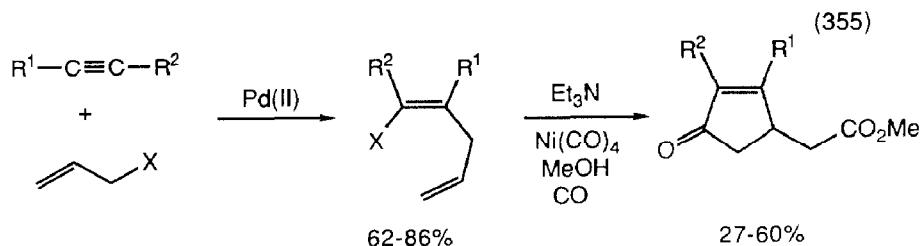


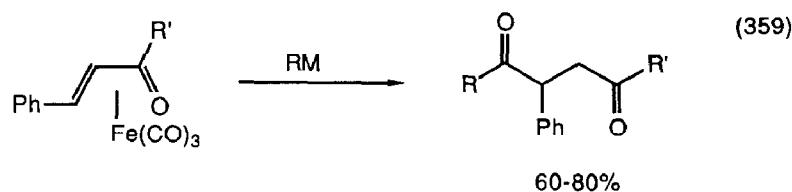
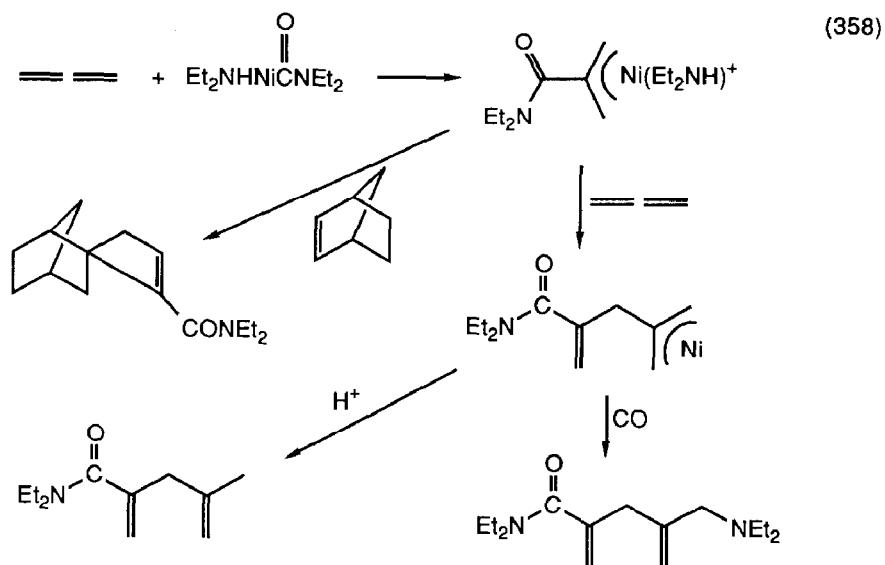
$\text{R}^1 = \text{Me}, \text{R}^2 = \text{ptol}$

80-90%

$\text{R}^1, \text{R}^2 = (\text{CH}_2)_5$

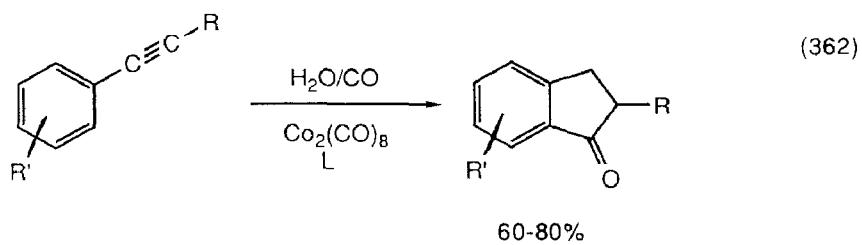
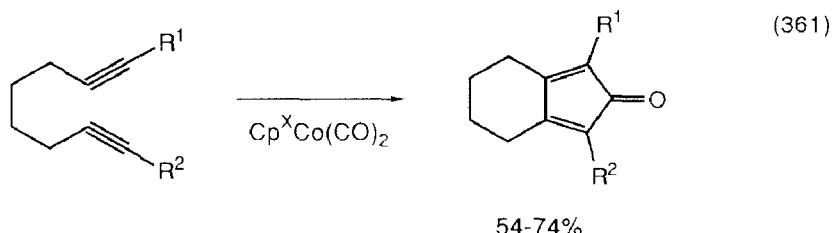
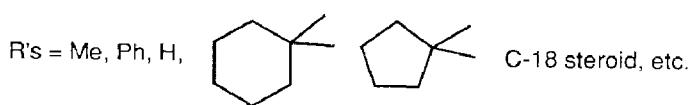
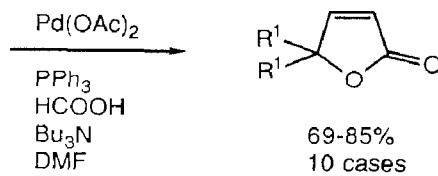
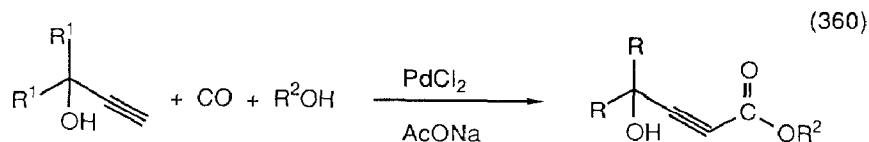
Cyclopentenones were made in a multistep process involving both palladium(II) and nickel(0) catalysis (equation 355) [437]. Dienes were methoxycarbonylated by palladium(II) catalysts (equation 356) [438]. Enynes were cyclocarbonylated using rhodium(0) catalysts (equation 357) [439]. Allene was acylated at the 2-position by nickel carbamoyl complexes and the resulting  $\pi$ -allyl ligand underwent further useful transformation (equation 358) [440]. Addition of organolithium reagents to iron-complexed conjugated enones resulted in acylation (equation 359) [441].

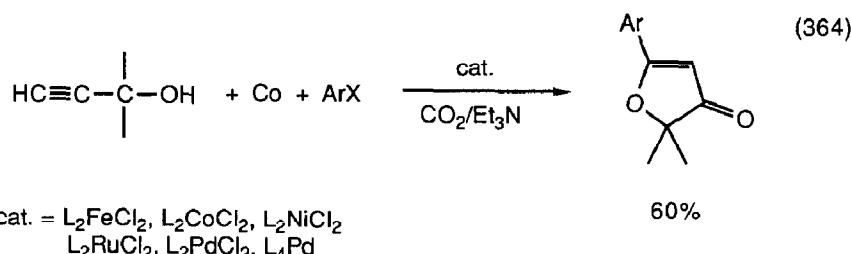
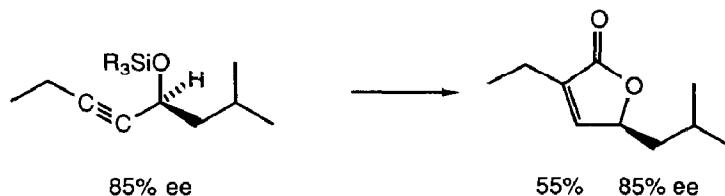
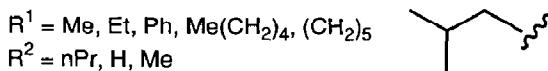
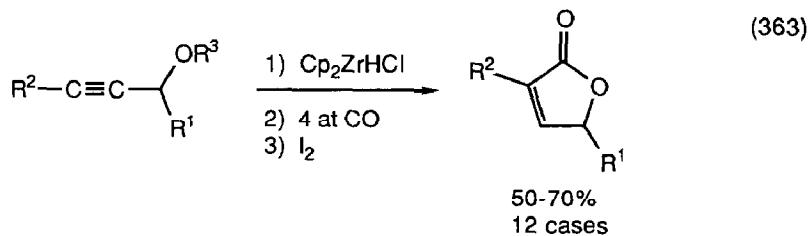




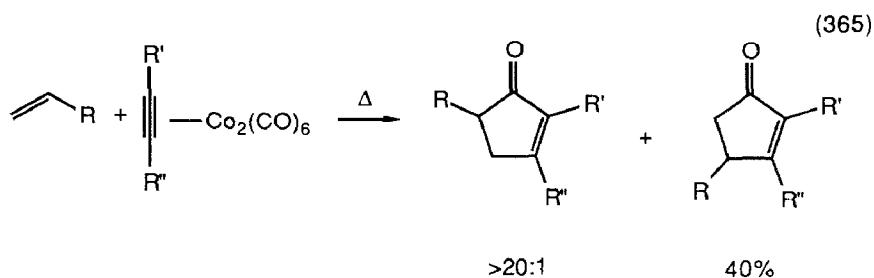
$\text{R} = \text{Me, Bn}$   
 $\text{Me} = \text{MgBr, Li, CuLi}$   
 $\text{R}' = \text{Bu, iBu, tBu, Me}$

Propargyl alcohols were carbonylated, then cyclized to butenolides using palladium catalysts (equation 360) [442]. Diynes and alkynes were cyclocarbonylated by cobalt (equation 361) [443], (equation 362) [444] and zirconium complexes (equation 363) [445]. Propargyl alcohols were arylated, carbonylated, and cyclized by a variety of transition metal catalysts (equation 364) [446]. Phenyl acetylene was reductively carbonylated to aryl alcohols using  $\text{Fe}_3(\text{CO})_{12}$  as a catalyst [447].



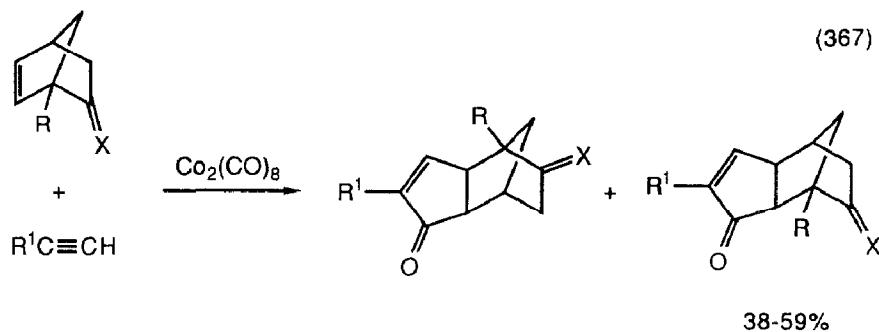
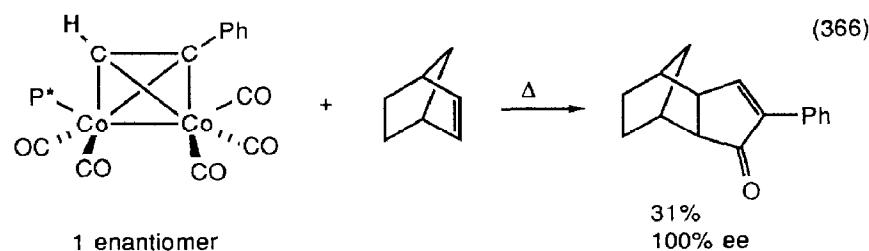


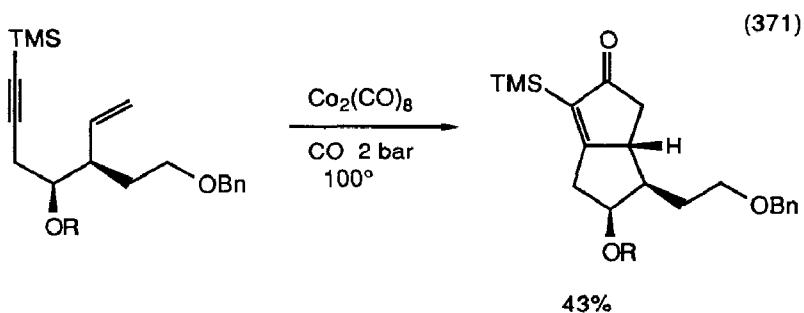
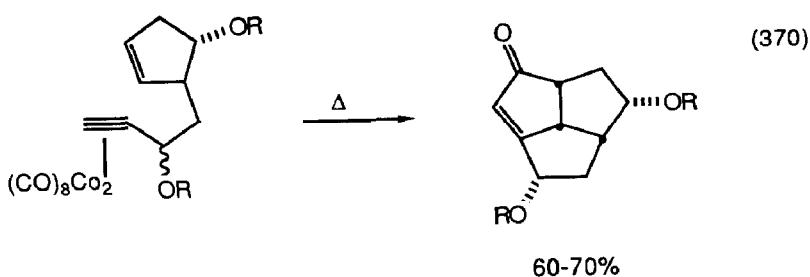
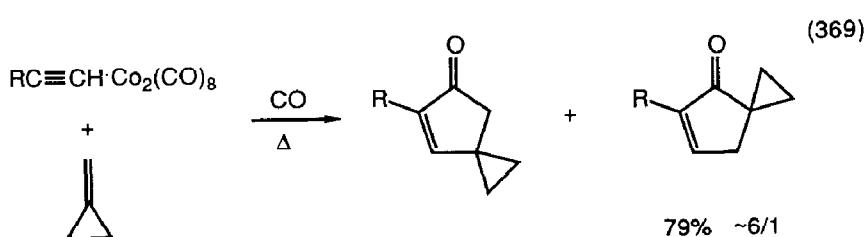
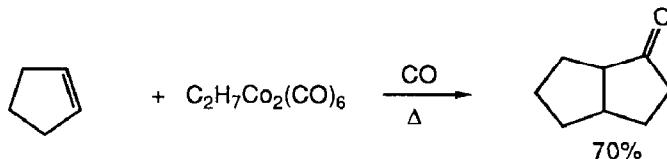
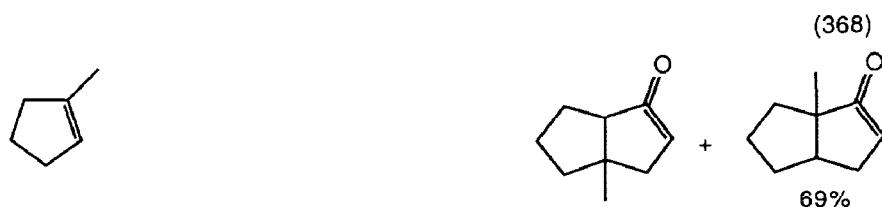
The temperature could be lowered and the yield of the Pauson-Khand reaction increased by using ultrasound and phosphine oxide as additives [448]. With disubstituted alkynes regio control was observed (equation 365) [449]. Using a chiral cobalt alkyne complex as a reactant resulted in very high asymmetric induction (equation 366) [450]. The effect of substituents on regiochemistry for the Pauson-Khand reaction has been studied (equation 367) [451]. Large excesses of olefin increased the yield (equation 368) [452]. Methylenecyclopropane participates in the process to give spiro derivatives (equation 369) [453]. A variety of polycyclic systems have been synthesized using this reaction (equation 370) [454], (equation 371) [455], (equation 372) [456].



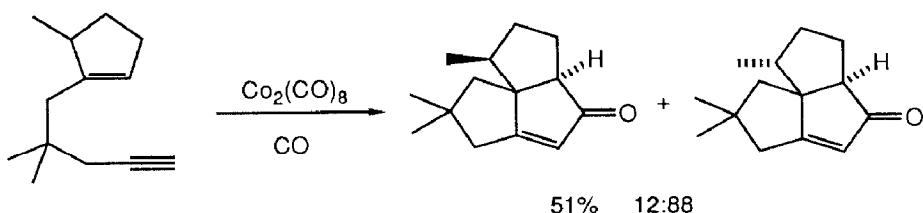
$R' = \text{Ph, Me, Bu}$   
 $R'' = \text{Me}$

$R'' = \text{H} \quad 1:1$



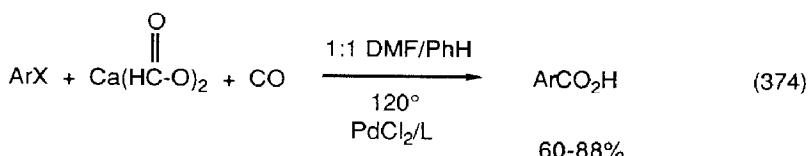
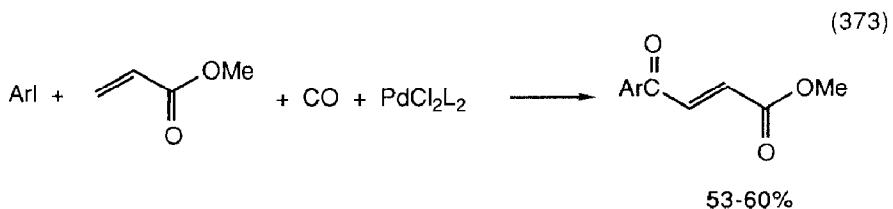


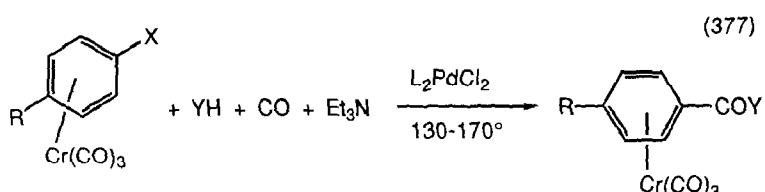
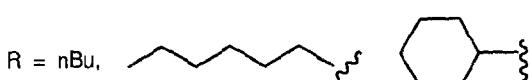
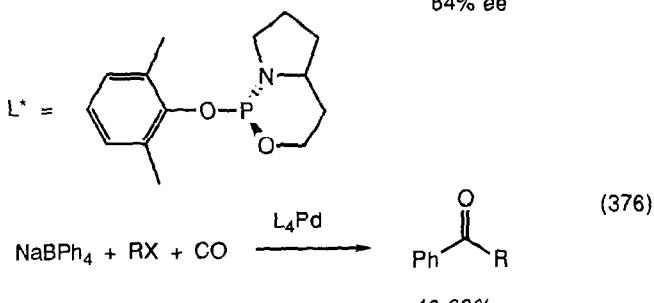
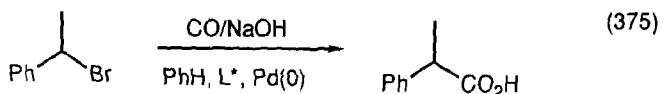
(372)



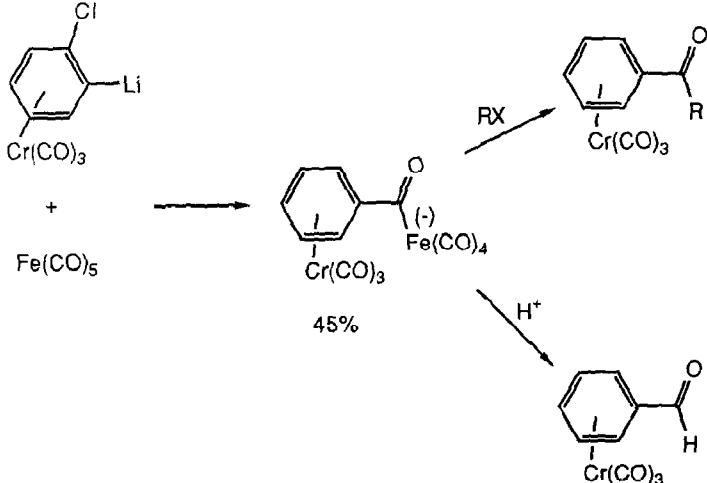
## 2. Carbonylation of Halides

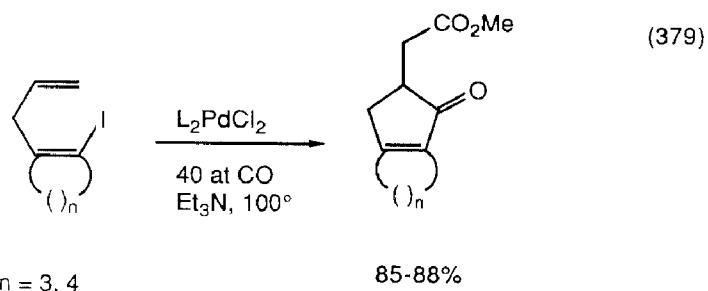
Aryl halides were carbonylatively vinylated using palladium(0) catalysts (equation 373) [457]. Aryl halides were converted to aryl carboxylates by calcium formate and palladium catalysts (equation 374) [458].  $\alpha$ -Phenethylbromide was carboxylated in 64% ee using chiral palladium catalysts (equation 375) [459]. Acylpalladium(II) complexes were converted to aldehydes, ketones and  $\alpha$ -diketones when treated with dialkylzinc reagents [460]. Alkyl halides were converted to phenylketones by sodium tetraphenylborate using palladium(0) catalysts (equation 376) [461]. Aryl halides complexed to chromium tricarbonyl were carbonylated using palladium catalysts (equation 377) [462]. Iron carbonyl carbonylated lithiated arene chromium tricarbonyls (equation 378) [463]. Palladium also catalyzed more complex acylations (equation 379) [465].



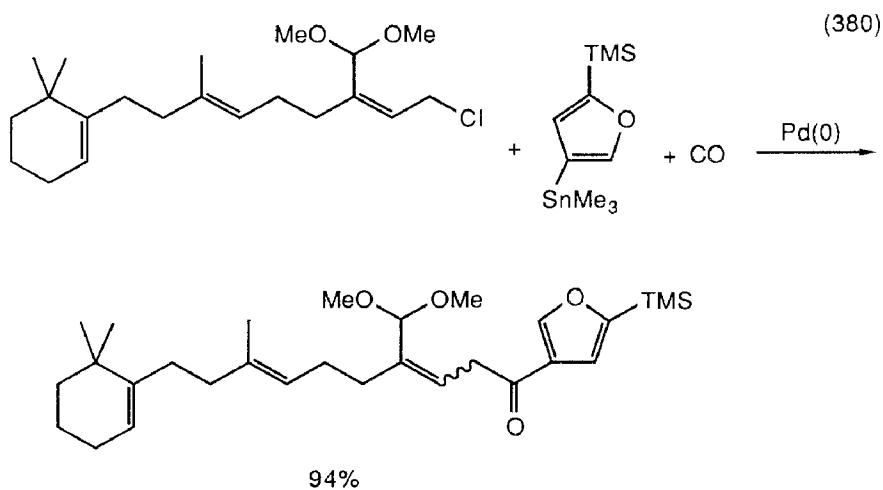


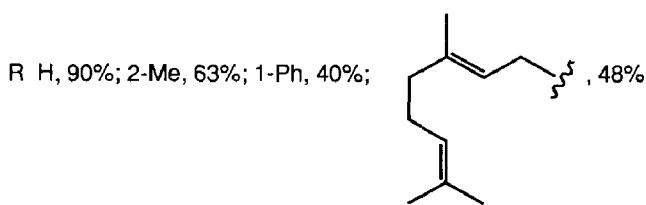
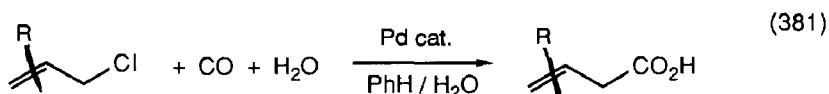
$X = \text{Cl, Br}$   
 $R = \text{H, CF}_3, \text{Me, MeO}^-, \text{Me}_2\text{N}$   
 $Y = \text{MeO, H, Et}_2\text{N}$



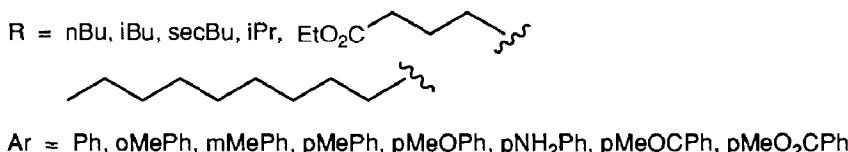
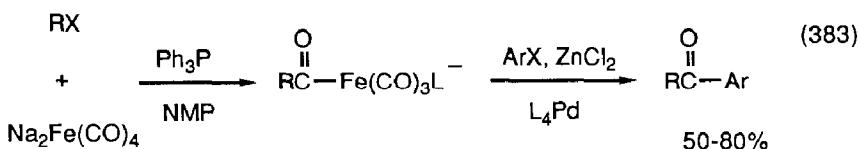
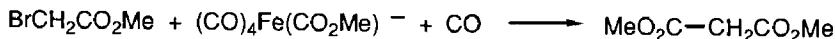


Allylic halides were carbonylated to  $\beta,\gamma$ -unsaturated acids by palladium catalysts (equation 381) [466]. Phase transfer conditions for the conversion of a broad range of aryl halides to the corresponding carboxylates using  $\text{Co}_2(\text{CO})_8$  or  $\text{Ni}(\text{CN})_2$  as catalysts have been worked out [467]. A detailed study of the phase-transfer carbonylation of benzyl halides to dibenzyl ketones and phenyl acetic acid using iron carbonyl as catalyst has been reported [468].  $\alpha$ -Halo esters were carboxylated by iron carboxylate species (equation 382) [469]. Aryl halides were acylated by acylate iron complexes in the presence of palladium catalysts (equation 383) [470]. Photolysis of alkyl iodides in alcohol under carbon monoxide pressure in the presence of a wide range of catalysts resulted in carbonylation [471].

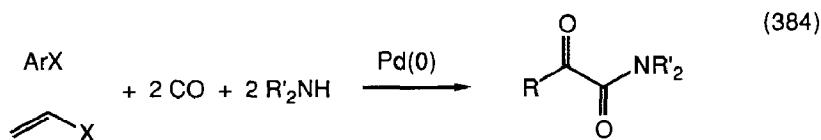


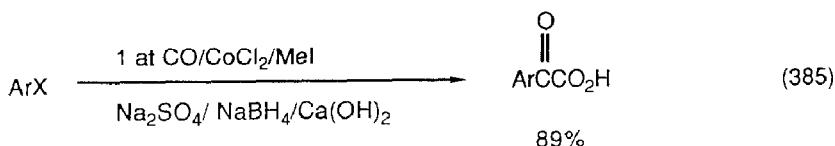


(382)



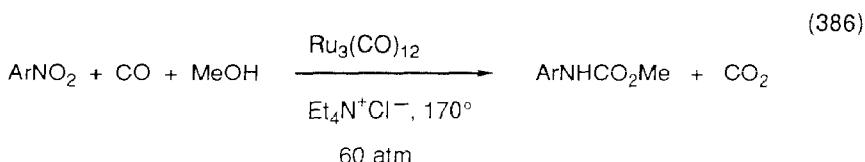
Detailed studies of the double carbonylation of aryl and vinyl halides using palladium(0) catalysts (equation 384) [472] and cobalt catalysts (equation 385) [473] have been published. Aryl iodides were coupled to benzils by L<sub>2</sub>PdCl<sub>2</sub> under carbon monoxide [474]. Inorganic bases promoted the double carbonylation of iodobenzene to phenylglyoxylic acid by palladium(II) phosphine complexes [475].



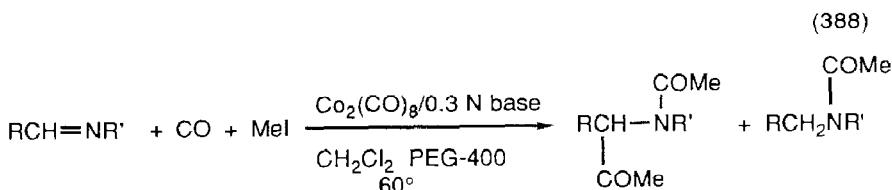
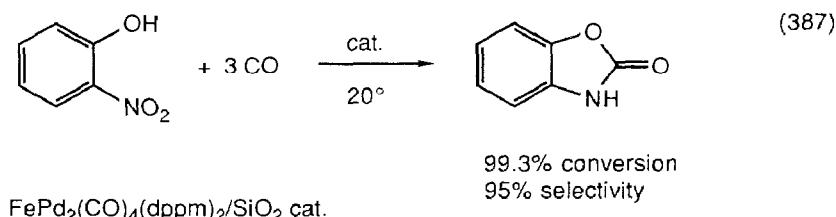


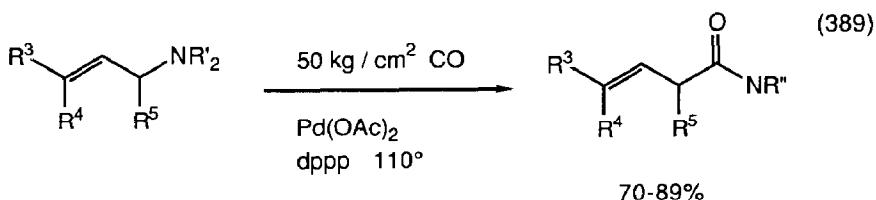
### 3. Carbonylation of Nitrogen Compounds

Ruthenium carbonyls [476] and palladium phenanthroline complexes [477] catalyzed the reductive carbonylation of nitroaromatics to urethanes (equation 386). Palladium chloride/copper chloride catalysts converted diphenyl urea into carbamate esters under CO and O<sub>2</sub> at 100° [478]. O-Nitrophenol was converted to a cyclic carbamate by iron/palladium catalysts (equation 387) [479]. Cobalt carbonyl catalyzed the carbonylation of imines (equation 388) [480]. Allyl amines were carbonylated to β,γ-unsaturated amides by palladium catalysts (equation 389) [481].



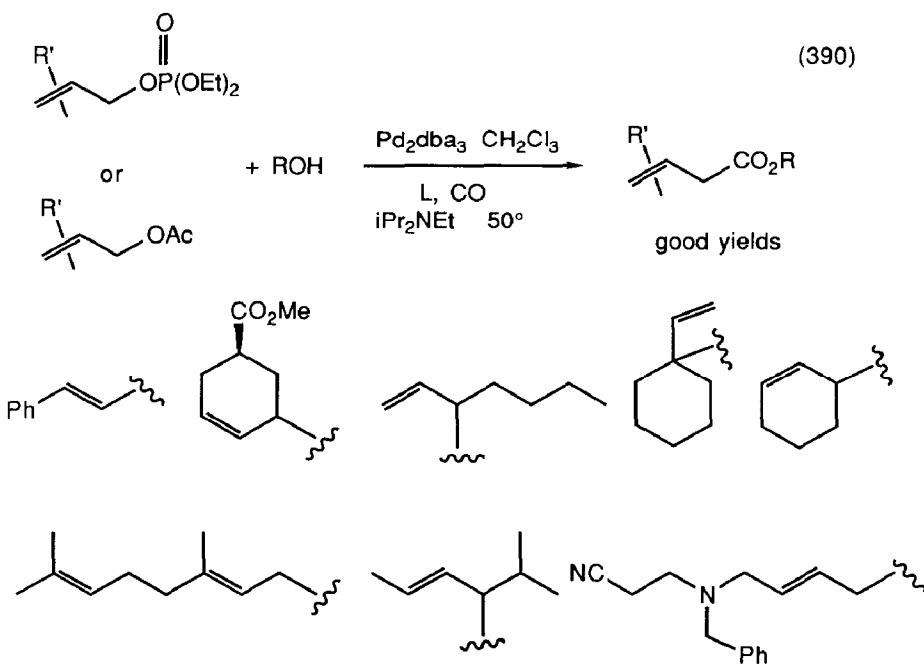
Ar = Ph, mMePh, mCF<sub>3</sub>Ph, pMePh, pMeO<sub>2</sub>CPh, 3,4-Cl<sub>2</sub>Ph, 3,5-Cl<sub>2</sub>Ph

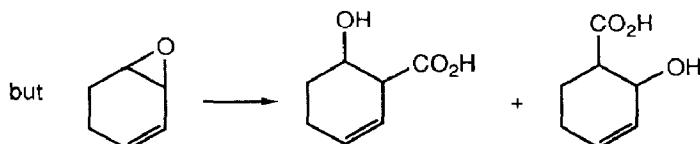
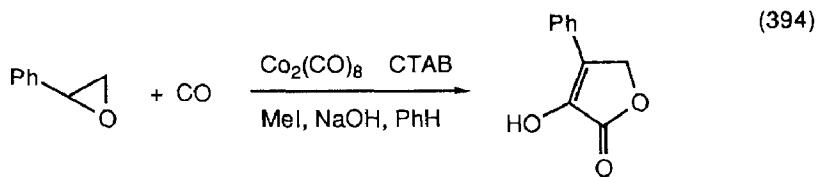
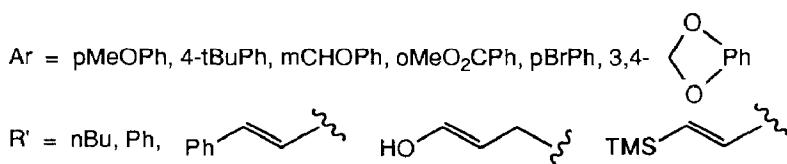
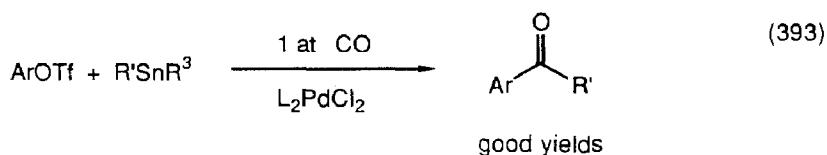
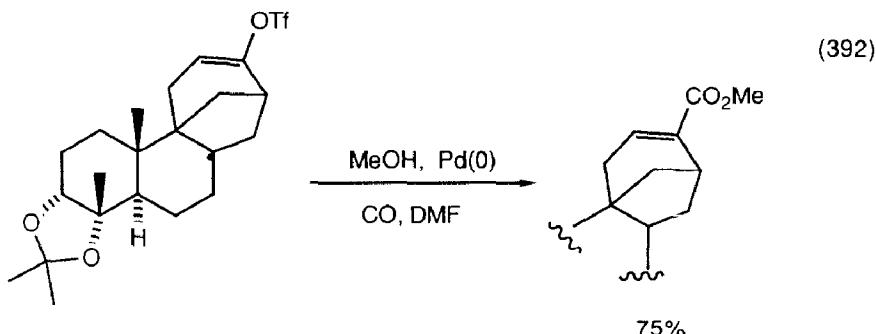
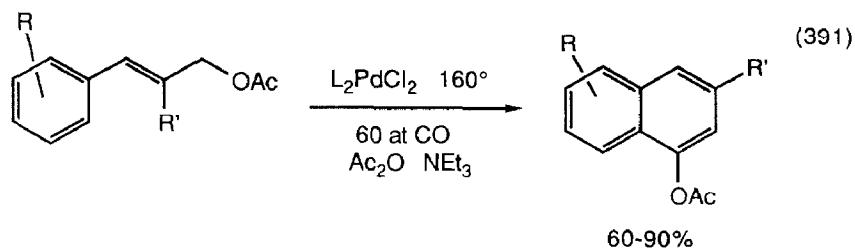


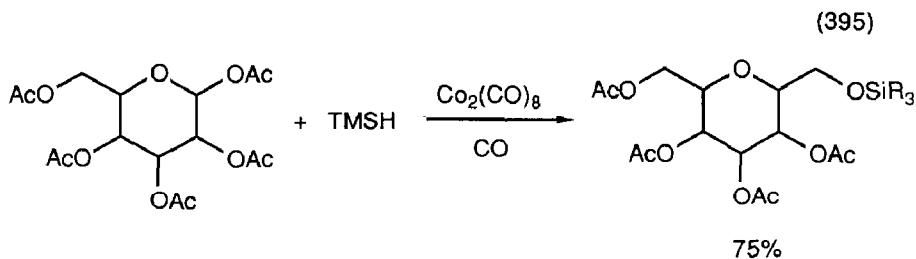


#### 4. Carbonylation of Oxygen Compounds

Mechanistic aspects of transition-metal catalyzed alcohol carbonylation have been reviewed (128 references) [482]. Palladium catalyzed the carbonylation of allylic acetates and phosphonates (equation 390) [483]. Cyclization occurred with cinnamyl acetates (equation 391) [484]. Palladium complexes also catalyzed the carbonylation of vinyl triflates (equation 392) [485] and aryl triflates (equation 393) [486]. Cobalt carbonyl carbonylated epoxides (equation 394) [487] and catalyzed the reductive carbonylation of carbohydrate acetates (equation 395) [488].

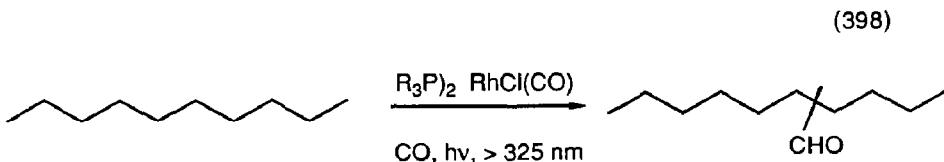
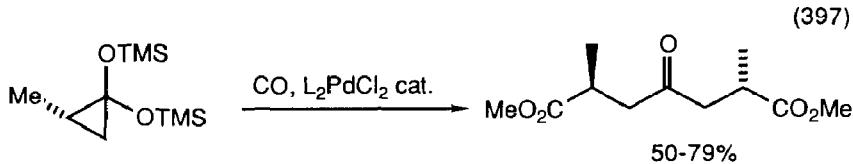
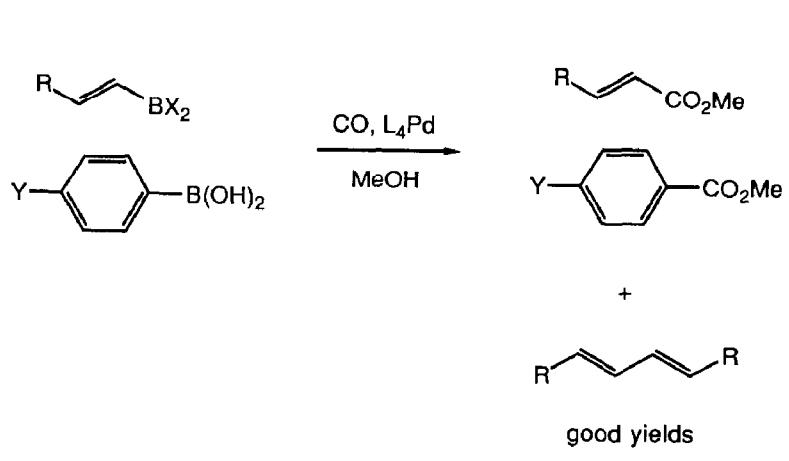


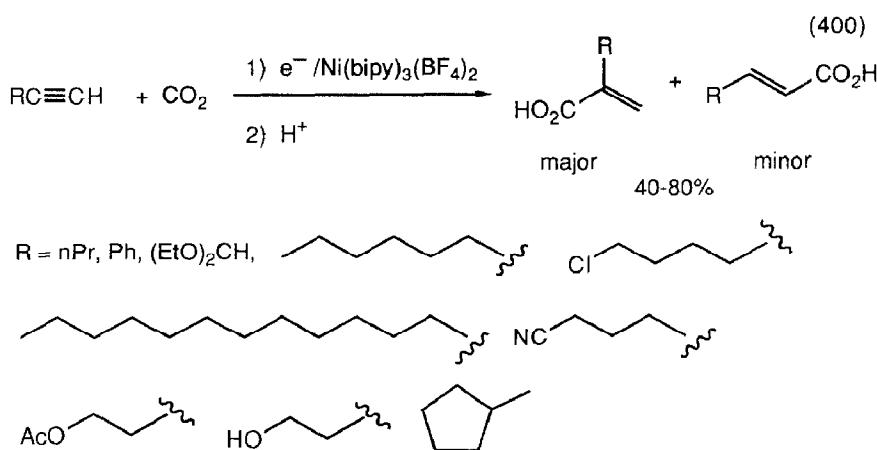
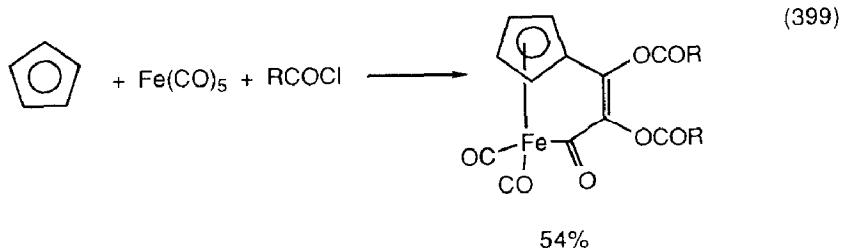




### 5. Miscellaneous Carbonylations

Palladium(0) complexes catalyzed the carbonylation of aryl and vinyl boranes (equation 396) [489], and acetylenic tellurides [490]. Cyclopropanone ketals were ring opened and carbonylated by CO with palladium catalysis (equation 397) [491]. Hydrocarbons were carbonylated by rhodium(I) complexes under photolytic conditions (equation 398) [492]. The strange carbonylation in equation 399 has been reported [493].



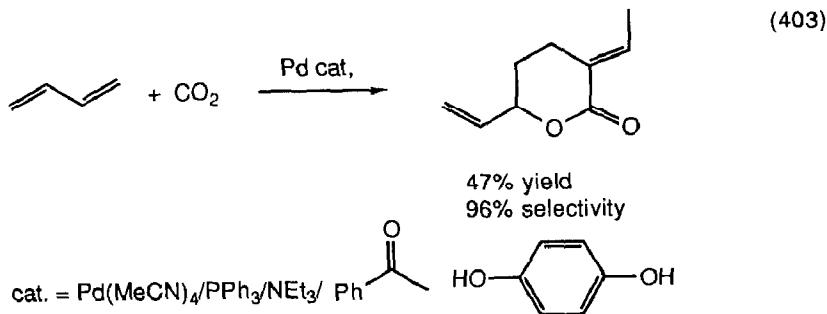
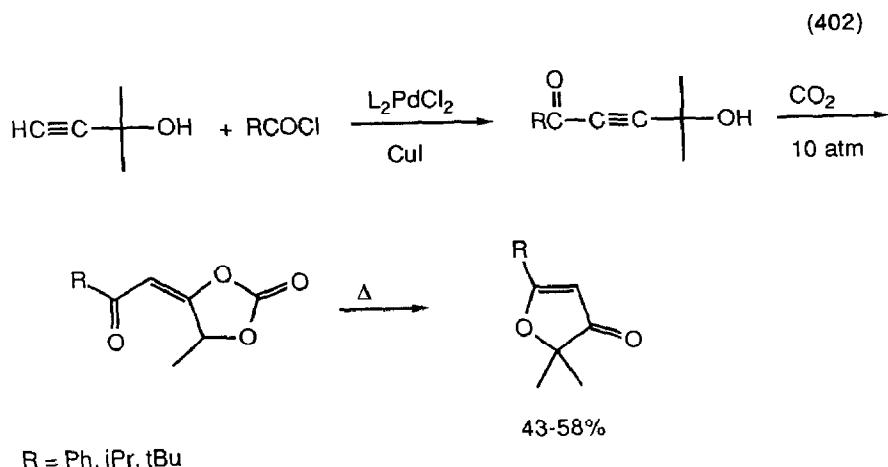
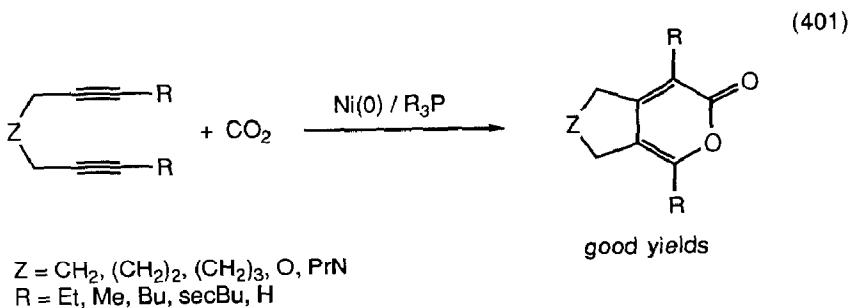


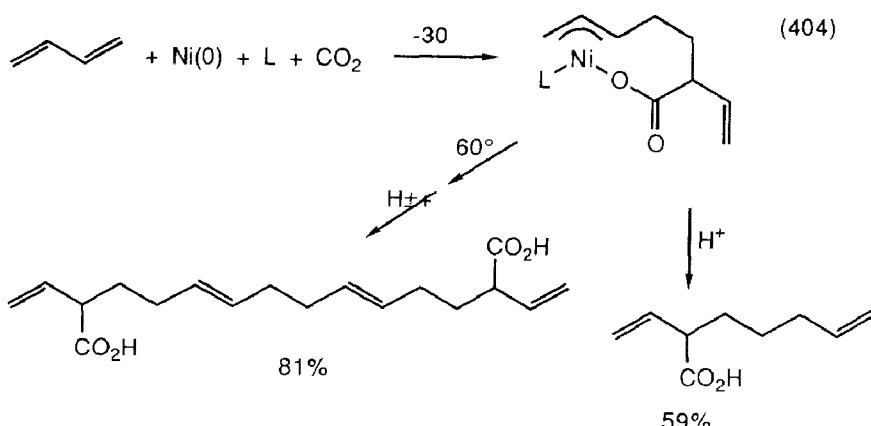
## 6. Decarbonylation Reactions

There were no synthetically significant decarbonylations reported this year.

## 7. Reactions of Carbon Dioxide

The following reviews have appeared: "Carbon dioxide as building block for fine chemicals synthesis by homogeneous catalysis" (98 references) [494]; "Carbon dioxide as an alternative C<sub>1</sub> synthetic unit: activation by transition metal complexes" (191 references); [495]; "Reactions of carbon dioxide with carbon-carbon bond formation catalyzed by transition-metal complexes" (121 references) [496]. Electrochemical reduction of terminal alkynes in the presence of carbon dioxide and a nickel catalyst produced acrylic acids (equation 400) [497][498]. Metal alkylcarbamates were prepared from primary amines and carbon dioxide, then decomposed by acid halides to give isocyanates [499]. Diynes were cyclocarboxylated by nickel(0) catalysts (equation 401) [500] while palladium catalysts formed cyclic carbamates from propargyl alcohols and carbon dioxide (equation 401) [501]. Palladium complexes catalyzed the cocyclooligomerization of dienes and carbon dioxide to give lactones (equation 403) [502], while nickel complexes produced open chain oligomers (equation 404) [503].

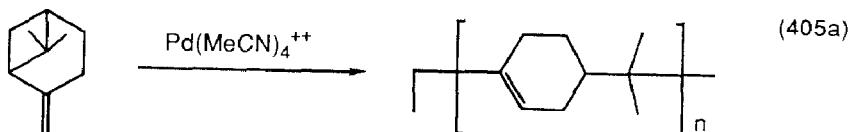




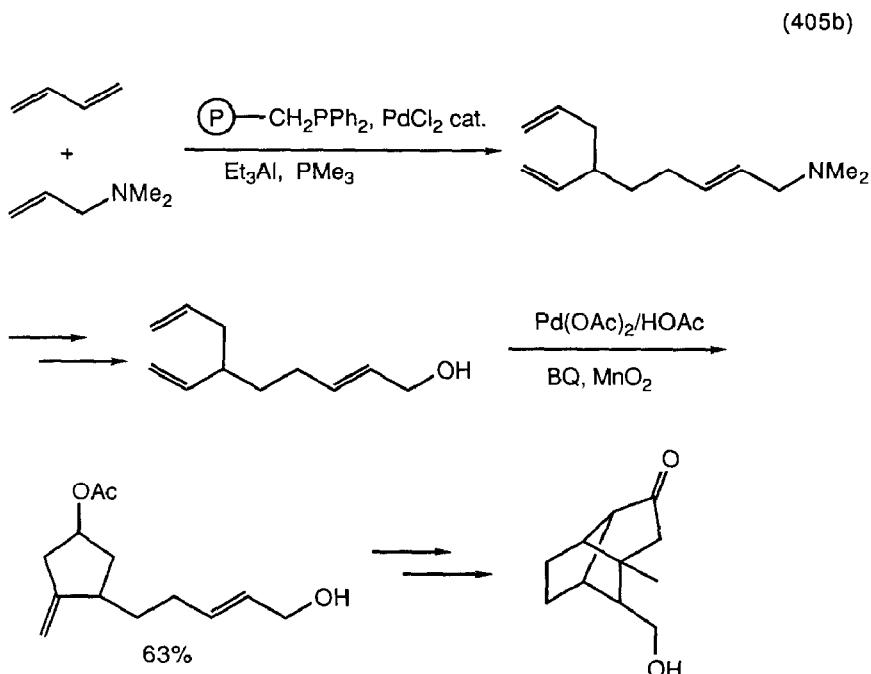
#### D. Oligomerizations

The influences of ligands on the catalytic properties of nickel containing systems in oligomerization and polymerization reactions has been reviewed (79 references) [504]. The mechanisms of SHOP process have been reviewed [505]. Dicyclopentadienyltitanium ethylene complexes catalyzed the dimerization of ethylene to 1- and 2-butene in a 1:1 ratio [506]. Butadiene diphos zirconium(I) ethyl complexes catalyzed the dimerization of ethylene to 1-butene [507]. The contents of a lecture, the dimerization of ethylene and propylene with tailor-made nickel complexes supported on silica have been published [508]. Cationic  $\pi$ -allyl nickel phosphine complexes catalyzed the trimerization of propene to trimethyl pentanes as major products [509]. Nitrogen and oxygen containing nickel chelates dimerized ethylene [510]. Rhodium(I) complexes dimerized methyl vinyl ketone [511]. Nickel(0) complexes catalyzed the dimerization and trimerization of methylene cyclopropane [512].

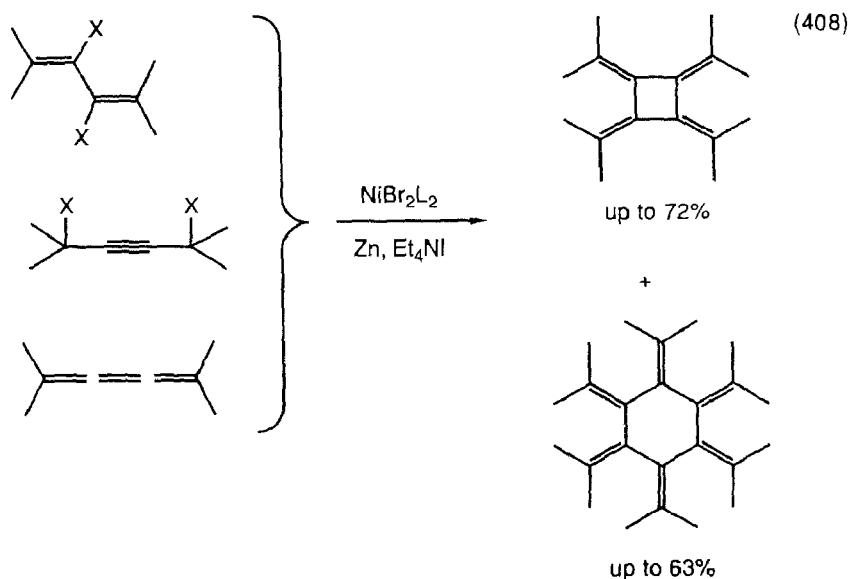
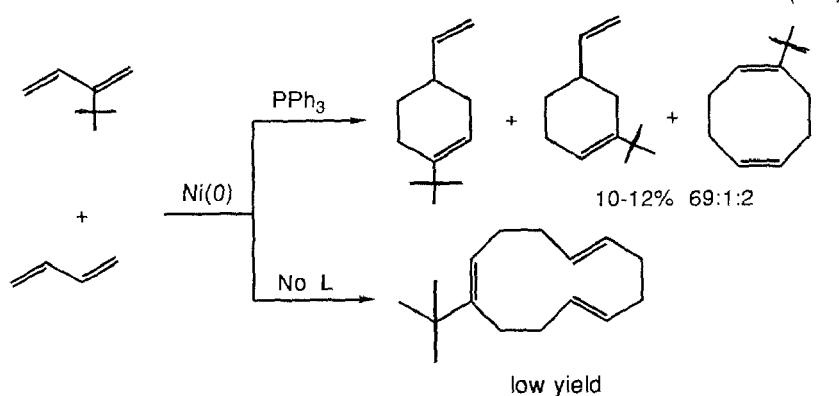
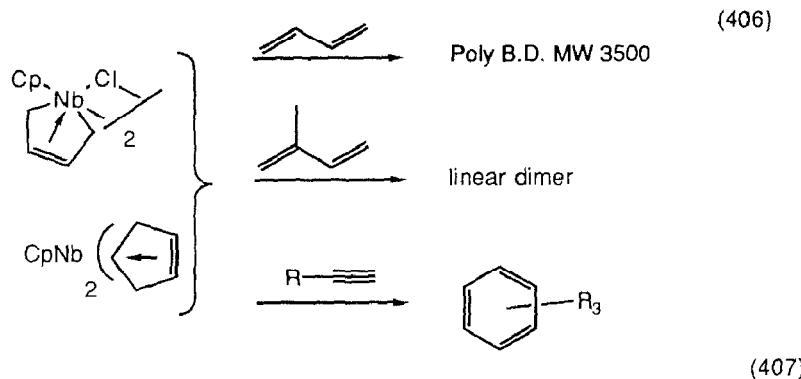
Oligomerization of monoolefins has been reviewed [513]. Treatment of dimethyl titanocene with dimethyl anilinium tetraphenyl borate produced a complex which catalyzed the polymerization of ethylene [514]. Cationic palladium(II) complexes catalyzed the polymerization of olefins (equation 405a) [515]. Cationic cyclopentadienylchromium methyl complexes catalyzed the polymerization of ethylene [516]. Styrene and propylene were codimerized to phenylpentenes by nickel-containing Ziegler-Natta catalysts [517].

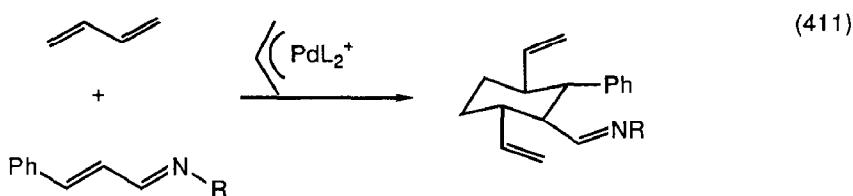
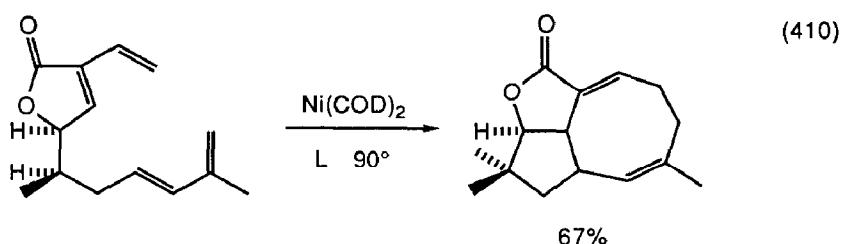
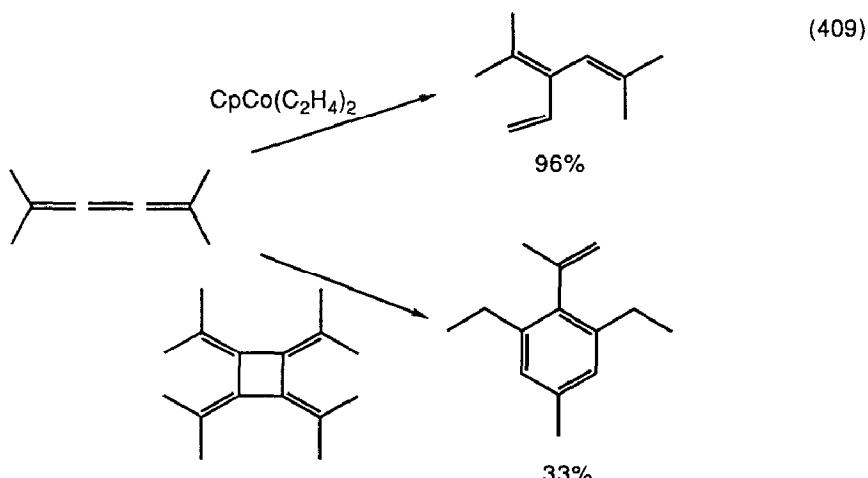


Application of telomerization and dimerization in the synthesis of fine chemicals has been reviewed [518]. Copper induced the telomerization of tetrafluoroethylene with fluoroalkyliodides [519]. Palladium(II) acetylacetonate catalyzed the telomerization of isoprene with phthalimides to give mixtures of seven isomeric terpene amines [520]. A report from a symposium dealing with selective palladium-catalyzed transformations of dienes has appeared [521]. Sepiolite-bound palladium complexes catalyzed the telomerization of butadiene with methanol [522]. A component of San Jose's scale's sex pheremone was synthesized by palladium catalyzed telomerization of isoprene with diethylamine [523]. A bridged polycyclic ketone was synthesized from a butadiene/allylamine telomerization product (equation 405b) [524].



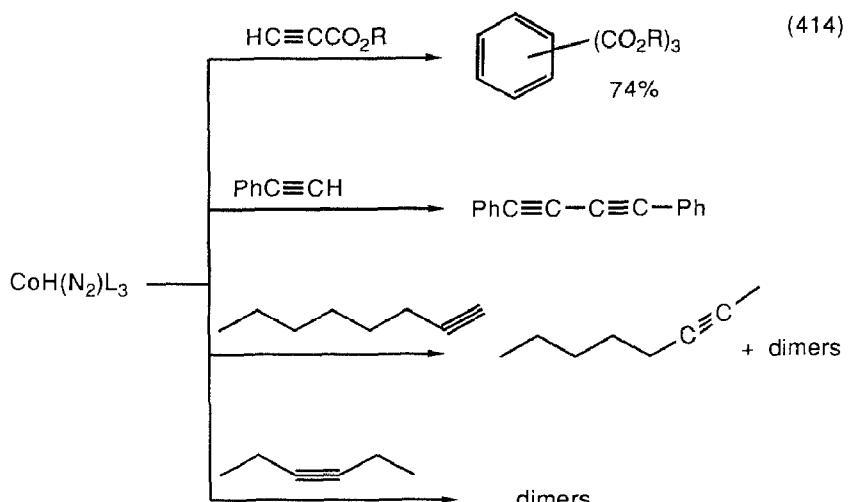
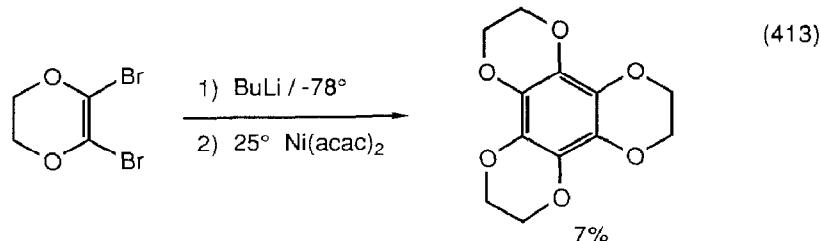
Modified nickel(0) catalysts linearly dimerized 1,3-butadiene [525]. Complex iron and nickel catalysts cyclooligomerized isoprene [526]. Butadienoniobium catalysts polymerized butadiene, dimerized isoprene and cyclotrimerized alkynes (equation 406) [527]. Nickel(0) complexes cocyclooligomerized butadiene and 2-t-butylbutadiene (equation 407) [528]. Allenes, alkynes and heavily substituted dienes were cyclooligomerized by nickel complex catalysts (equation 408) [529] and linearly oligomerized by cobalt catalysts (equation 409) [530].  $\text{Ni}(\text{COD})_2$  cyclized dienes to give a tricyclic system (equation 410) [531]. Cationic  $\pi$ -allylpalladium complexes co-cyclooligomerized butadiene and 1-azadienes (equation 411) [532].





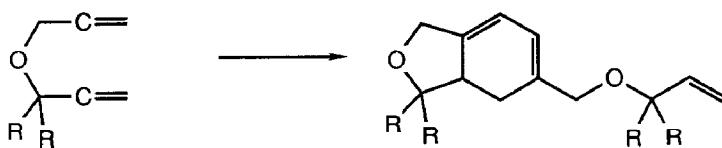
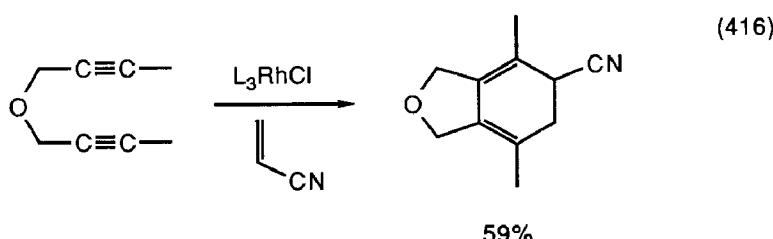
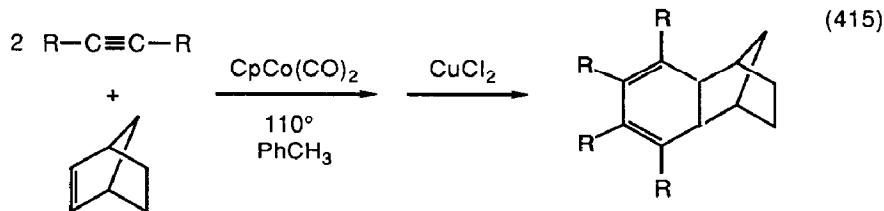
A dissertation investigating the role of metallocyclopentadienes in transition metal catalyzed cyclooligomerization of acetylenes has appeared [533]. Cyclobutadienes were shown not to be intermediate in the cobalt-catalyzed cyclotrimerization of diaryl acetylene [534]. Mixtures of tantalum(V) chloride and EtAlCl<sub>2</sub> generated effective catalysts for cyclotrimerization of acetylene [535]. Molecular mechanics studies on the cyclotrimerization of acetylene to benzene on palladium(III) have been carried out [536]. Nickel complex catalysts cyclotetramerized propargyl alcohol [537].

Alkynes were cyclotrimerized by reduced nickel catalysts (equation 412) [538], (equation 413) [539]. Cobalt nitrogen complexes cyclotrimerized or oligomerized alkynes (equation 414) [540].

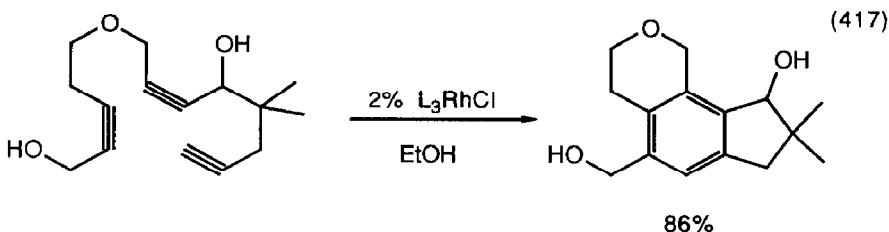


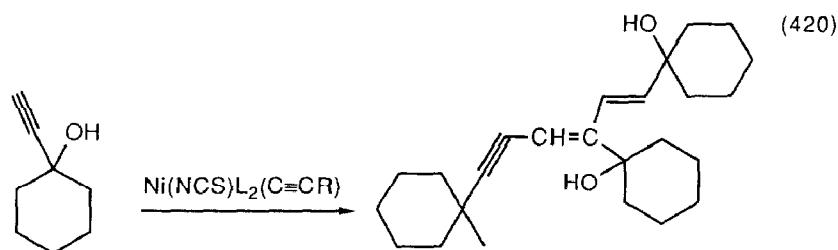
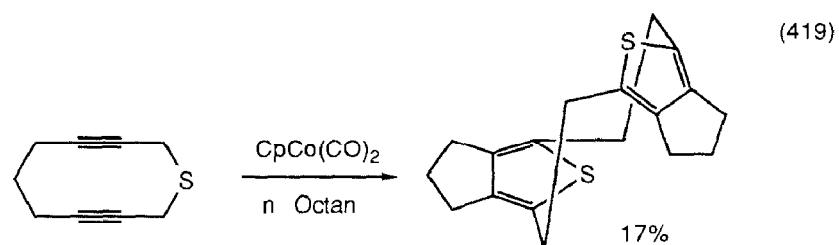
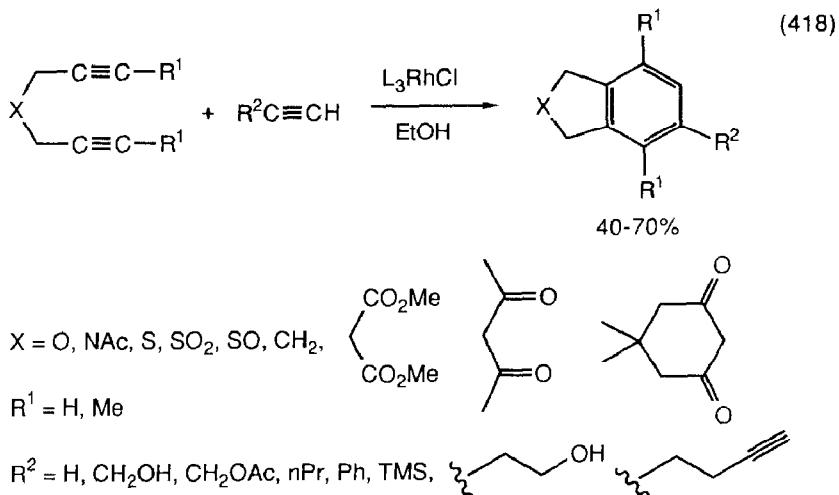
Cyclopentadienylcobalt carbonyl co-cyclotrimerized norbornene with internal alkynes (equation 415) [541]. Low valent cobalt complexes catalyzed the co-cyclodimerization of norbornadiene with diacetyles to give a host of bicyclic compounds [542]. Rhodium(I) complexes catalyzed the cyclotrimerization of alkynes and the co-cyclotrimerization of diynes with acrylonitrile (equation 416) [543]. The same complex catalyzed intramolecular cyclotrimerization of alkynes (equation 417) [544], (equation 418) [545]. Cobalt complexes catalyzed the cyclodimerization of diynes (equation 419) [546].

Nickel(II) complexes linearly trimerized 1-ethynyl-1-cyclohexanol (equation 420) [547]. Photoactivated tungsten hexacarbonyl polymerized ethynyl (trifluoromethyl) naphthalenes [548]. Terminal alkynes such as phenylacetylene and 1-hexyne were polymerized by  $\text{W}(\text{CO})_3(\text{MeCN})_3/\text{Cp}_2\text{Fe}^+$  catalysts [549].

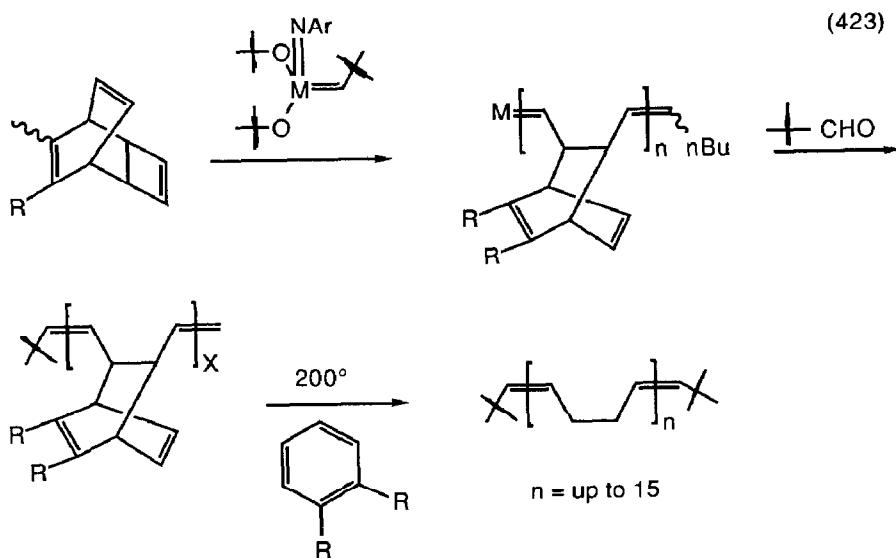
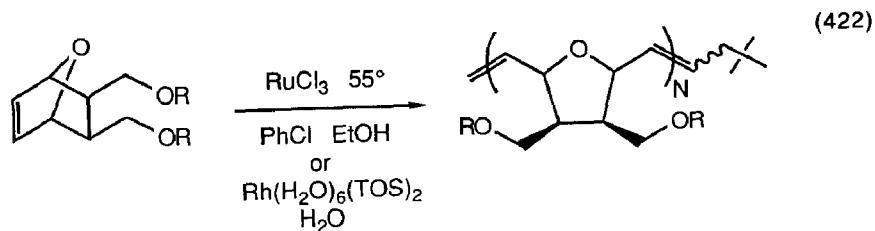
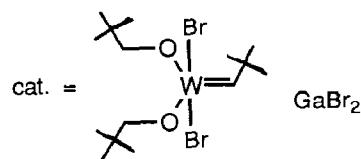
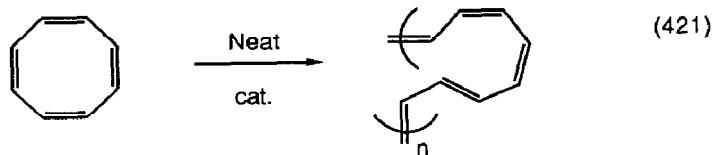


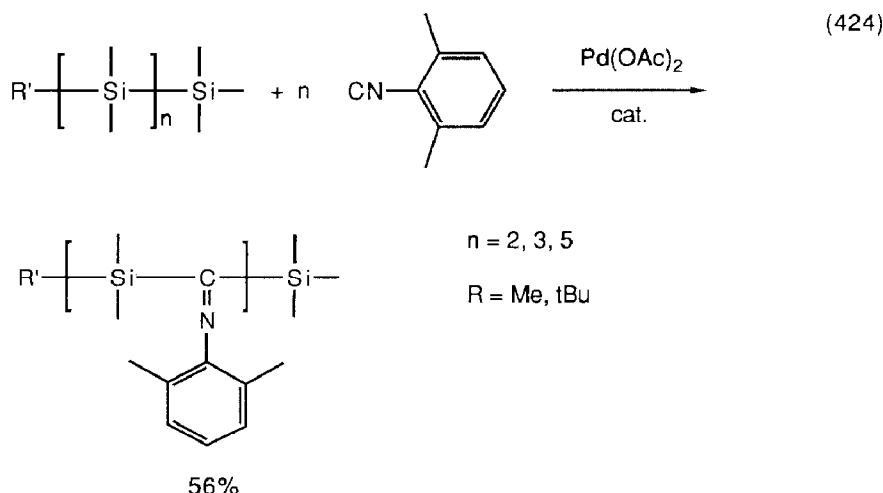
R = OH, 60%; R = Me, 89%





Tungsten alkylidene/gallium bromide mixtures polymerized cyclooctatetraene to polyacetylene (equation 421) [550]. Functionalized bicyclic olefins underwent ROMP polymerization using ruthenium catalysts in water (equation 422) [551]. Alkylidene complexes were used to prepare polyacetylenes from bicyclic polyenes (equation 423) [552]. Palladium(II) acetate catalyzed the co-polymerization of isocyanides with polysilanes (equation 424) [553].



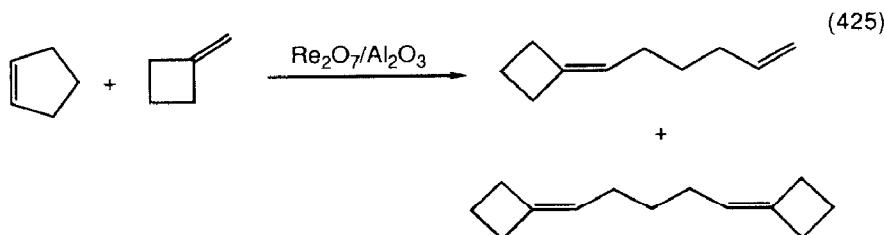


## E Rearrangements

### 1. Metathesis

The following reviews dealing with olefin metathesis have appeared: "Metathesis of alkenes" (7 references) [554]; "Metathesis: a breakthrough in organic chemistry" [555]; "The seventh international symposium on olefin metathesis" [556]; "Running the impossible reaction, metathesis of cyclohexene" (41 references) [557]. The proceedings of a symposium on design of selective olefins metathesis catalysts have been published [558].

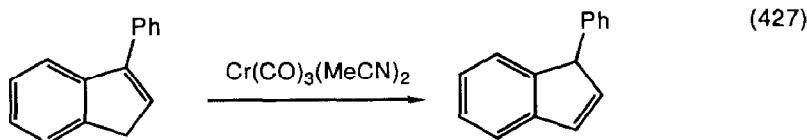
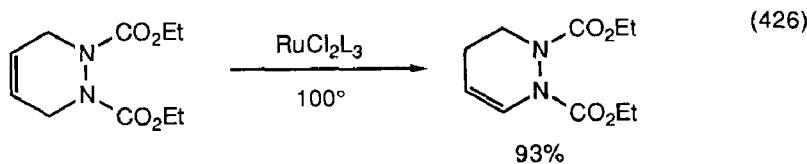
Photolysis of tungsten hexacarbonyl and polysilanes produced active olefin metathesis catalysts [559], as did photolysis of tungsten hexacarbonyl/titanium tetrahalide [560]. Precursors to olefin metathesis catalysts were produced by treatment of  $\text{Re}_2\text{O}_7$  with tetramethyl tin or dimethyl zinc [561]. Homogeneous metathesis catalysts were made from  $\text{CpW}(\text{CO})_3/\text{iBuAlCl}_2/\text{O}_2/\text{RnX}_{3-n}\text{Sn}$  mixtures [562]. Rhenium catalysts cometathesized methylene cyclobutane and cyclopentane (equation 425) [563].



Tungsten and molybdenum hexachloride/EtAlCl<sub>2</sub> or R<sub>4</sub>Sn systems immobilized on polymers were effective catalysts for the metathesis of 1-hexene [564]. Molybdenum oxide films produced by vacuum sublimation were highly active metathesis catalysts [565]. Treatment of silica-bound chromium with Fischer carbene complexes produced an immobilized olefin metathesis catalyst [566]. Tungsten and molybdenum hexacarbonyl were adsorbed on NaY zeolites or Vycor glass and then photolyzed to produce metathesis catalysts [567]. Functionalized olefin metathesis has been reviewed (46 references) [568]. Allyl cyanide was metathesized by the catalyst system 1,1,3,3-tetramethyl-1,3-disilacyclobutane-tungsten hexachloride [569].

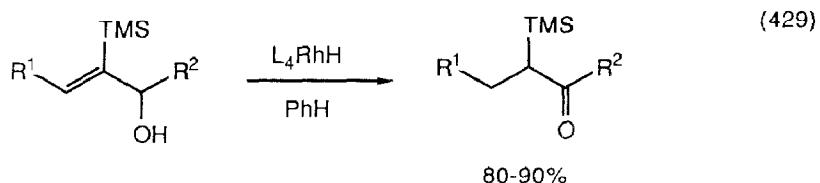
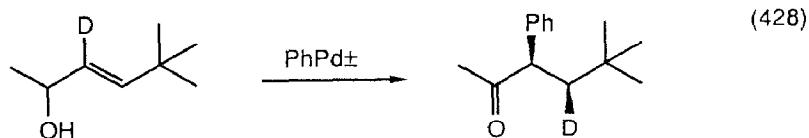
## 2. Olefin Isomerization

Rhenium(V) complexes with cyclic sulfides catalyzed olefin isomerizations [570]. The isomerization of nonconjugated cyclic dienes by Cp<sub>2</sub>TiH was subjected to molecular mechanics calculations [571]. Silica-polyalumazane palladium complexes catalyzed olefin isomerization [572], as did supported polynuclear Cu(II)-Al, Cr, or Fe complexes [573]. Other useful olefin isomerizations are shown in equation 426 [574] and equation 427 [575].

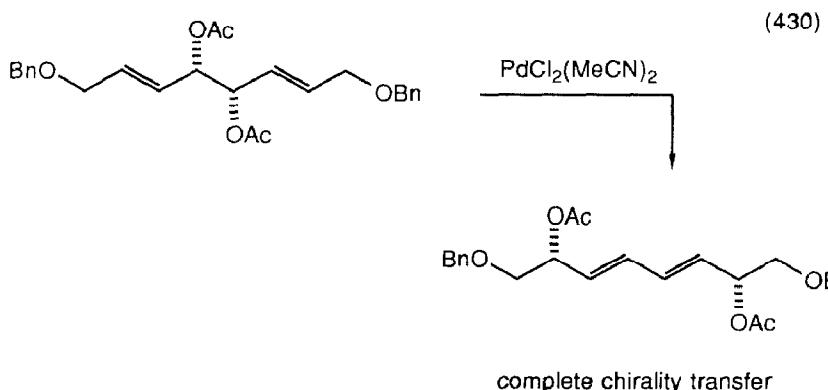
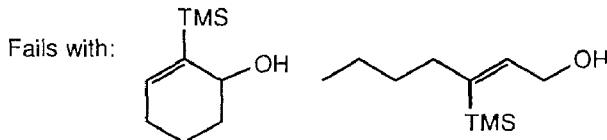
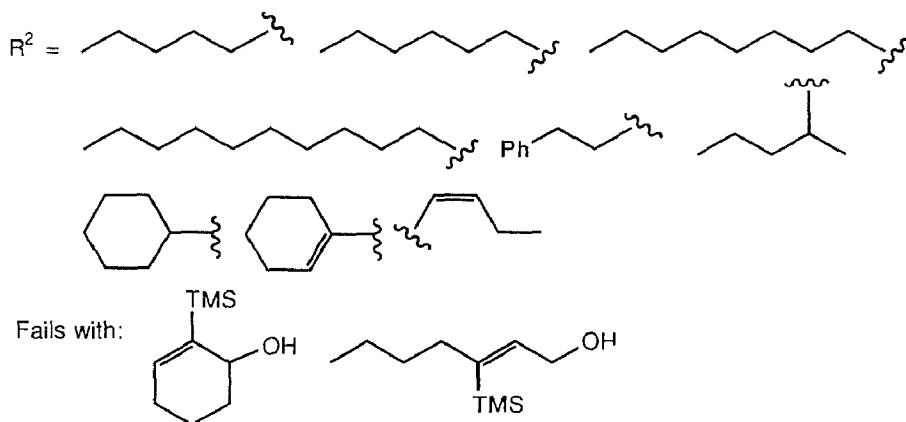


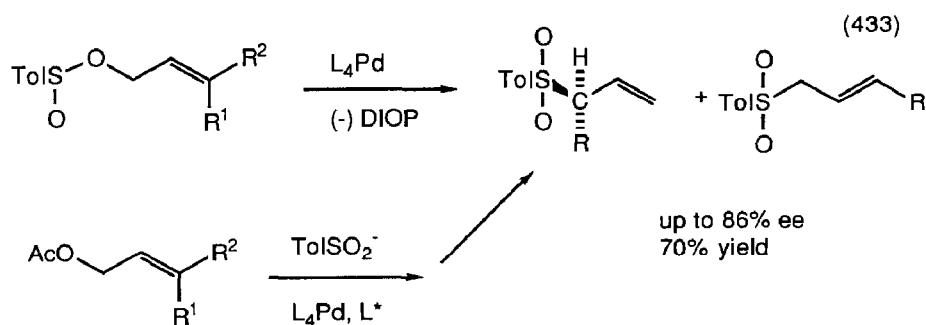
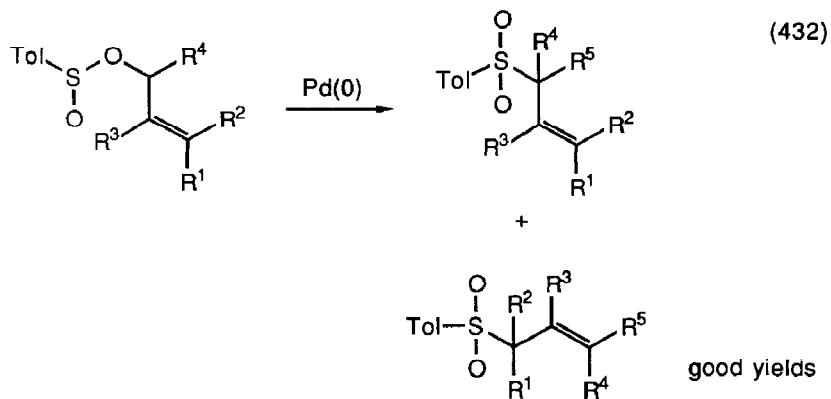
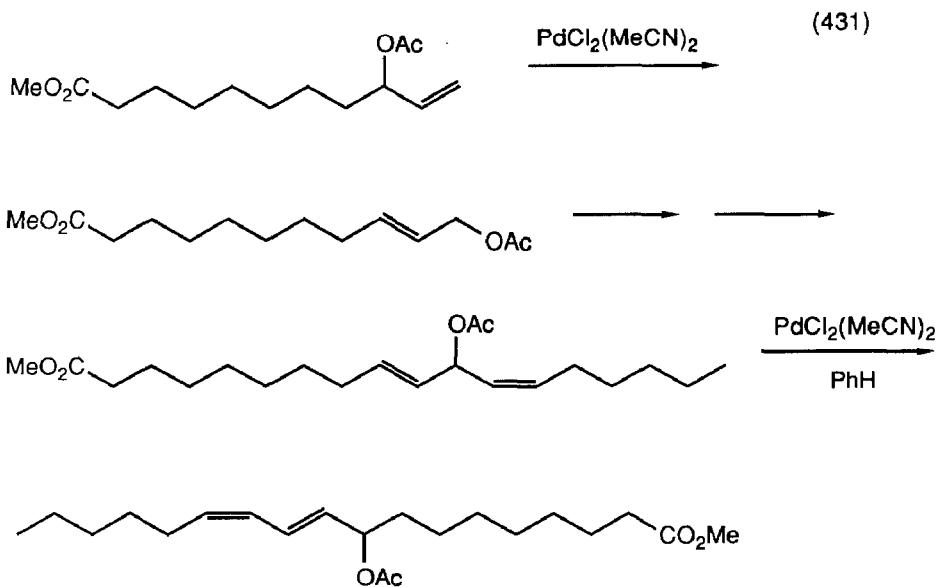
## 3. Rearrangement of Allylic and Propargylic Oxygen and Nitrogen Compounds

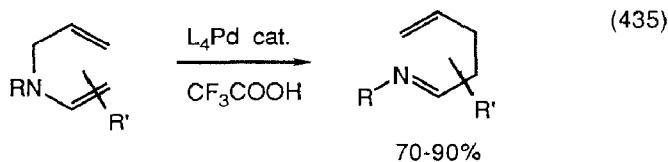
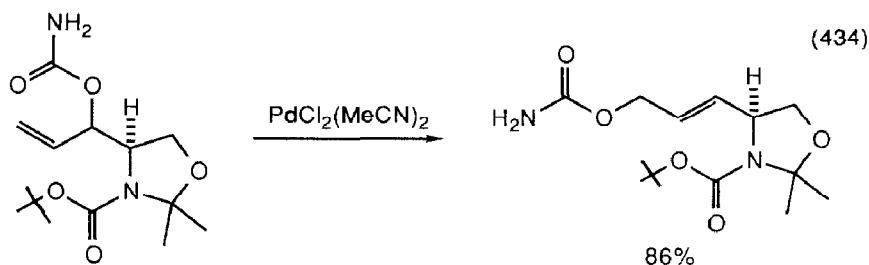
Arylation of allylic alcohols by arylpalladium(II) complexes resulted in a stereospecific 1,2-hydrogen shift (equation 428) [576]. Rhodium hydrides catalyzed the rearrangement of allylic alcohols to ketones (equation 429) [577]. Palladium chloride catalyzed the rearrangement of allylic acetates (equation 430) [578], (equation 431) [579], allylic sulfonates (equation 432) [580], (equation 433) [581], allyl carbamates (equation 434) [582], and N-allylenamines (equation 435) [583].



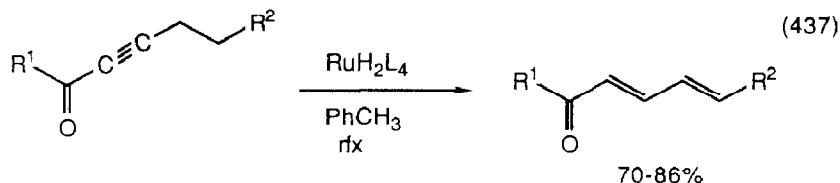
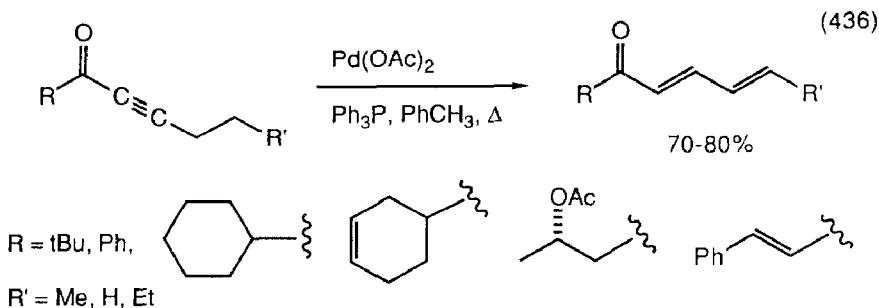
$\text{R}^1 = \text{H, Me, nBu}$



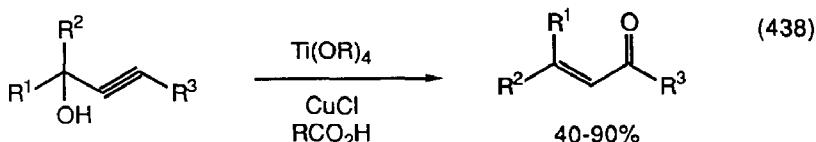




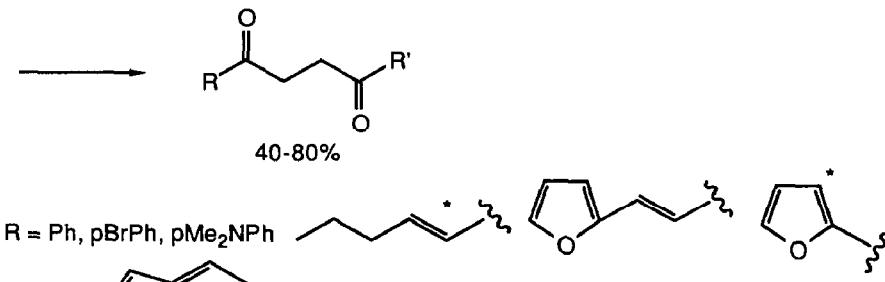
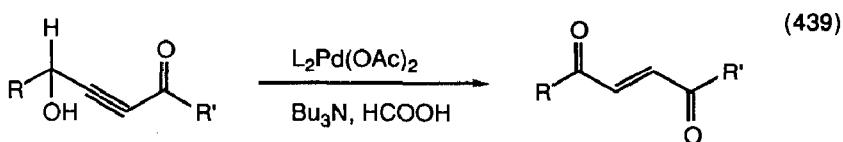
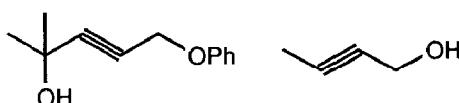
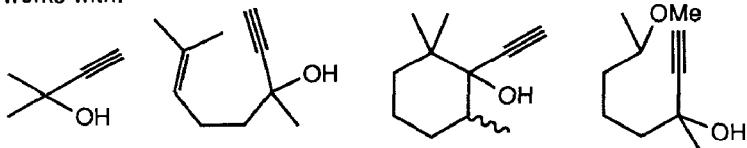
Acetylenic ketones were isomerized to dienones by palladium (equation 436) [584] and ruthenium (equation 437) [585] catalysts. Propargyl alcohols isomerized to conjugated ketones with titanium (equation 438) [586] and palladium (equation 439) [587] catalysts.



$R^1 = \text{Ph, Me, pMePh, pClPh, pMeOPh, Et}$   
 $R^2 = n\text{Pr, nBu}$



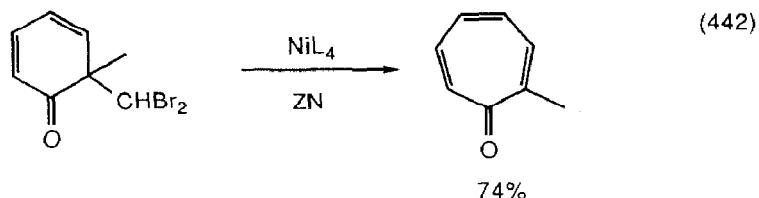
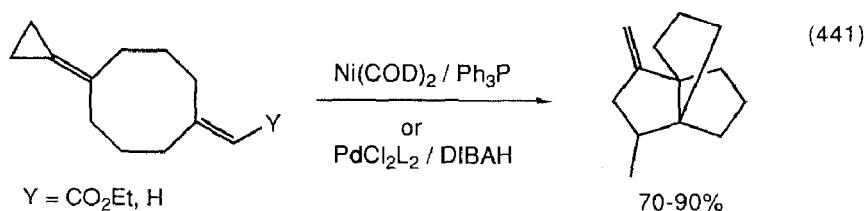
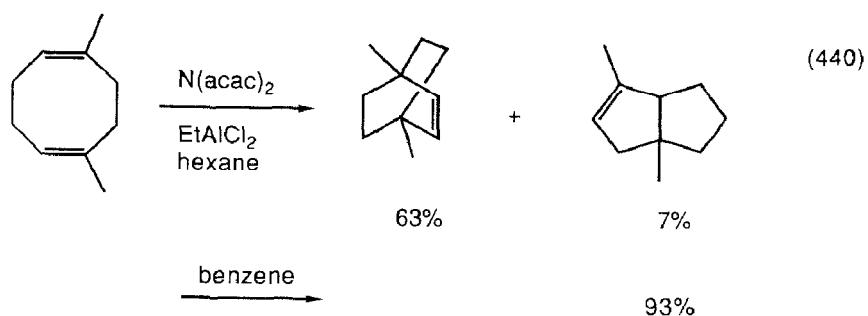
works with:



$R' = \text{OEt, } ^*\text{Ph}$

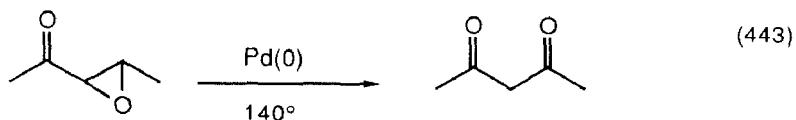
#### 4. Skeletal Rearrangements

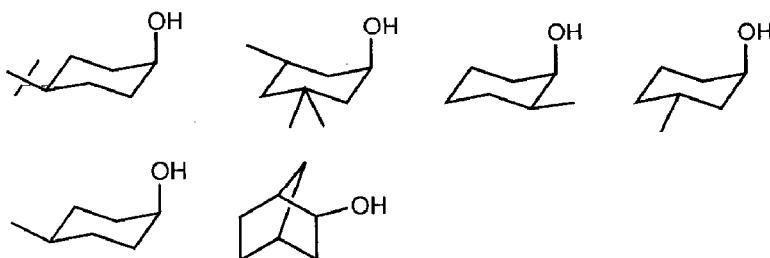
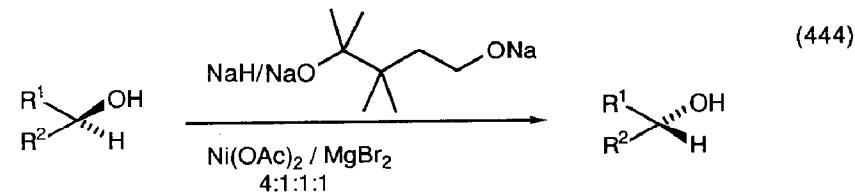
Reduced nickel species isomerized cyclooctadiene to bicyclooctenes (equation 440) [588] and exocyclic cyclooctadienes to tricyclic compounds (equation 441) [589]. Dibromomethyl cyclohexadienones isomerized to tropanones when treated with nickel(0)/zinc reagents (equation 442) [590].



### 5. Miscellaneous Rearrangements

Palladium(0) catalyzed the rearrangement of  $\alpha$ -ketoepoxides to  $\beta$ -diketones (equation 443) [592]. Alcohols were epimerized by nickel complex reducing agents (CRA) (equation 444) [593]. Epoxides epimerized when treated with ruthenium catalysts (equation 445) [594].

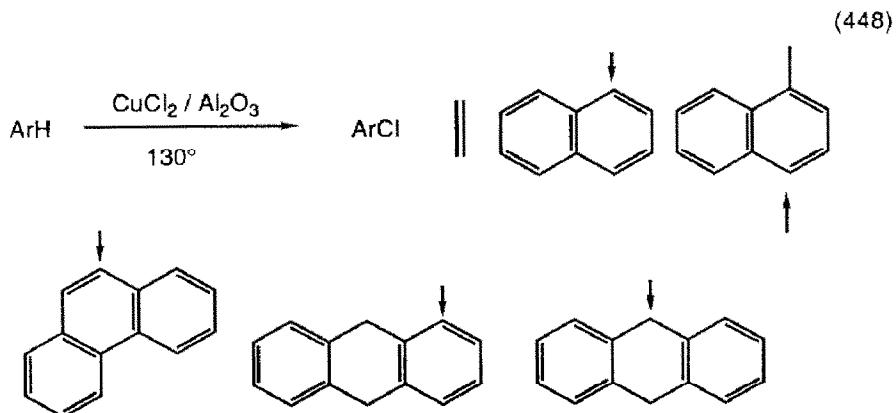
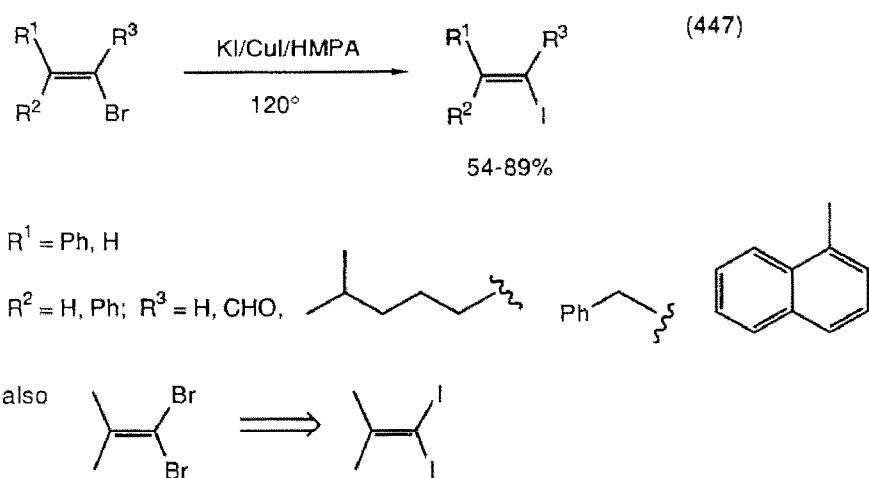
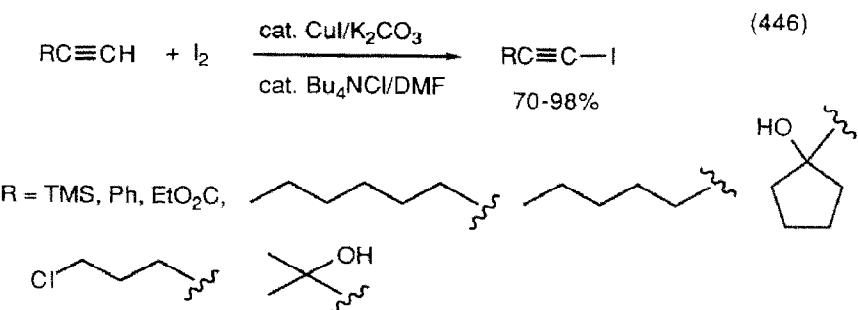


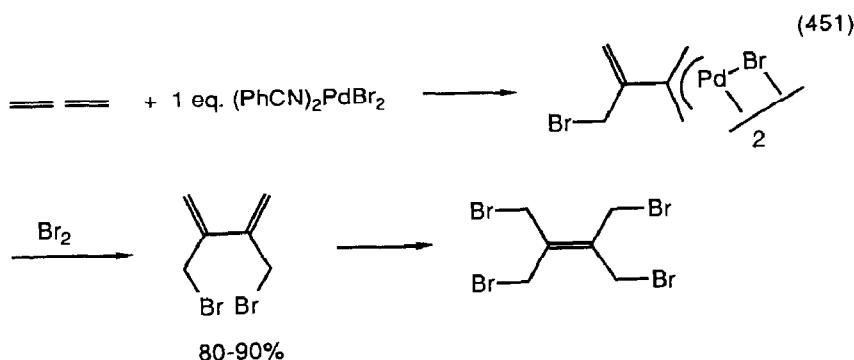
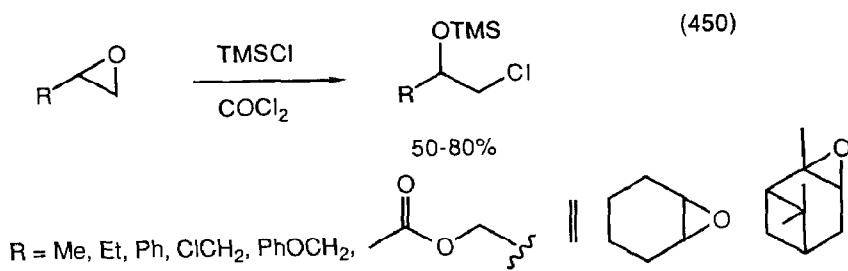
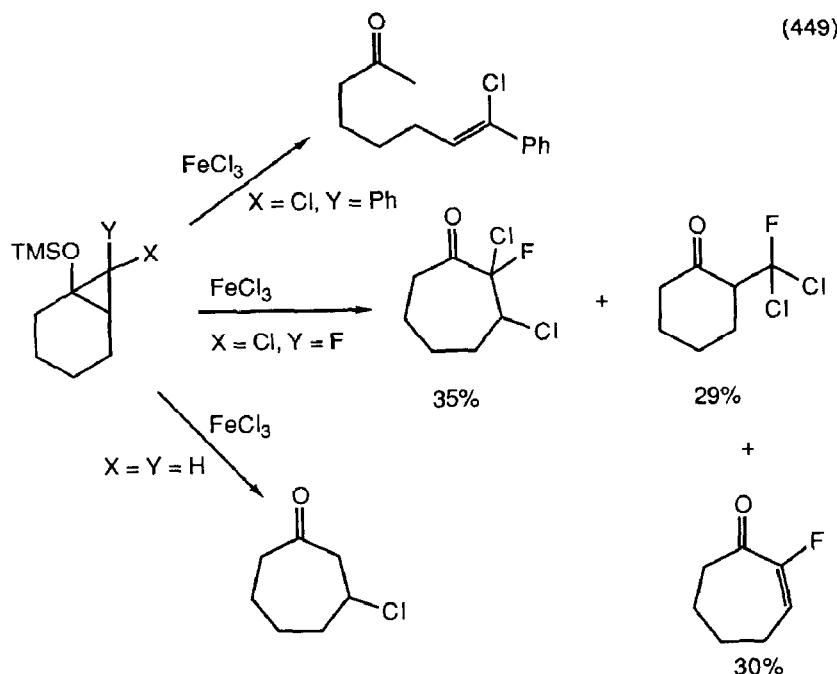


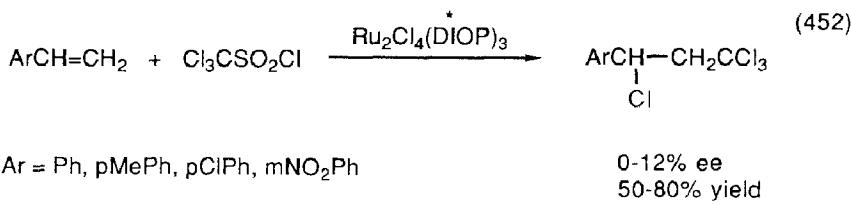
### III. Functional Group Preparations

#### 1. Halides

Terminal alkynes were iodinated by iodine and copper iodide (equation 446) [595]. Vinyl bromides were converted to vinyl iodides by potassium iodide/copper(I) iodide (equation 447) [596]. Toluene was randomly fluorinated by  $\text{CsCoF}_4$  at  $400^\circ$  [597]. Arenes were ring chlorinated by copper(II) chloride on alumina (equation 448) [598]. Iron(III) chloride oxidatively ring opened cyclopropanols with halogenation (equation 449) [559]. Epoxides were opened to trimethylsilyl chlorohydrins by  $\text{TMSCl}$  and cobalt(II) chloride (equation 450) [600]. Allene was coupled and brominated by palladium(II) bromide (equation 451) [601]. Ruthenium complexes catalyzed the addition of trichloromethyl sulfonyl chlorides to olefins (equation 452) [602]. Iron carbonyl telomerized chloroform with ethylene [603].

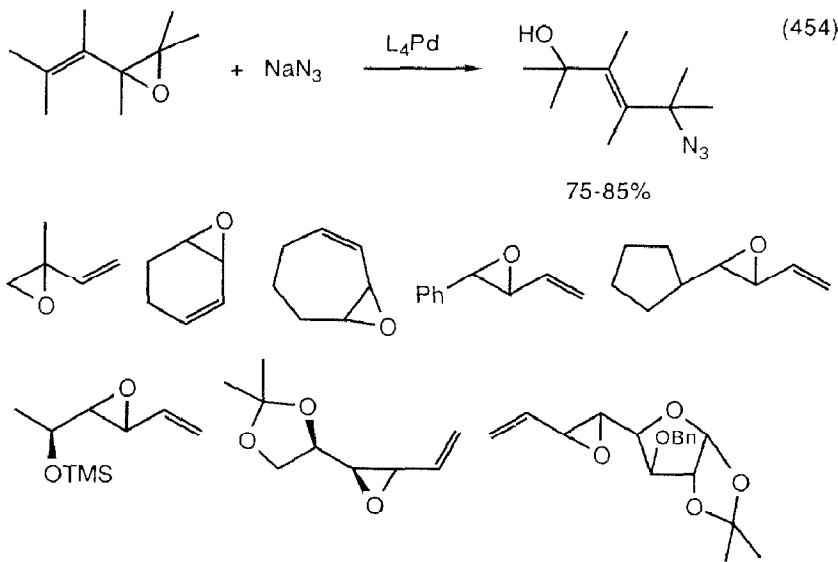
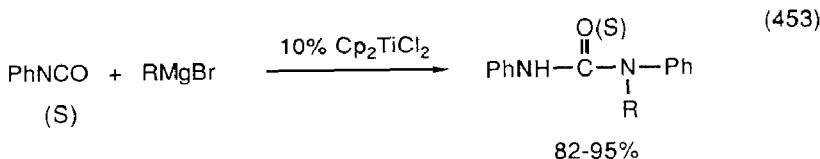




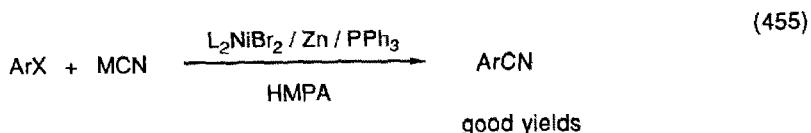


## 2. Amides, Nitriles

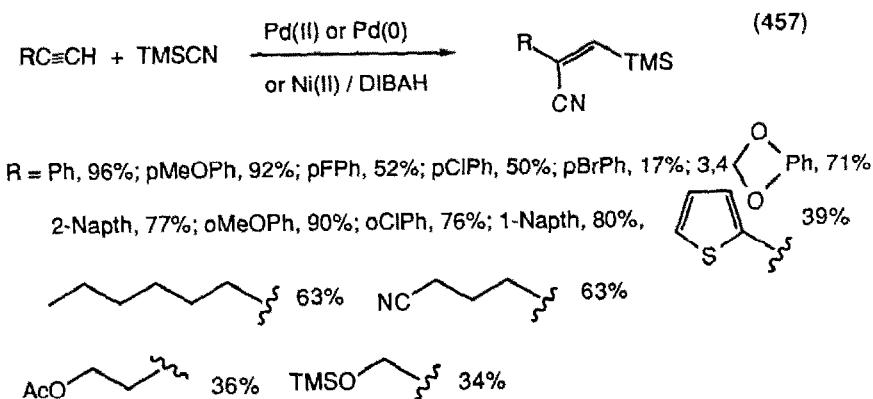
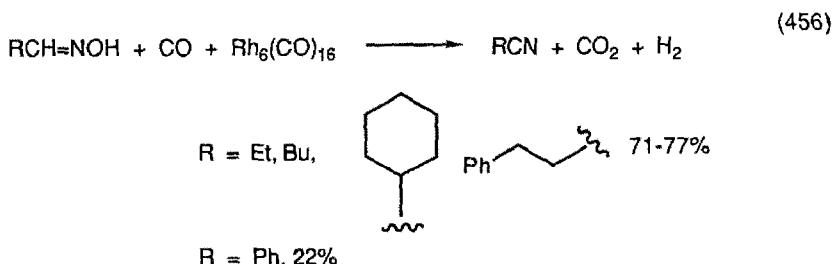
Alcohols were formylated by dimethylformamide by polymer supported phosphine-halogen complexes [604]. Reduced palladium complexes catalyzed the addition of sulfonamides to allylic alcohols, ethers, acetates, and amines to give N-allylsulfonamides [605]. Titanocene dichloride catalyzed the addition of Grignard reagents to isocyanates to give ureas (equation 453) [606]. Palladium(0) complexes catalyzed the reaction of allyl epoxides with sodium azide (equation 454) [607].

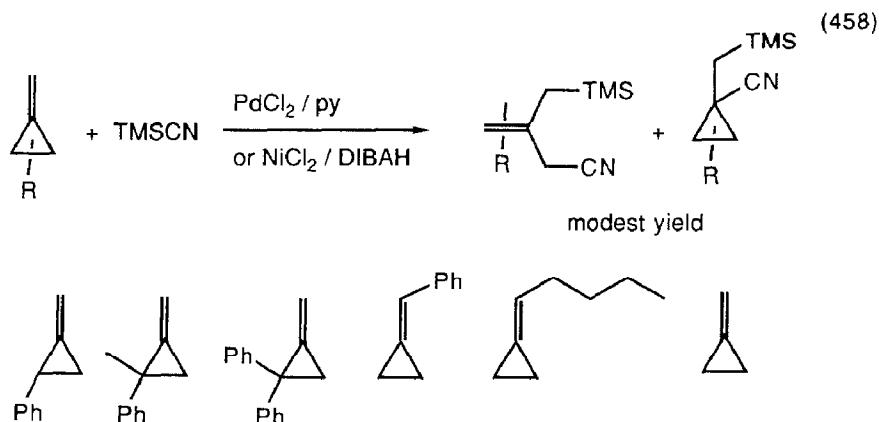


Aryl halides were converted to aryl nitriles by nickel(II) phosphine/zinc/sodium cyanide systems (equation 455) [608]. Aldoximes were converted to nitriles by  $\text{Rh}_6(\text{CO})_{12}$  (equation 456) [609]. Palladium or nickel complexes catalyzed the addition of trimethylsilyl cyanide to terminal alkynes (equation 457) [610], and the ring opening of vinyl cyclopropanes by trimethylsilyl cyanide (equation 458) [611].

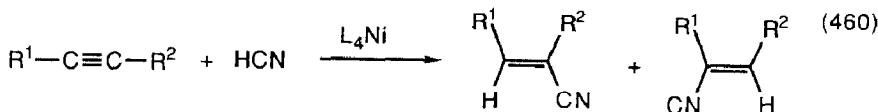
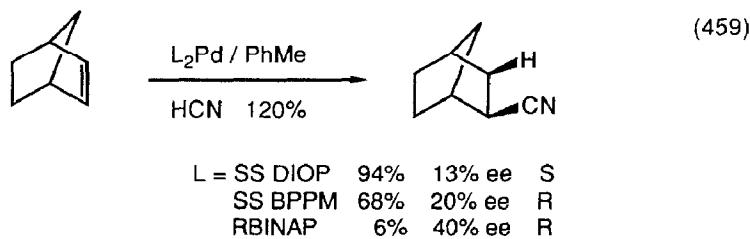


$\text{Ar} = \text{Ph}, p\text{Me}_2\text{NPh}, p\text{MeOPh}, m\text{MeOPh}, o\text{MeOPh}, m\text{MePh}, o\text{MePh}, m\text{FPh},$   
 $p\text{MeO}_2\text{CPh}, m\text{CF}_3\text{Ph}, p\text{CF}_3\text{Ph}, p\text{MeOCPH}, p\text{NCPh}, o\text{NCPh} \parallel p\text{O}_2\text{NPh}$  fails.





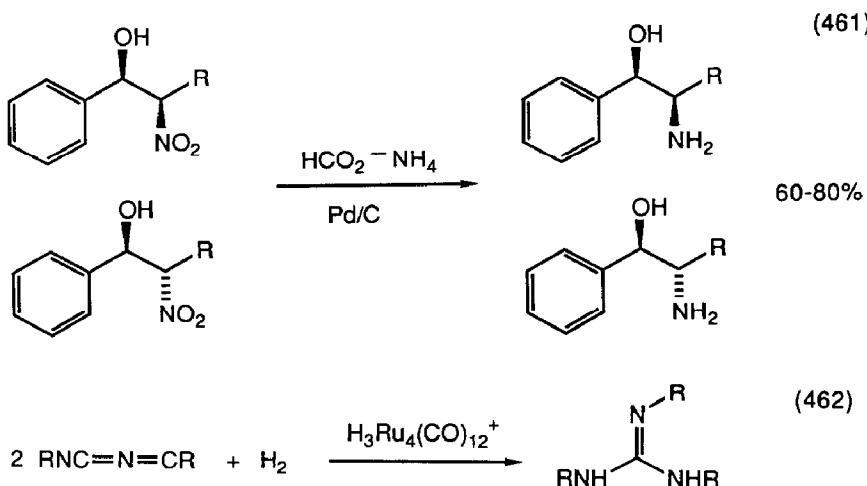
Palladium(0) complexes catalyzed the stereoselective addition of HCN to norbornene (equation 459) [612]. The effects of ligands, and of substituents bearing oxygen or nitrogen on the alkyne, on the nickel-catalyzed addition of HCN to alkynes has been extensively studied (equation 460) [613] [614] [615] [616].



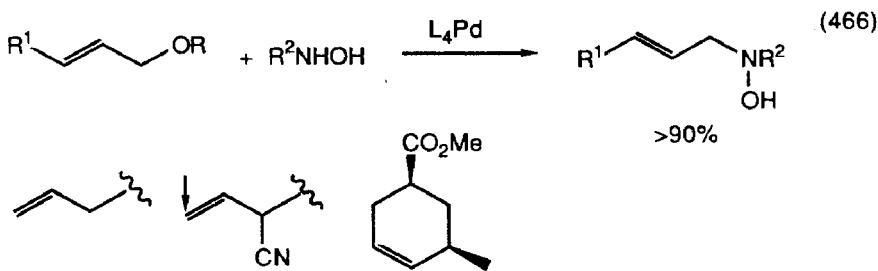
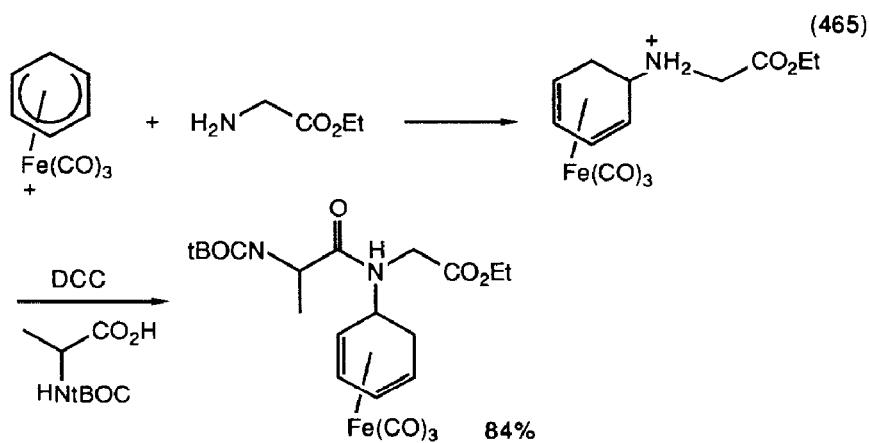
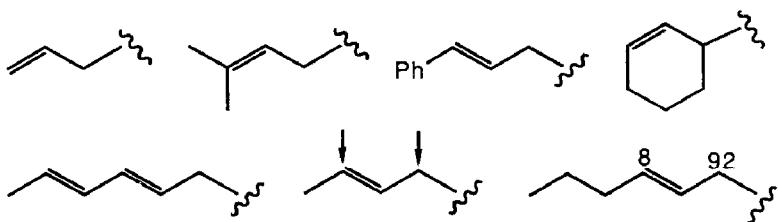
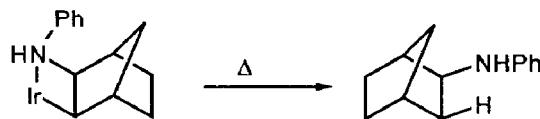
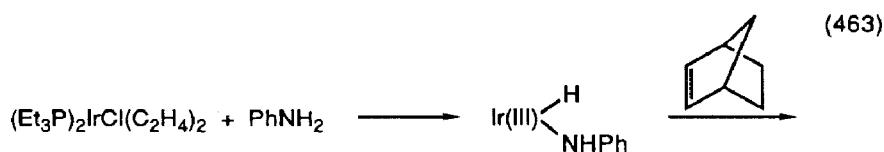
R's = MeOCH<sub>2</sub>, HOCH<sub>2</sub>, etc.

### 3. Amines, Alcohols

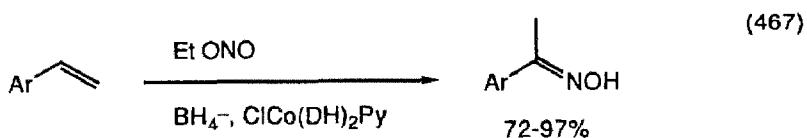
Hydrogenation of aromatic nitro compounds in the presence of transition metal complexes has been reviewed (96 references) [617], as has metal catalyzed deoxygenation reactions by carbon monoxide of nitroso and nitro compounds (328 references) [618]. Aromatic nitro compounds were reduced to anilines by carbon monoxide NaOH/H<sub>2</sub>O/benzene in the presence of RCCo<sub>3</sub>(CO)<sub>9</sub> clusters [619]. Nitrobenzene was reduced to aniline by HFe(CO)<sub>4</sub><sup>-</sup>, a reagent which tolerated PhCHO, PhCOCl and PhCH<sub>2</sub>Cl without reducing them [620].  $\alpha$ -Hydroxynitro compounds were reduced to amino alcohols with no loss of stereochemistry (equation 461) [621]. Carbodimides were reduced to amidines by ruthenium hydride catalysts (equation 462) [622].



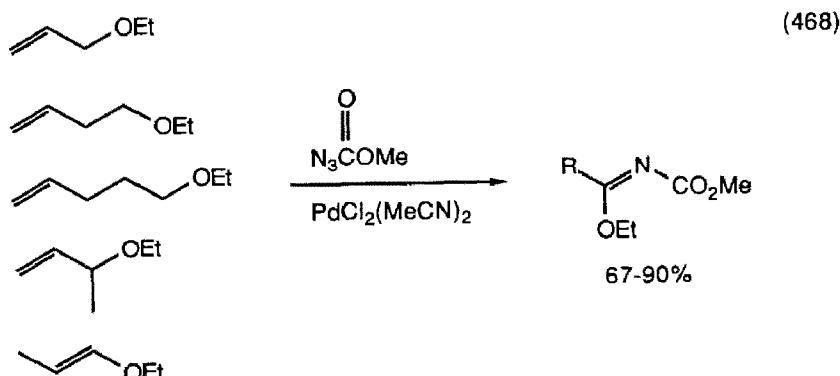
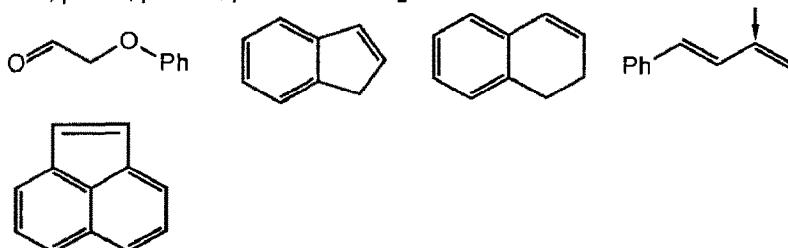
Rhodium complexes catalyzed the amination of olefins [623]. Stepwise activation of N-H was used to amine norbornene (equation 463) [624]. Allyl acetates were aminated by *bis* benzyloxycarbonyl amides (equation 464) [625]. Cationic iron dienyl complexes were readily aminated (equation 465) [626]. Allyl ethers and acetates were aminated by hydroxylamines in the presence of palladium(0) catalyst (equation 466) [627].

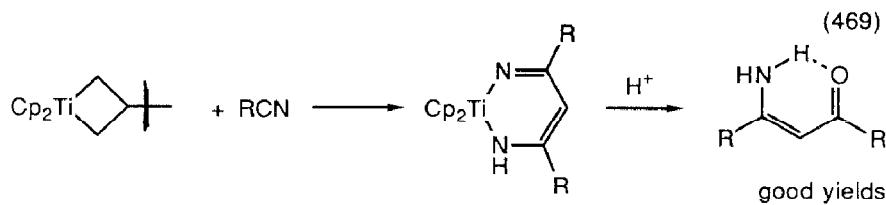


Aryl amines were methylated by methanol in the presence of ruthenium trichloride catalysts at 180° [628]. Styrenes were converted to benzyl oximes by ethyl nitrile in the presence of cobalt catalysts (equation 467) [629]. Benzyl amines were oxidized to imines of benzaldehyde by iodosyl benzene in the presence of RuCl<sub>2</sub>L<sub>3</sub> catalysts [630]. A variety of unsaturated ethers were converted to N-acylimidates by reaction with methyl azidoformate in the presence of palladium(II) catalysts (equation 468) [631]. Titanocyclobutanes dimerized nitriles to β-iminoketones (equation 469) [632]. Chiral α-amino acids were synthesized using a chiral nickel template (equation 470) [633]. Nitriles inserted into Fischer carbene complexes to give imidato carbenes which could be alkylated (equation 471) [634].

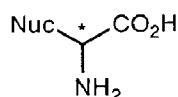
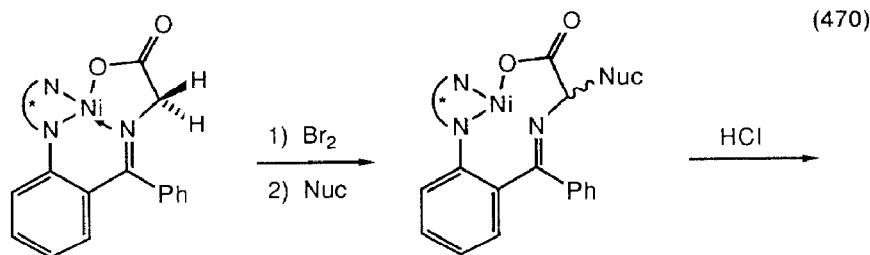


$\text{Ar} = \text{Ph}, \text{pClPh}, \text{pMePh}, \text{pMeOPh}, \text{mNO}_2\text{Ph}, \text{oBrPh}, \text{oClPh}, \text{oMePh},$





R = Me, tBu, Ph, pMePh, pMeOPh, pCF<sub>3</sub>Ph, C<sub>6</sub>F<sub>5</sub>

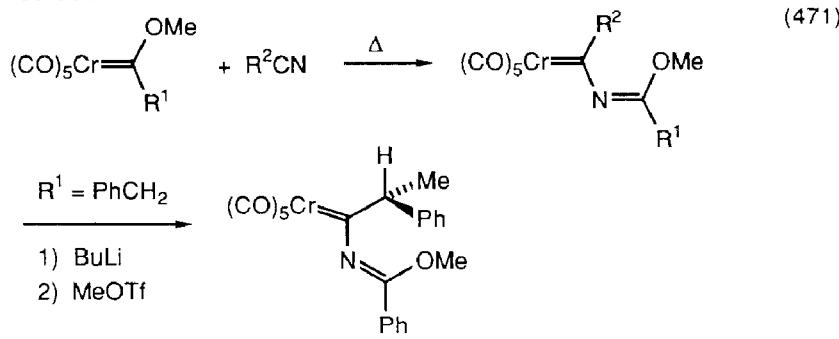


Nuc = MeO<sup>-</sup>, PhO<sup>-</sup>, Me<sub>2</sub>NH, CH<sub>2</sub>(CO<sub>2</sub>Et)<sub>2</sub>

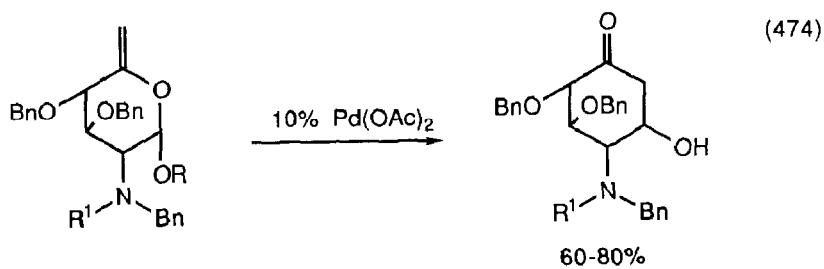
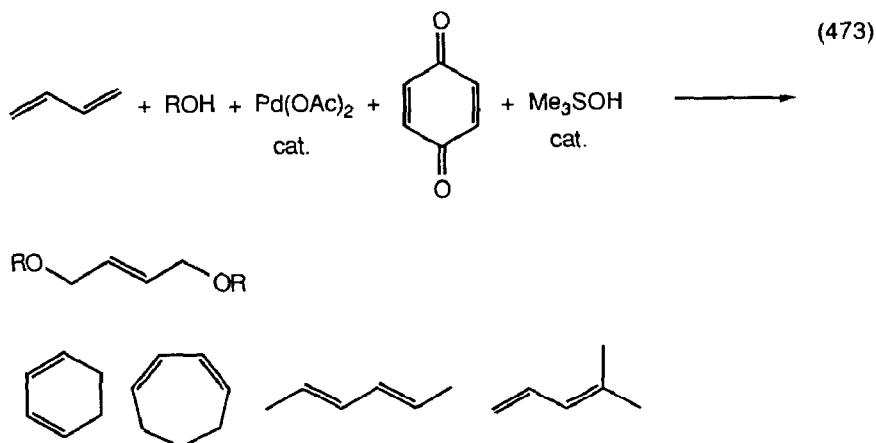
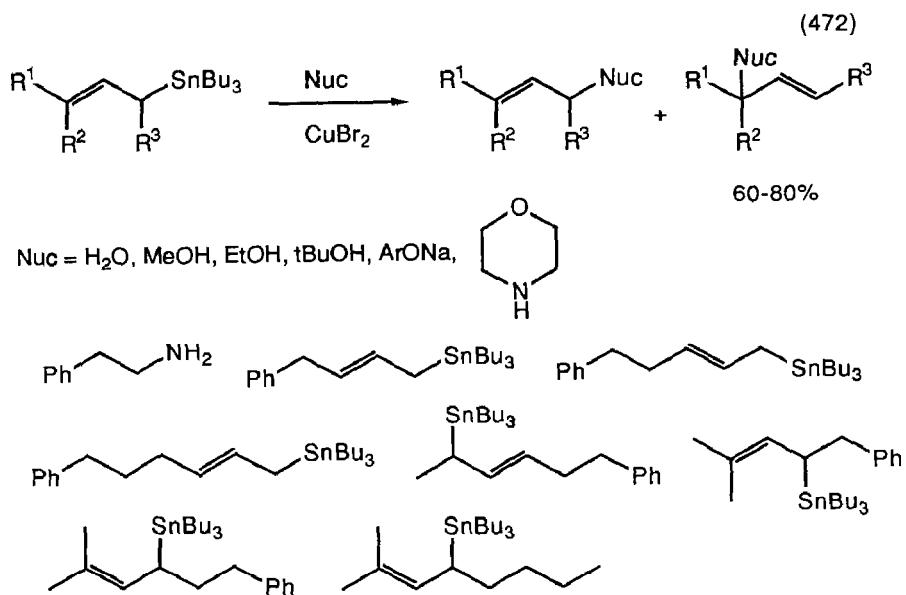
L-stereochemistry

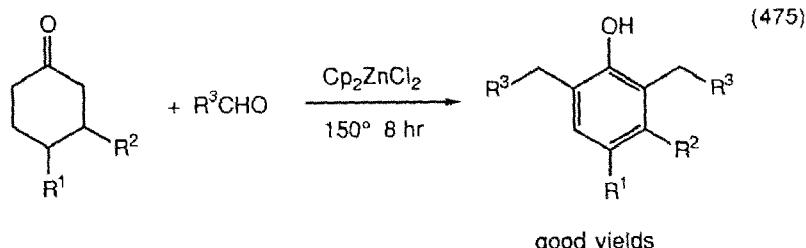
>90:10

60-90%



Allyl tin reagents were aminated and alkoxylated in the presence of copper(II) bromide (equation 472) [635]. Dienes were 1,4-bis-alkoxylated using palladium(II) catalysts and catalytic amounts of methanesulfonic acid (equation 473) [636]. Palladium acetate rearranged cyclic enol ethers to hydroxy ketones (equation 474) [637]. Zirconium dichloride catalyzed the condensation of aldehydes with cyclohexanone (equation 475) [638]. Aldohexoses were degraded to pentaols by Wilkinson's catalyst (equation 476) [639].

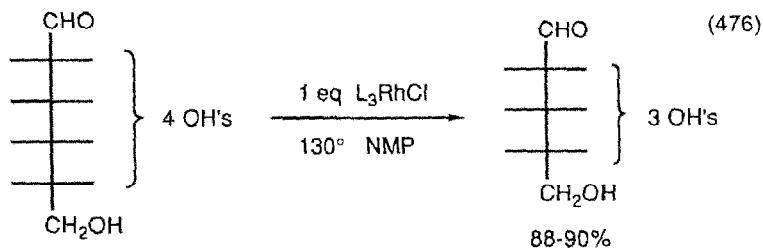
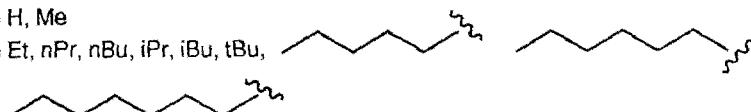




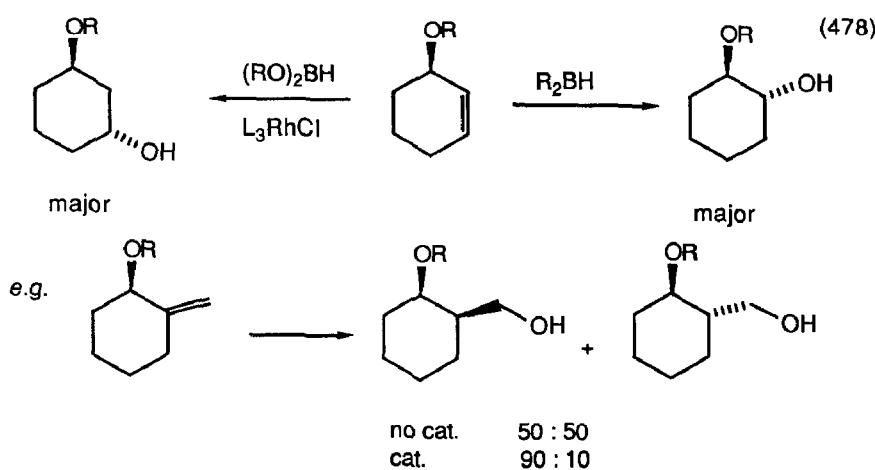
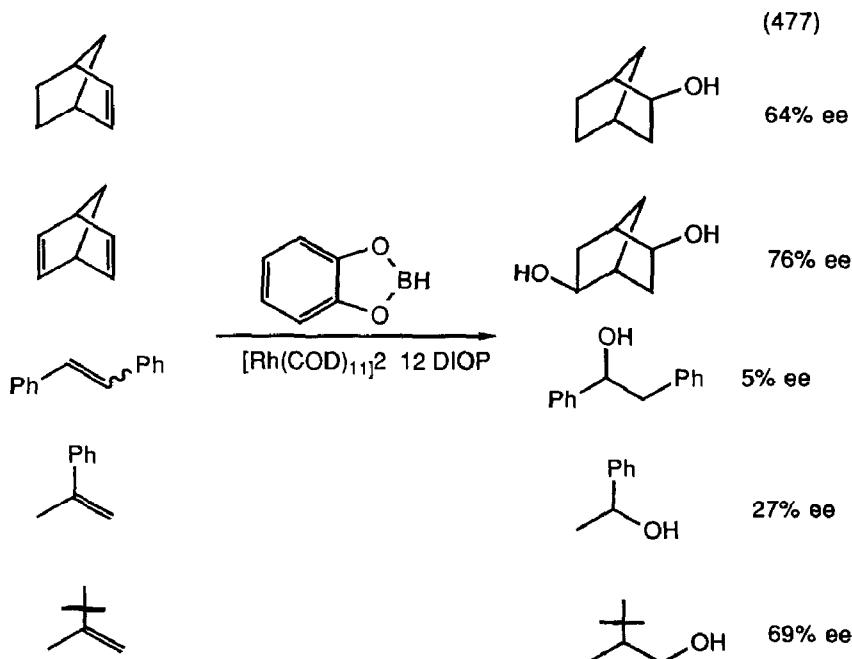
$\text{R}^1 = \text{H, Me, tBu}$

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

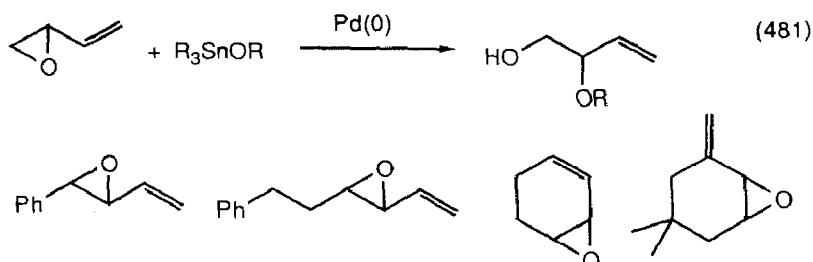
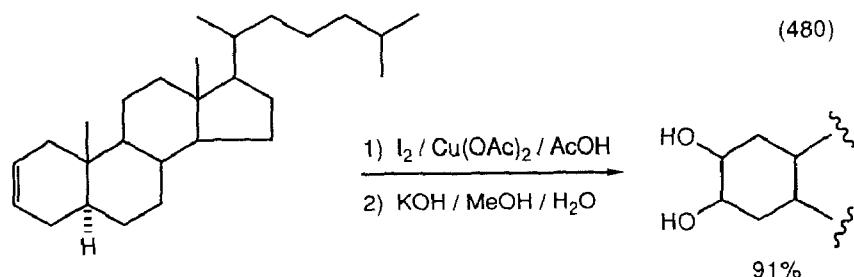
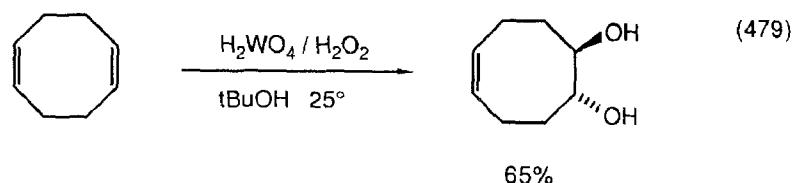
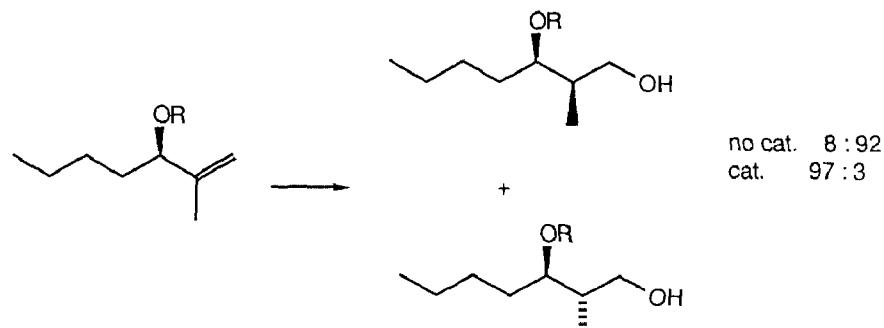
$\text{R}^3 = \text{Et, nPr, nBu, iPr, iBu, tBu}$



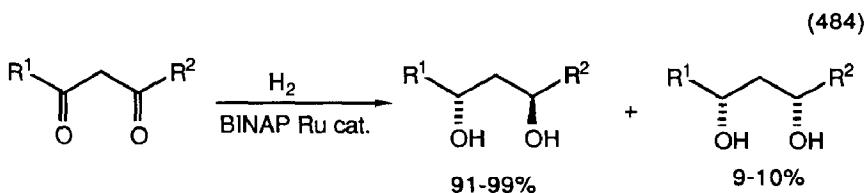
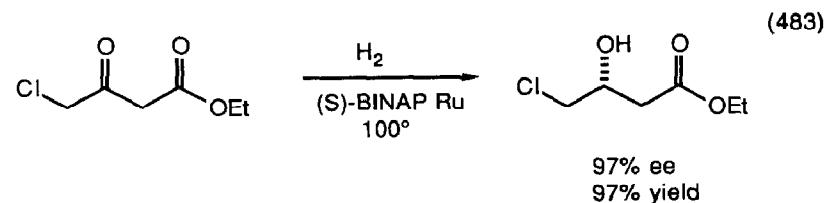
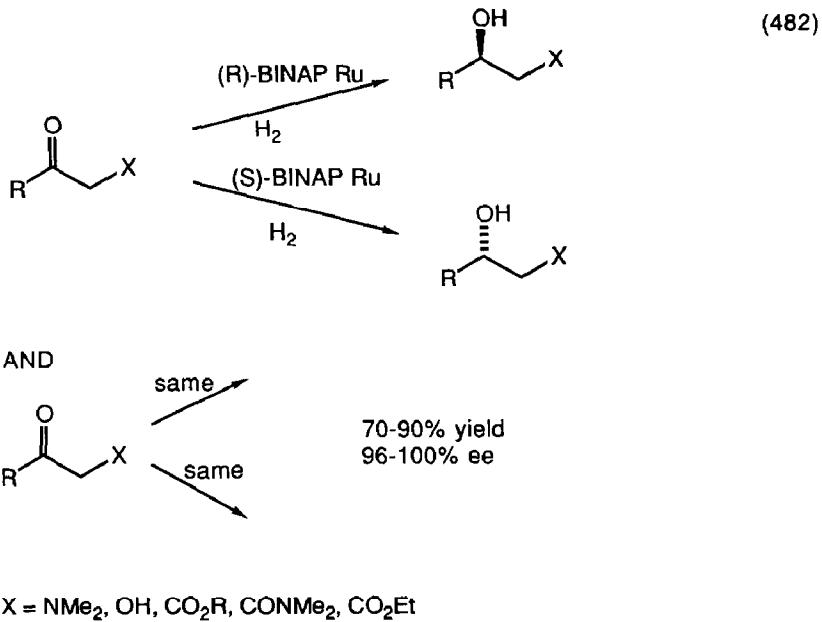
Chiral rhodium(I) complexes catalyzed the hydroboration of alkenes with decent enantiomeric excesses (equation 477) [640]. Allylic ethers were hydroborated with different regiochemistry in the presence of rhodium(I) catalysts (equation 478) [641]. 1,5-Cyclooctadiene was oxidized to the *trans* monodiol by tungsten acid/hydrogen peroxide (equation 479) [642]. Steroid A-ring olefins were oxidized to the diol by iodine/copper acetate (equation 480) [643]. Allyl epoxides were converted to diol monoethers by reaction with tin alkoxides in the presence of palladium(0) catalysts (equation 481) [644].



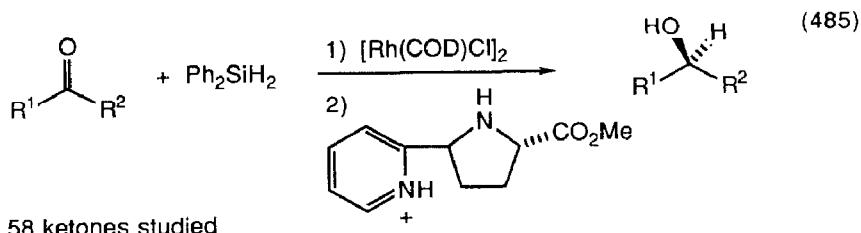
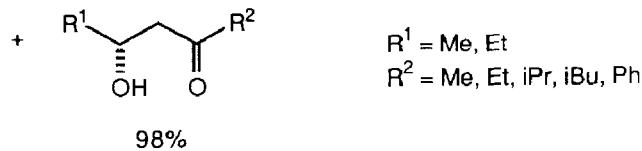
(equation 478 continued)



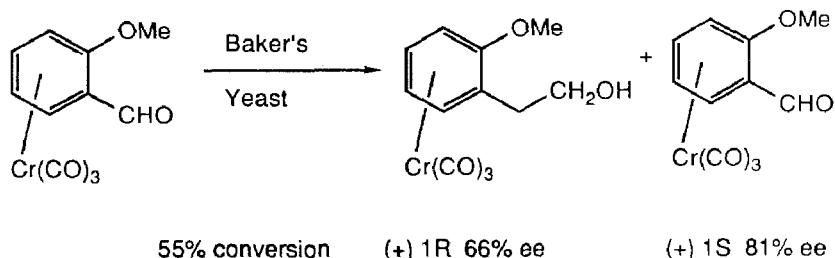
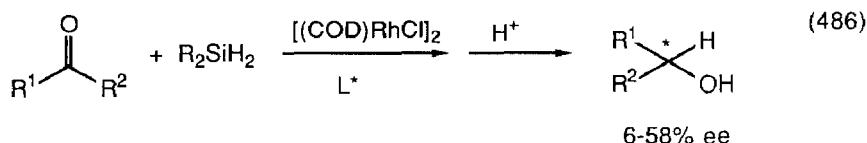
Ruthenium binaph catalysts reduced  $\alpha$ - and  $\beta$ -functionalized ketones to alcohols with high enantiomeric excesses (equation 482) [645], (equation 483) [646], (equation 484) [647]. Rhodium(I) complexes catalyzed the asymmetric hydrosilylation of ketones (equation 485) [648], (equation 486) [649]. Baker's yeast resolved chromium complexed aromatic aldehydes (equation 487) [650] and reduced complexed phenyl ketones (equation 488) [651] with high enantioselectivity. Chiral alcohols were resolved by kinetically consuming one enantiomer with a chiral catalyst (equation 489) [652], (equation 490) [653].

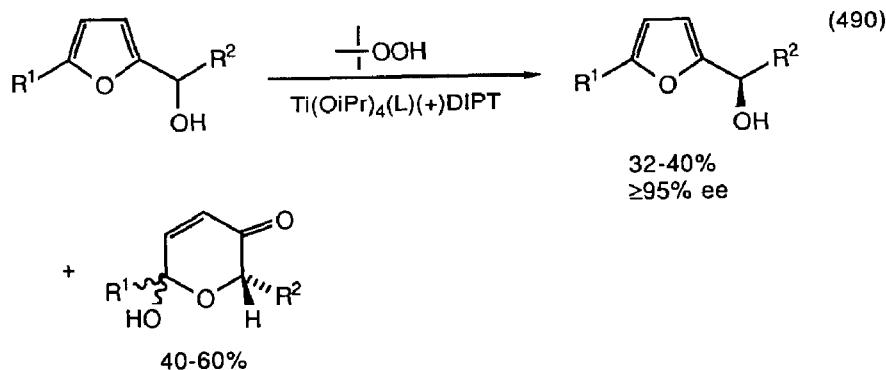
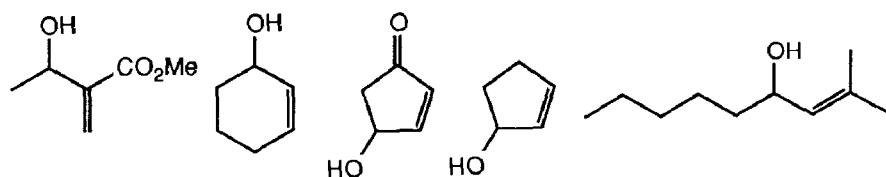
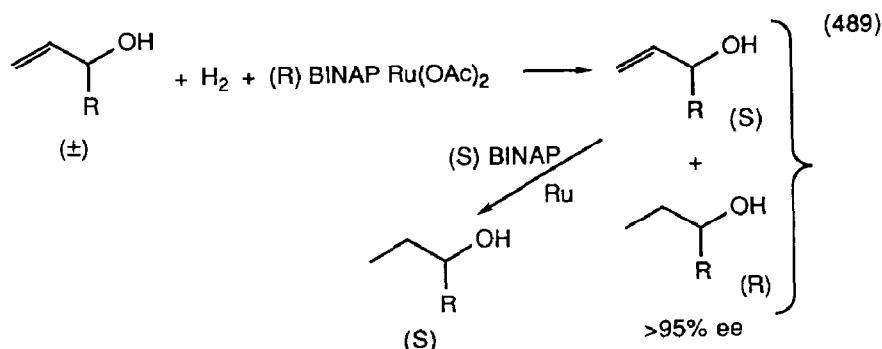
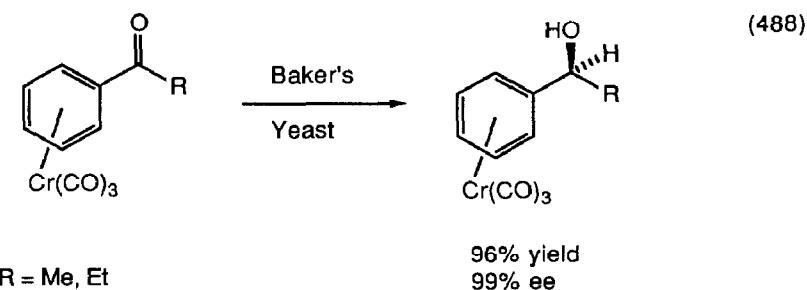


(equation 484 continued)



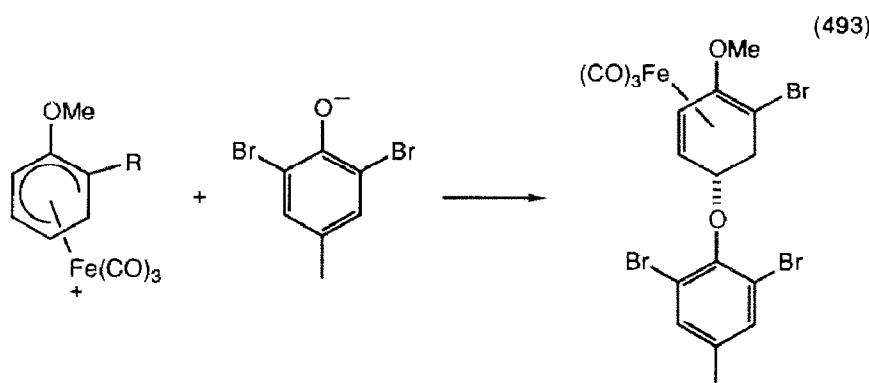
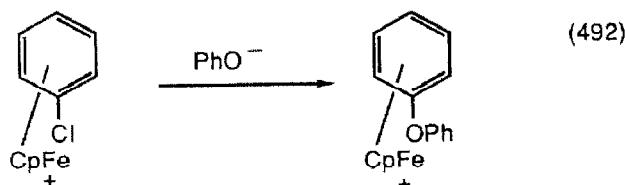
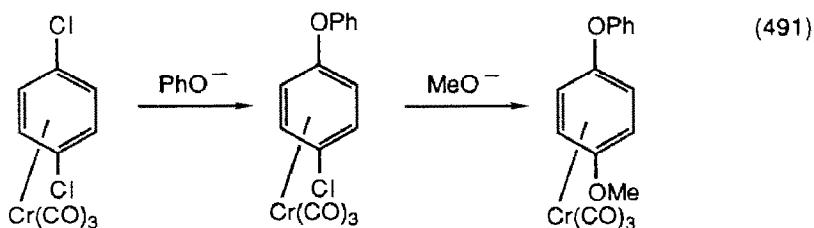
$R^1 = \text{Ar}; R^2 = \text{Me}; >80\% \text{ ee}$   
 $R^1 = \text{hetAr}; R^2 = \text{Me}; >80\% \text{ ee}$   
 $R^1 = \text{alkyl}; R^2 = \text{Me}; \sim 50\% \text{ ee}$

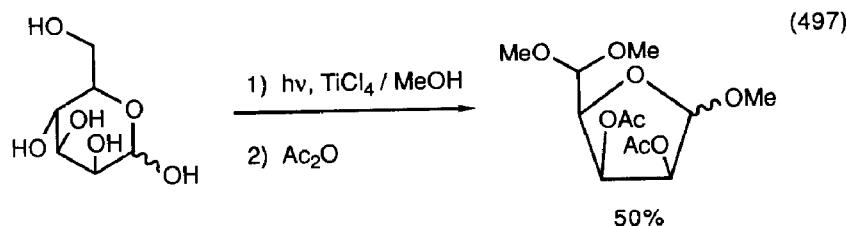
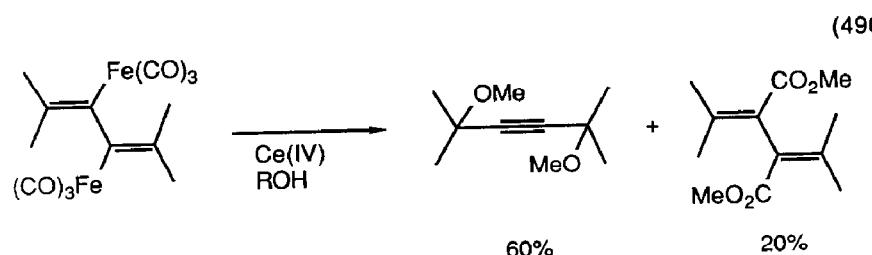
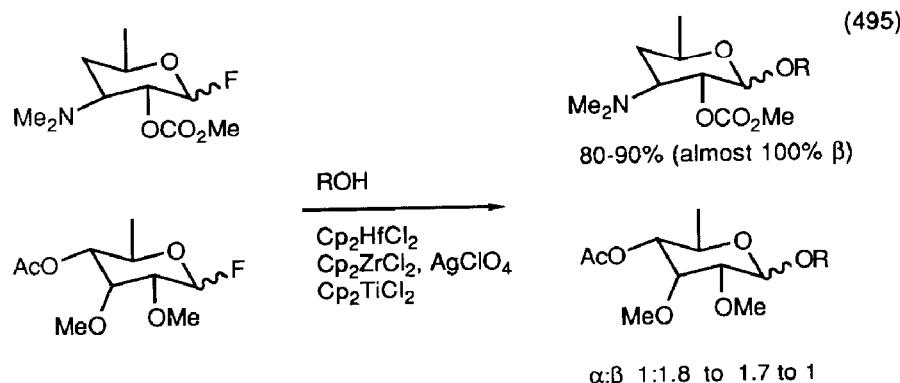
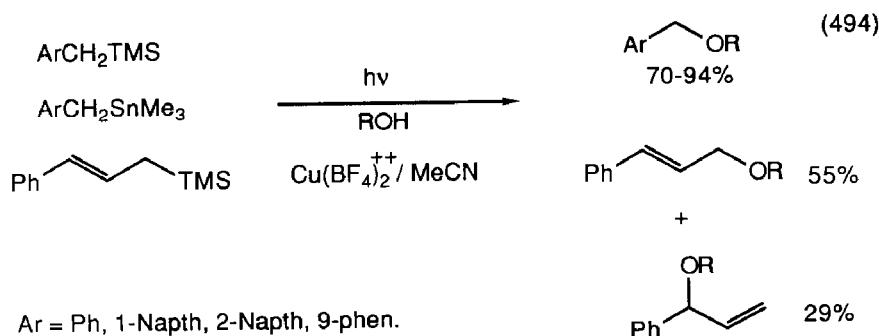


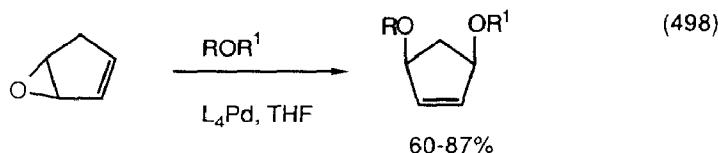


#### 4. Ethers, Esters, Acids

Metal complexed aryl halides underwent reaction with alkoxides to produce aryl ethers (equations 491, 492, and 493) [654]. Benzyl and allylsilanes were converted to ethers by photolysis in alcohol in the presence of copper(II) (equation 494) [655]. 1-Fluoroglycosides were alkoxylated by treatment with alcohols and titanium, zirconium or hafnum cationic complexes (equation 495) [656] [657]. Cerium(IV) oxidation of vinyliron complexes produced propargyl ethers (equation 496) [658]. Sugars were selectively methylated by irradiation in methanol in the presence of titanium(IV) chloride (equation 497) [659]. Allyl epoxides were converted to 1,4-diethers by reaction with siloxanes and palladium(0) catalysts (equation 498) [660].



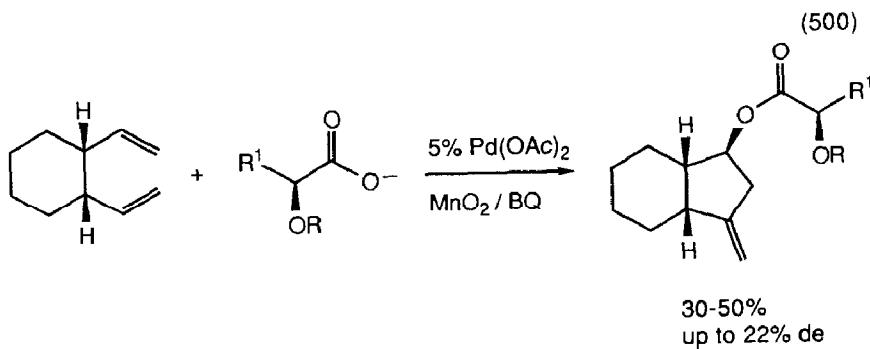
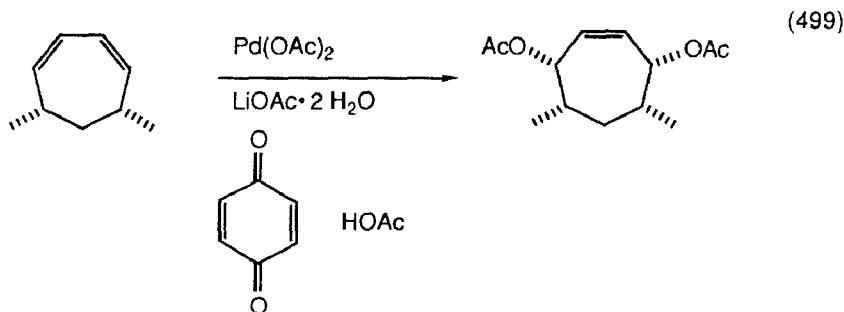


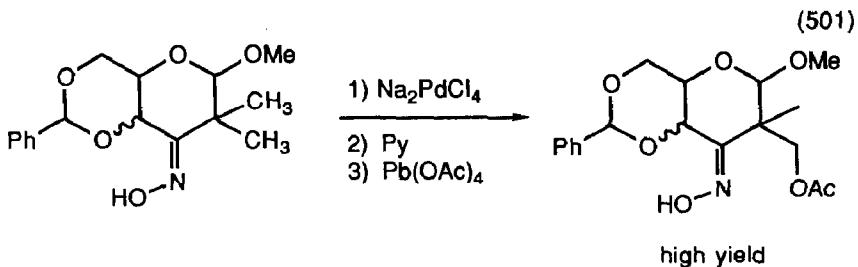


$R = \text{TMS, Et}_3\text{Si, PhMe}_2\text{Si}$

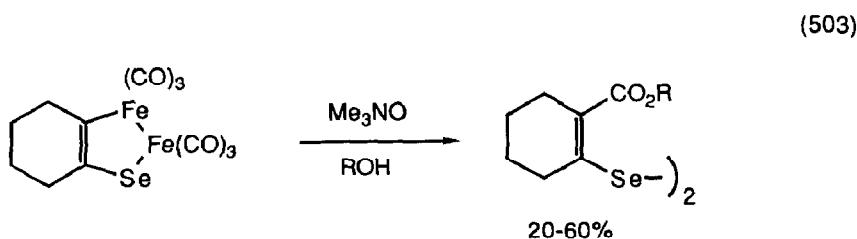
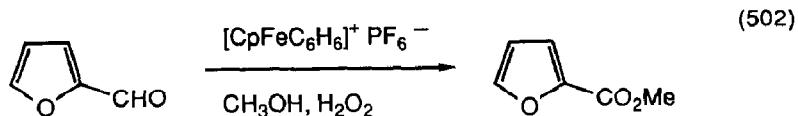
$R^1 = \text{Ac, PhCH}_2, \text{Ph, pNO}_2\text{Ph}$

The spectroscopic and physiochemical properties of the catalyst in the  $\text{Cu}(\text{OAc})_2\text{-LiBr}_2$  catalyzed diacetoxylation of butadiene were examined [661]. Palladium catalyzed homogenous diacetoxylation of butadiene was studied yet again [662]. Palladium(II) catalyzed the stereospecific diacetoxylation of dimethyl cyclohepta-1,3-diene (equation 499) [663].  $\pi$ -Allylpalladium-quinone complexes were involved in the *bis* acetoxylation of butadiene [664]. 1,5-Dienes were acetoxylated and cyclized by palladium(II) catalysts (equation 500) [665]. An unactivated methyl group was acetoxylated by ortho palladation/oxidation (equation 501) [666].

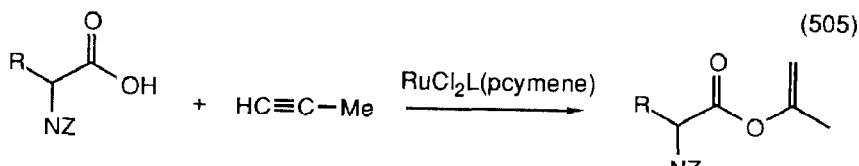
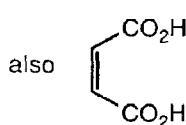
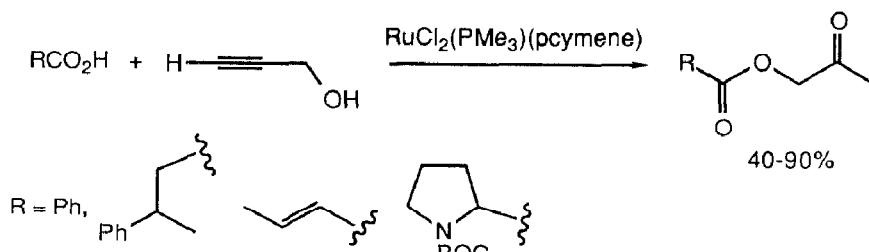




Iron complexes catalyzed the oxidation of furfural (equation 502) [667]. Oxidation of a vinyl iron species produced the corresponding ester (equation 503) [668]. Ruthenium complexes catalyzed the addition of carboxylic acids to propargyl alcohols to give keto esters (equation 504) [669]. The addition of amino acids gave enol acetates (equation 505) [670]. Photolysis of methyl acrylate and methyl acetate or propionate in the presence of rhodium(I) phosphine complexes resulted in very unusual addition products (equation 506) [671]. Cyclic ketones were oxidatively cleaved to diacids by oxygen and rhenium catalysts (equation 507) [672].

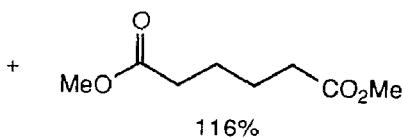
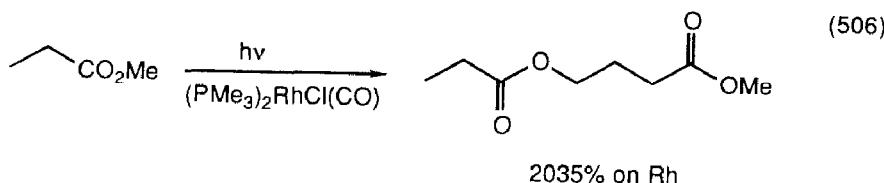


(504)

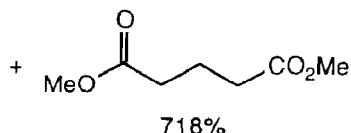


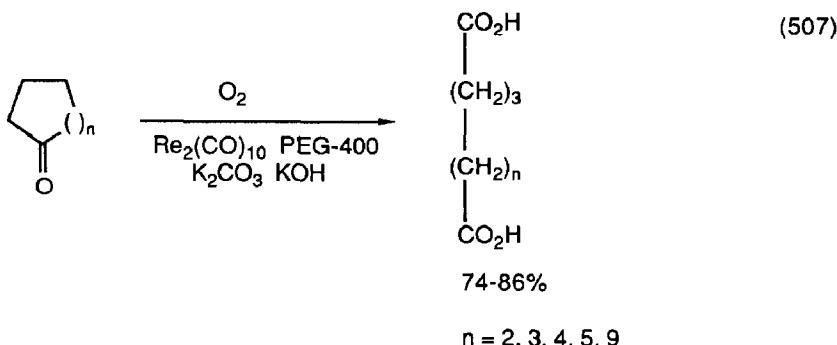
$\text{R} = \text{H}, \text{Me}, \text{PhCH}_2$ ; also proline, aspartine

56-80%



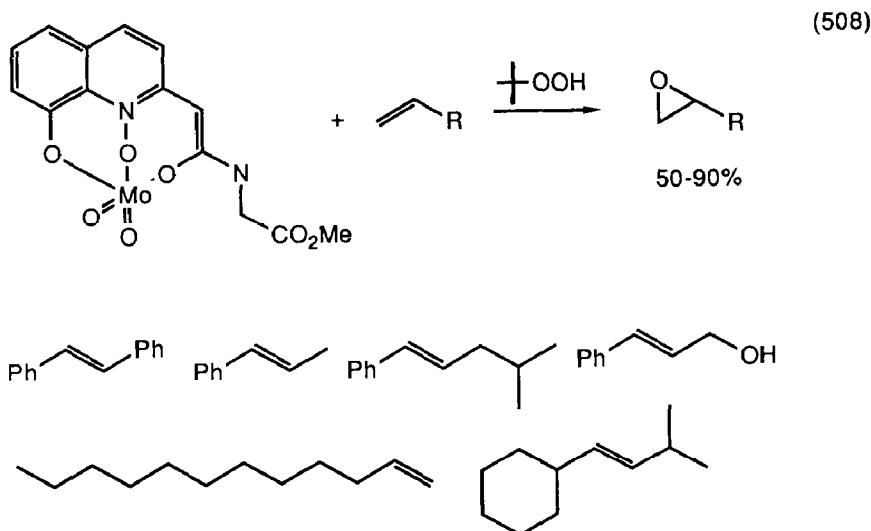
and

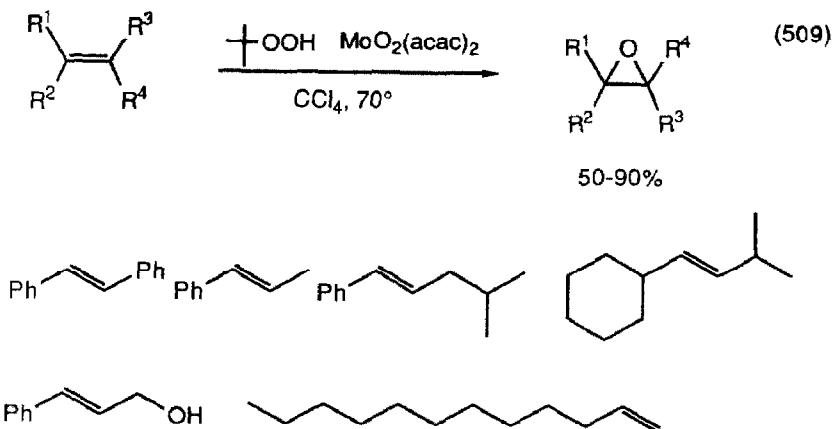




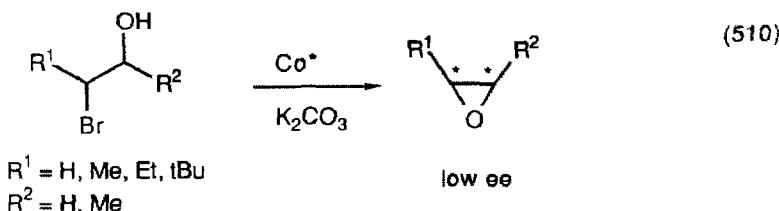
## 5. Heterocycles

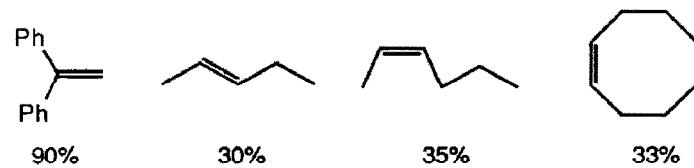
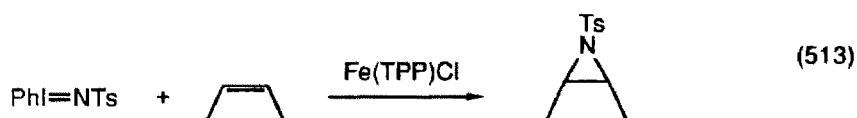
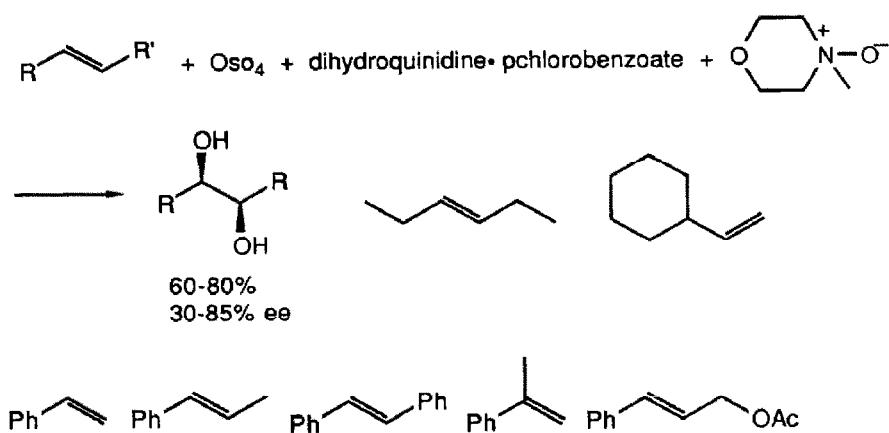
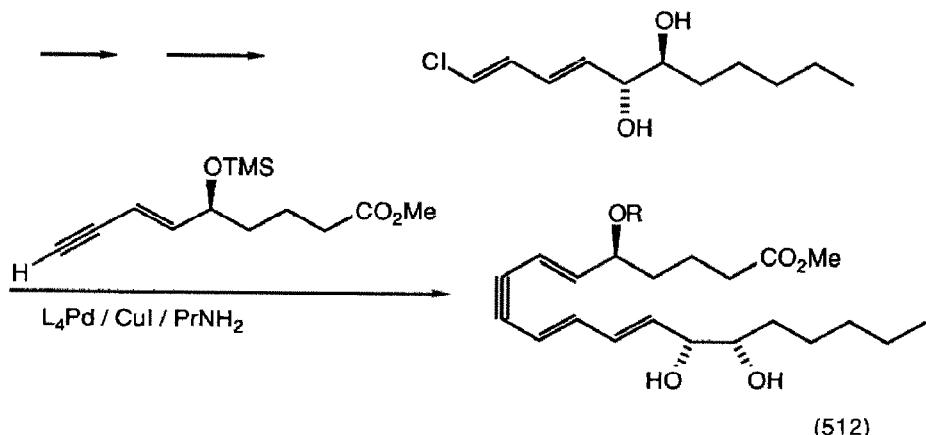
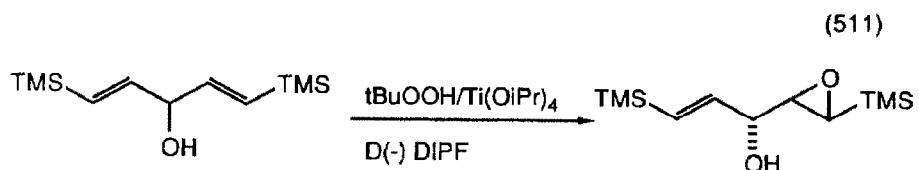
Epoxidation of olefins continues to be extensively explored. A review with no references dealing with catalytic epoxidation has appeared [673]. The kinetics of oxygen transfer in the ruthenium catalyzed epoxidation of olefins by iodosyl benzene have been studied [674]. Cis-dioxomolybdenum species with tetradentate ligands were effective epoxidation catalysts [675]. Immobilized molybdenum complexes were used to catalyze the epoxidation of  $\alpha$ -methyl styrene [676], and cyclohexene [677], while molybdenum carbide powder catalyzed the epoxidation of 1-octene [678], as did molybdenum-containing heterogeneous catalysts [679]. Propylene dimers were epoxidized [680]. Iodosylbenzene epoxidized alkenes and terpenes in the presence of vanadyl acetylacetone [681], and a variety of other metal complexes (symposium report) [682]. Molybdenum oxo catalysts efficiently epoxidized olefins by t-butylhydroperoxide (equation 508) [683], (equation 509) [684].



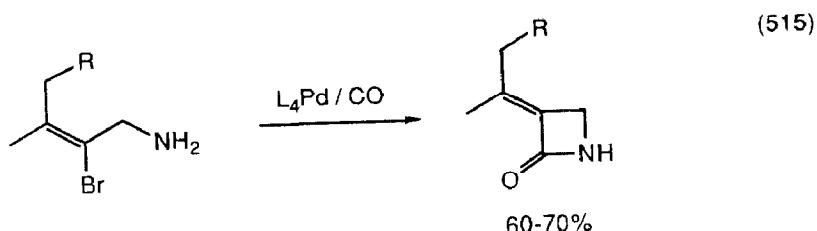
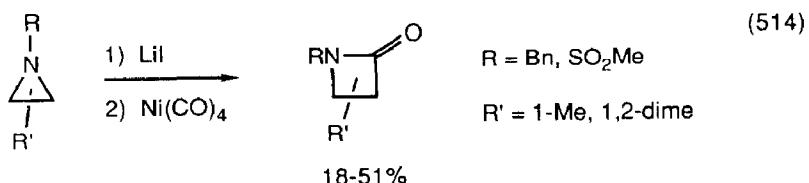


Two reviews dealing with asymmetric epoxidation have appeared [685] (31 references) [686] (15 references). Chiral cobalt catalysts cyclized bromohydrins to epoxides with only low ee's at reasonable conversion (equation 510) [687]. Asymmetric epoxidation was used in the synthesis of polyunsaturated acids (equation 511) [688]. Sharpless epoxidation of methyl E-7-hydroxy-5-heptenoate proceeded with high ee's [689]. Sodium tungstate catalyzed the epoxidation of half esters of maleic acids with asymmetric centers [690]. Nickel(II) complexes of chiral cyclams catalyzed alkene epoxidation in low yields and with no ee's [691]. Alkenes were asymmetrically *cis* hydroxylated by OsO<sub>4</sub> in the presence of chiral amines (equation 512) [692]. N-Tosylaziridines were synthesized by iron-porphyrin catalyzed reaction of alkenes with PhI-NTs (equation 513) [693].

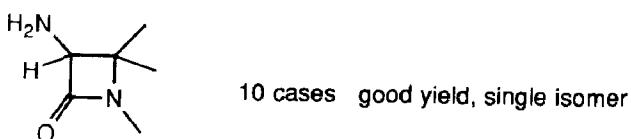
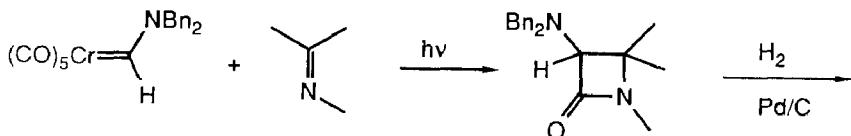
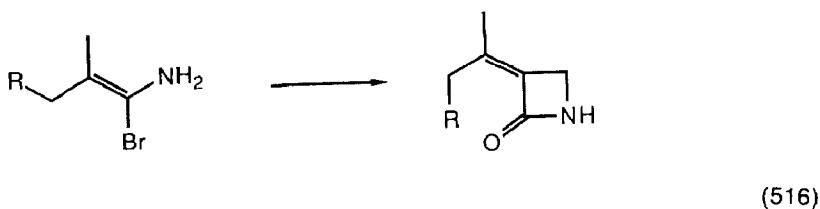


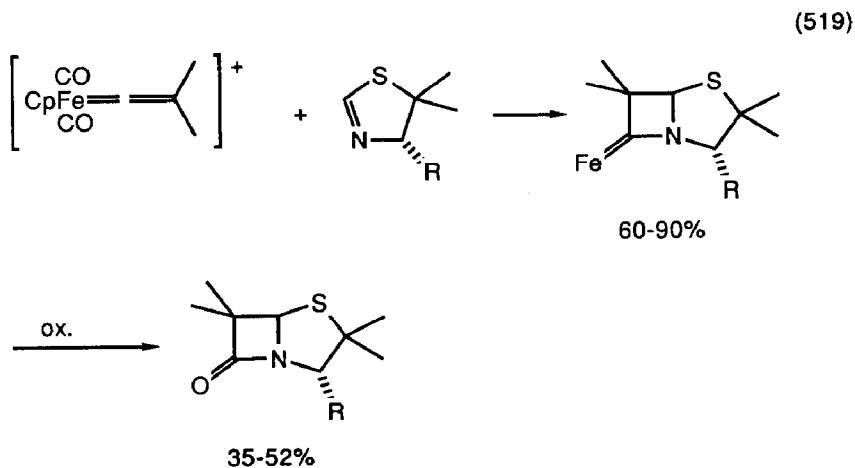
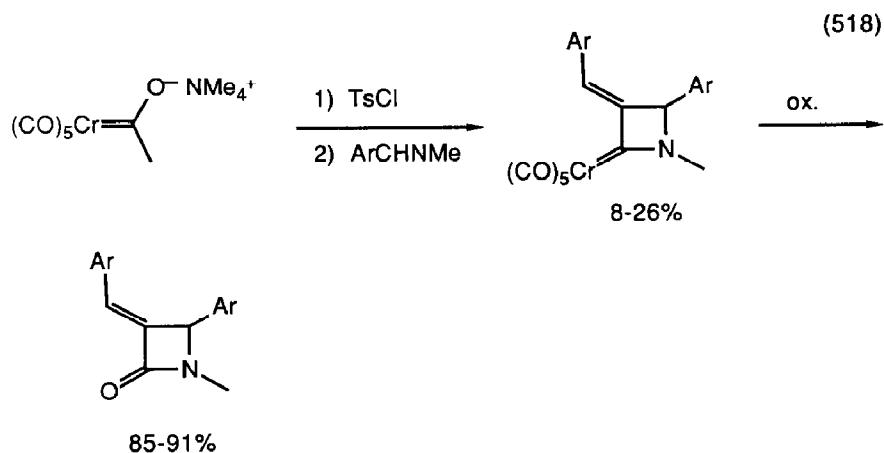
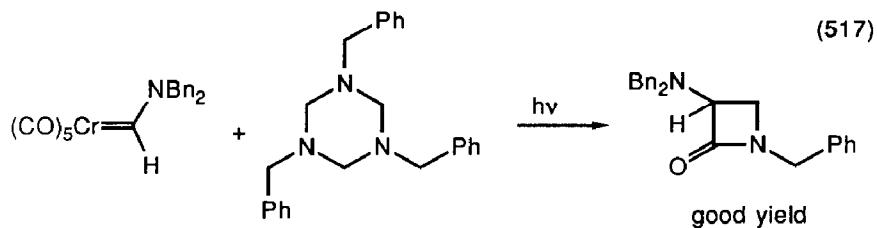


The application of organometallic reagents in  $\beta$ -lactam chemistry has been reviewed (100 references) [694]. Aziridines were carbonylated to  $\beta$ -lactams by nickel carbonyl (equation 514) [695].  $\beta$ -Bromo allyl amines underwent carbonylative cyclization to  $\beta$ -lactams using palladium(0) catalysts (equation 515) [696].  $\beta$ -Lactams were prepared by the photolytic reaction of chromium amino carbene with imines (equation 516) [697]. The same complexes underwent reactions with S-1,3,5-triazines to give monocyclic  $\beta$ -lactams (equation 517) [699]. Chromium carbene (equation 518) and iron vinylidene complexes condensed with imines to give  $\beta$ -lactams after oxidative removal of the metal (equation 519) [699].

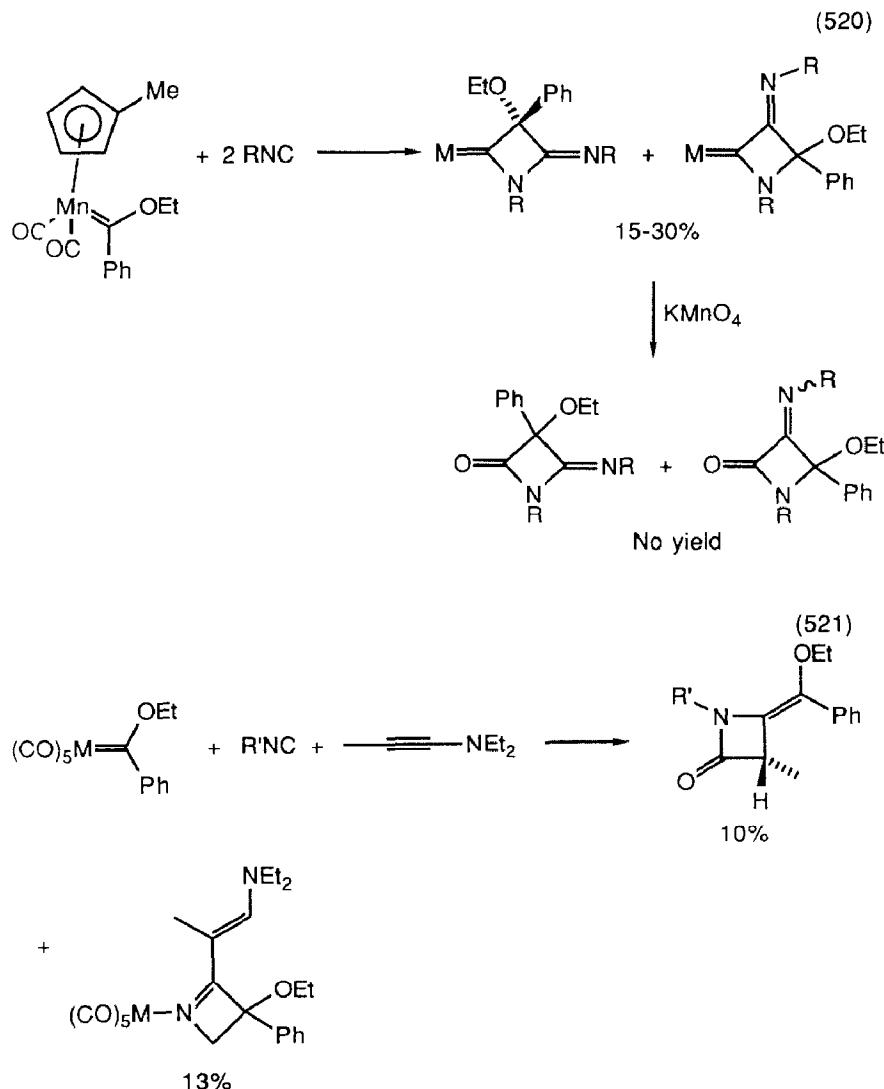


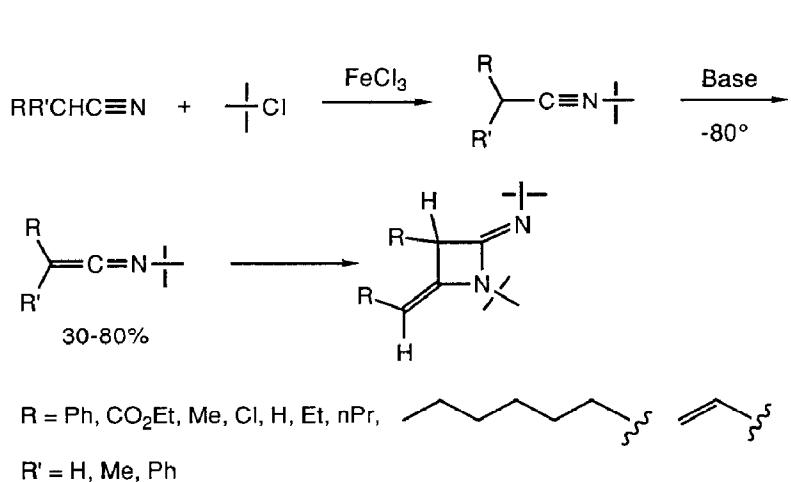
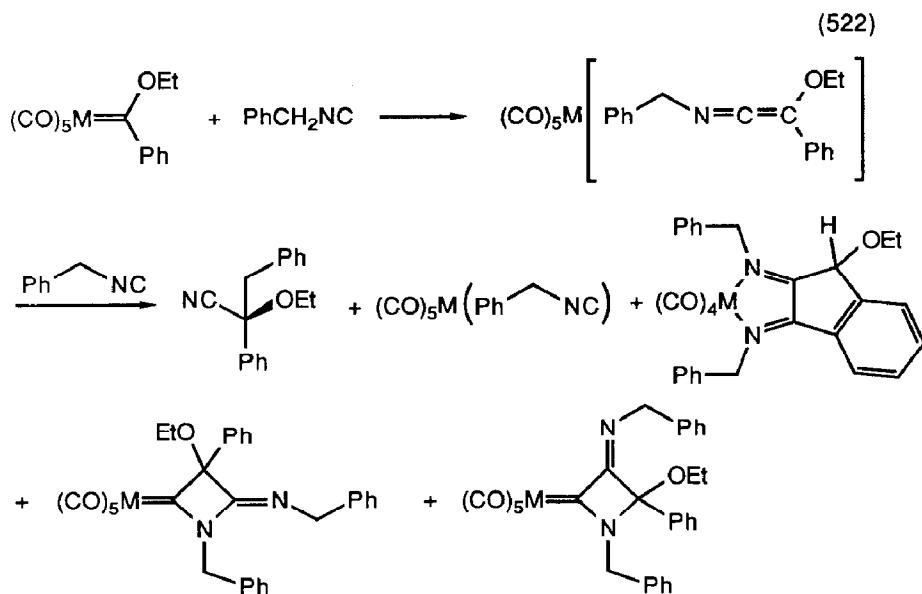
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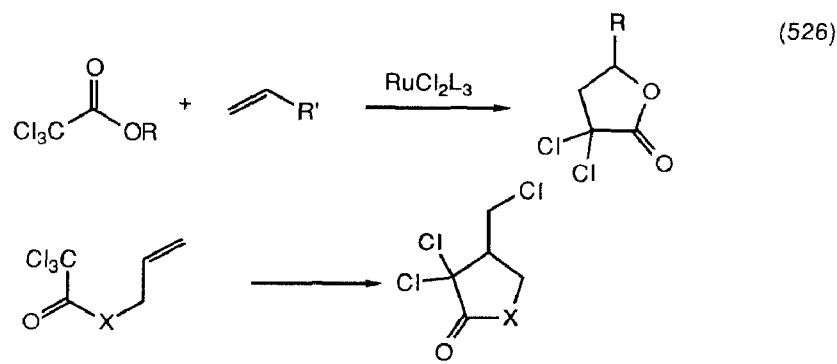
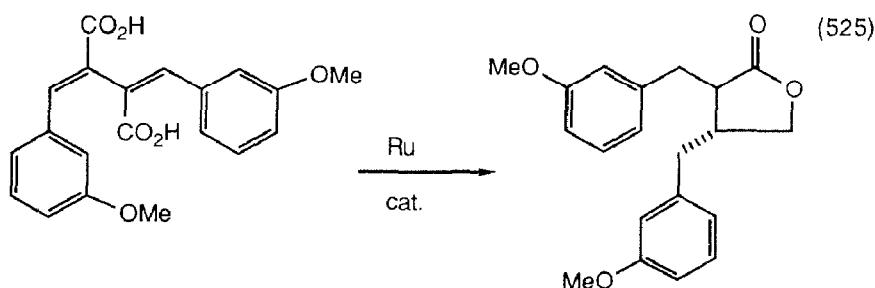
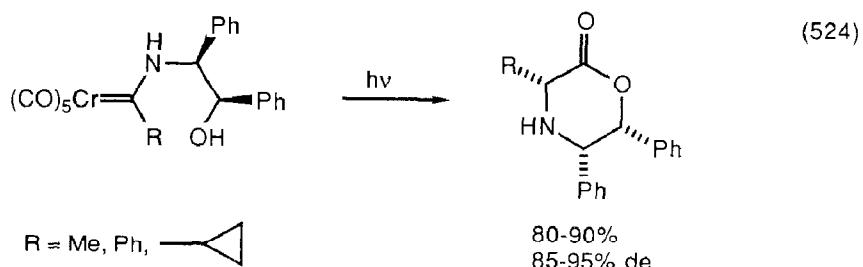
"Ketenimine complexes from carbene complexes and isocyanides: versatile building blocks for carbocycles and heterocycles" was the title of a recent review (59 references) [700]. Manganese carbenes underwent reaction with isocyanides to produce complexes which were oxidized to 4-imino-2-azetidinones (equation 520) [701]. Group(VI) alkoxy carbenes combined with isonitriles and ynamines to give  $\beta$ -azetidinones in low yield (equation 521) [702], as well as more complex carbene complexes (equation 522) [703]. Iron(III) chloride catalyzed a Ritter reaction to give nitrilium salts which dimerized (equation 523) [704].



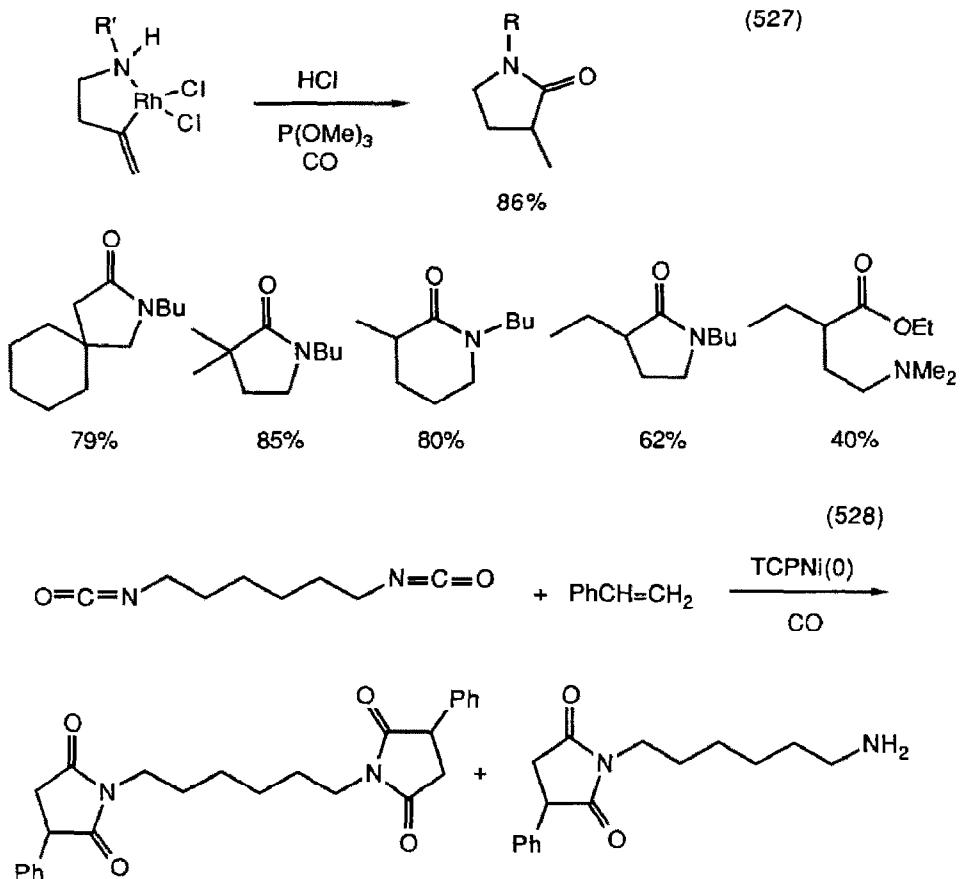


Photolysis of chiral amino carbene complexes of chromium having a pendant hydroxyl group produced amino lactones convertible to amino acids, in high yield and diastereomeric excess (equation 524) [705]. Enterolactone was synthesized by a ruthenium catalyzed reaction (equation 525) [706]. Ruthenium(II) chloride complexes catalyzed the addition of trichloroacetic acid esters to olefins to give lactones (equation 526) [707]. Rhodium(I) complexes of homoallyl amines underwent facile cyclocarbonylation to lactams (equation 527)

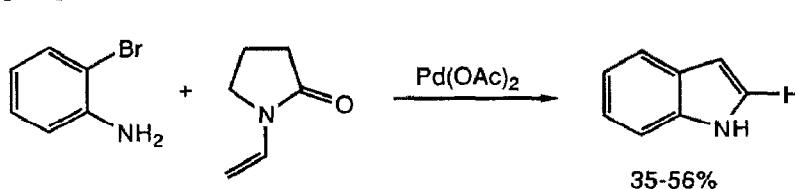
[708]. A cobalt mediated route to acromelic acid A has been reported [709]. Nickel(0) complexes catalyzed the reaction of styrene with *bis*-isocyanates to produce *bis*-imides (equation 528) [710].

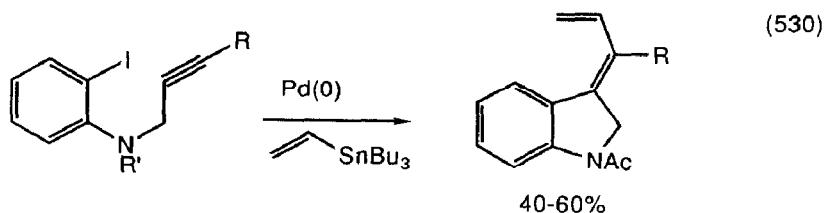


$X = O, N$

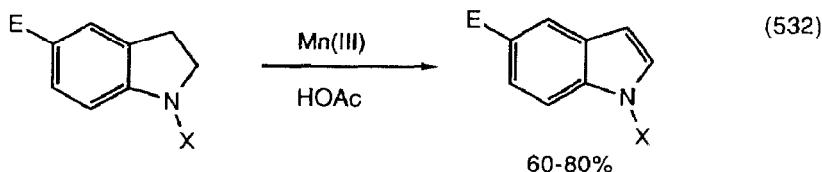
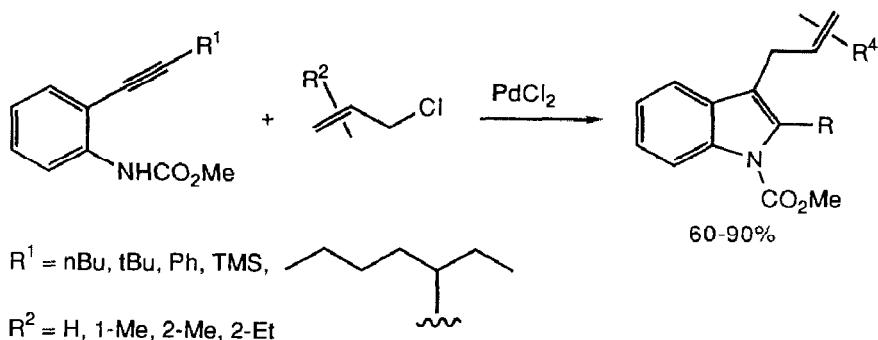
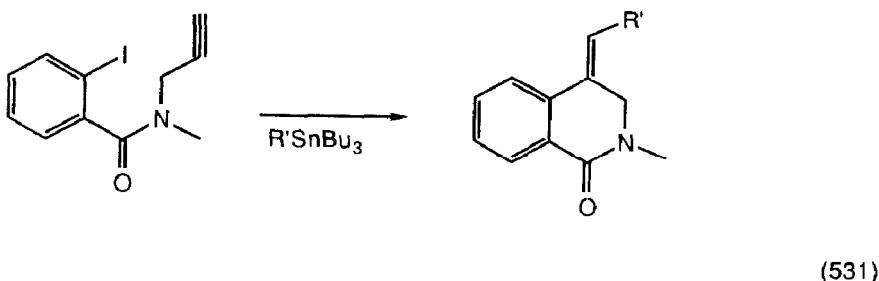


Transition metals in the synthesis and functionalization of indoles has been reviewed (111 references) [711]. Palladium(II) acetate catalyzed the reaction of 2-bromoanilines with enamines to give indole (equation 529) [712]. Palladium(0) complexes catalyzed the cyclization of o-iodopropargyl anilines to indoles (equation 530) [713]. Palladium(II) chloride catalyzed the reaction of allyl chlorides with o-acetylenic anilines to give indoles (equation 531) [714]. Manganese(III) salts catalyzed the oxidation of indolines to indoles (equation 532) [715].



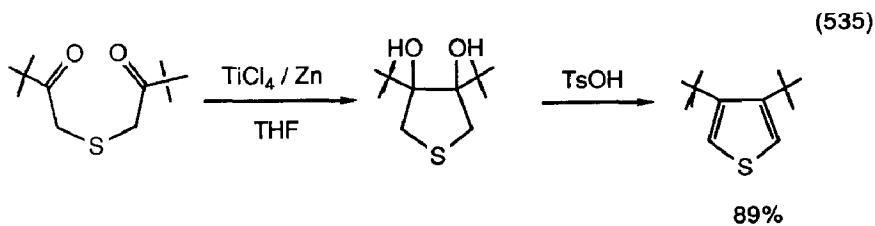
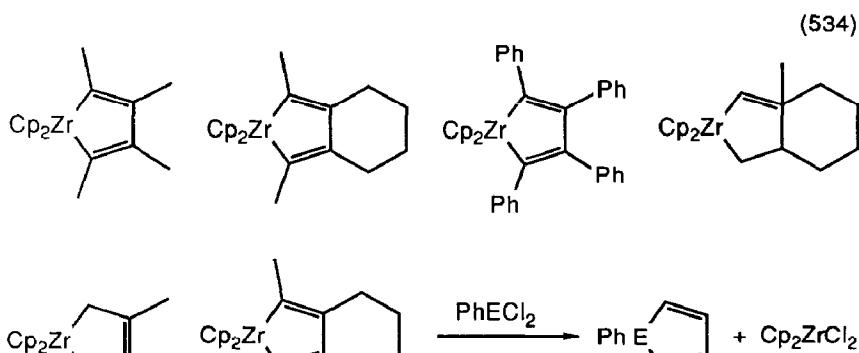
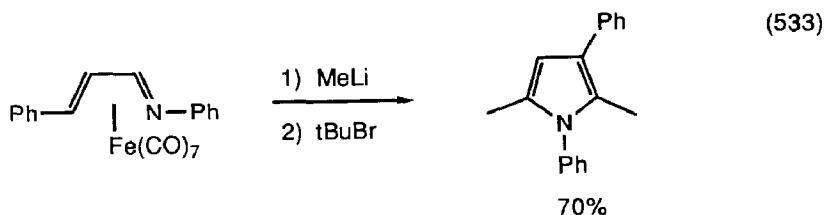


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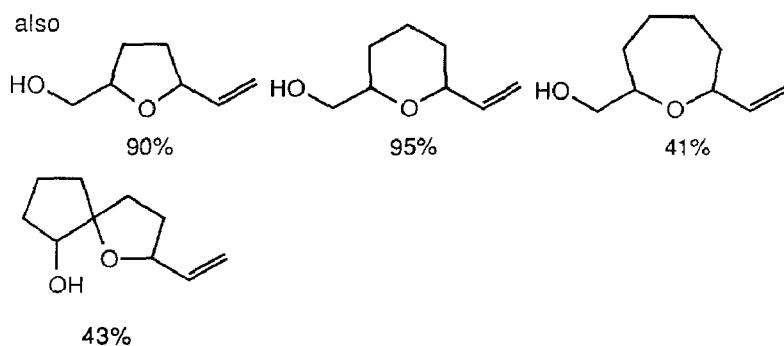
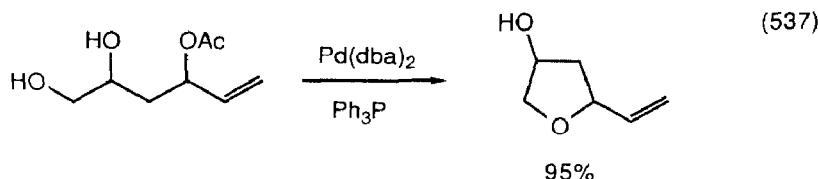
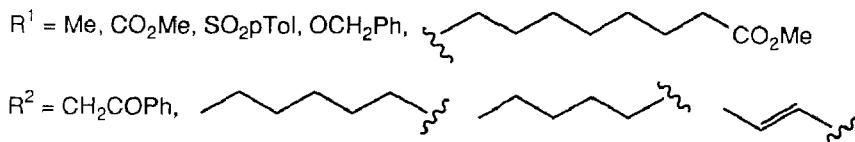
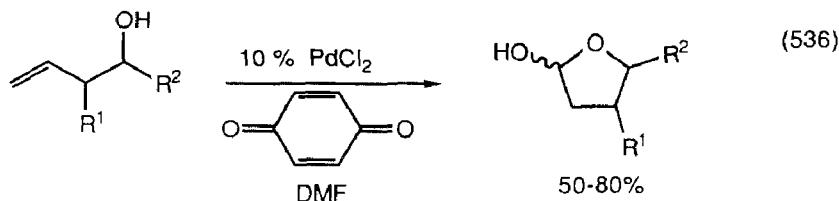


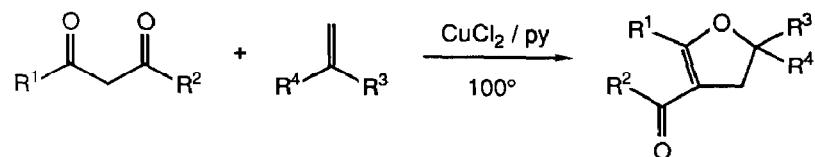
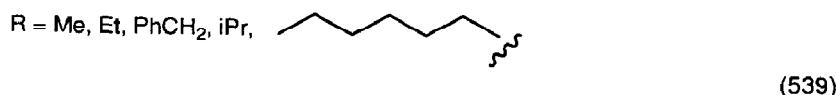
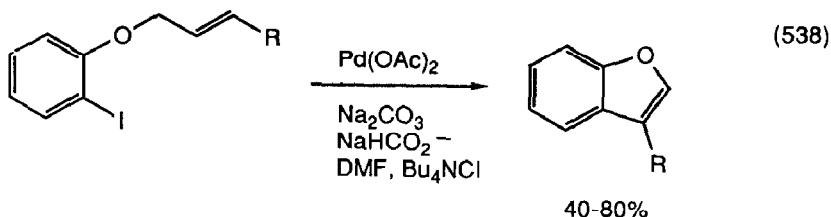
$X = \text{CONEt}_2, \text{PhSO}_2$   
 $E = \text{H}, \text{Br}, \text{Et}, \text{PhCH}_2, \text{CO}_2\text{NEt}_2$

Pyrroles were synthesized from 1-azadiene iron complexes (equation 533) [716]. A variety of 5-membered heterocycles were produced from zirconocycles (equation 534) [717]. Low valent titanium ketone coupling was used to synthesize 3,4-ditert-butyl thiophene (equation 535) [718].



Palladium(II) complexes catalyzed the oxidative cyclization of allyl alcohols to furans (equation 536) [719]. Allyl acetates having remote hydroxyl groups cyclized to tetrahydrofurans with palladium(0) catalysts (equation 537) [720]. O-Iodophenyl allyl ethers cyclized to benzofurans under palladium catalysis (equation 538) [721]. Copper(II) chloride catalyzed the addition of  $\beta$ -dicarbonyl compounds to olefins to give dihydrofurans (equation 539) [722]. Molybdenum carbonyl converted 7-oxabicyclo[2.2.1]heptadiene into furans (equation 540) [723].



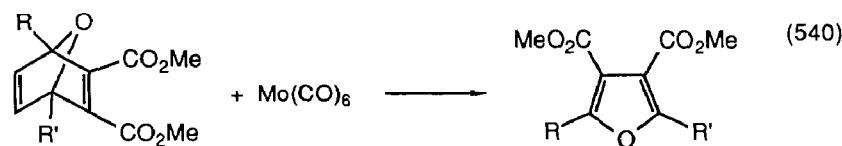


$R^1 = \text{Me}$

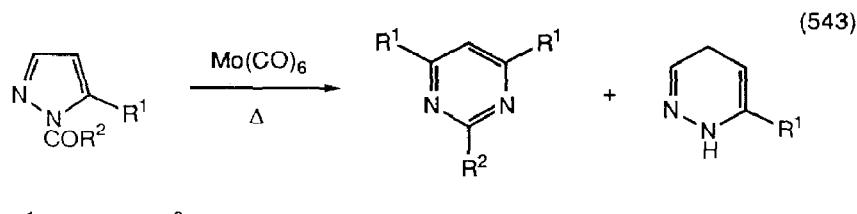
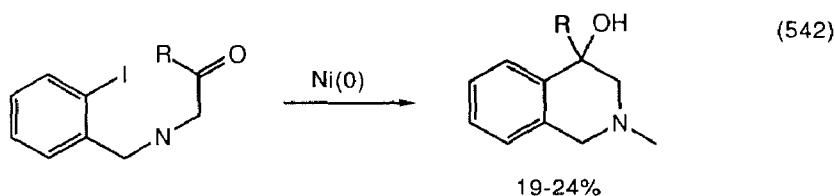
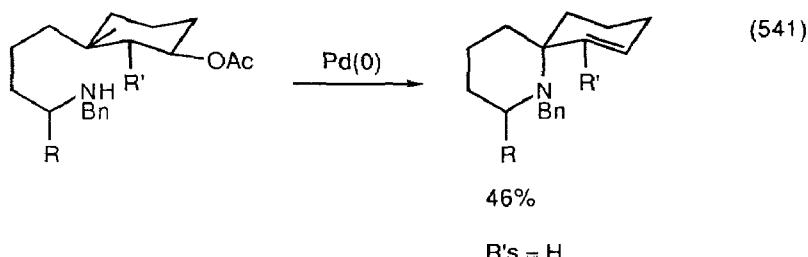
$R^2 = \text{Me, OEt}$

$R^3 = \text{H, Me, } \text{CH}_2=\text{CH-}$

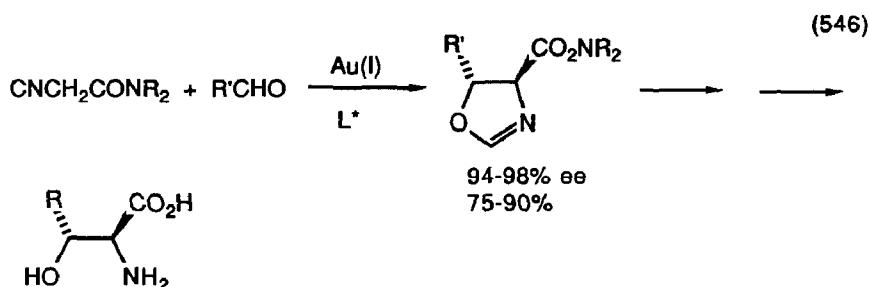
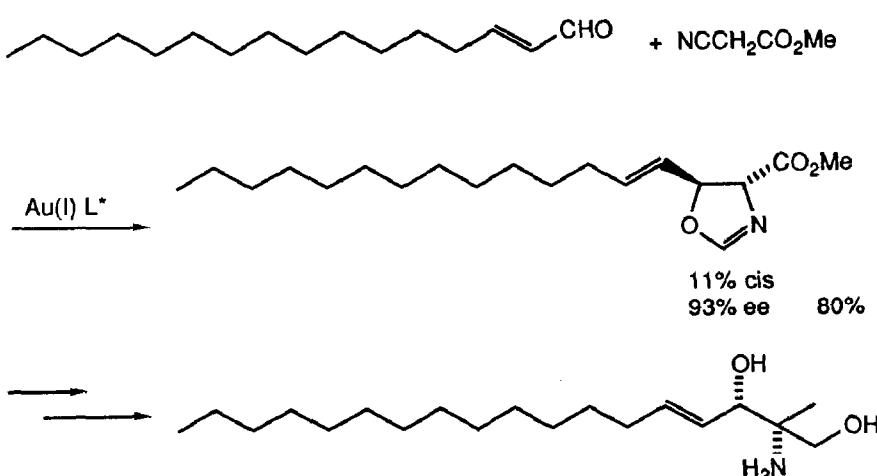
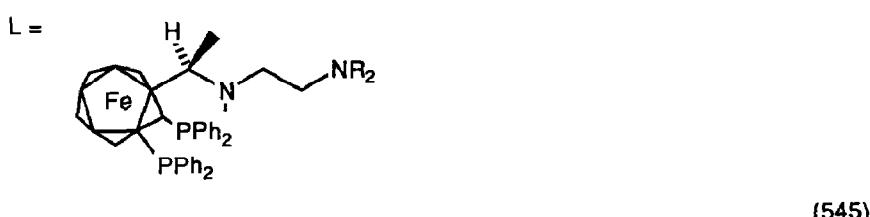
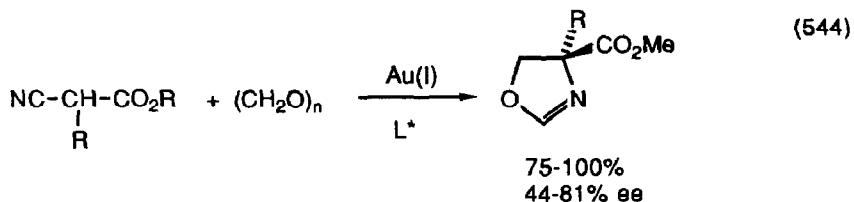
$R^4 = \text{Ph, } \text{CH}_2=\text{CH-}, \text{CH}_2=\text{CH-CH}_2-\text{CH}_2-\text{CH}_2-\text{CH}_2-$



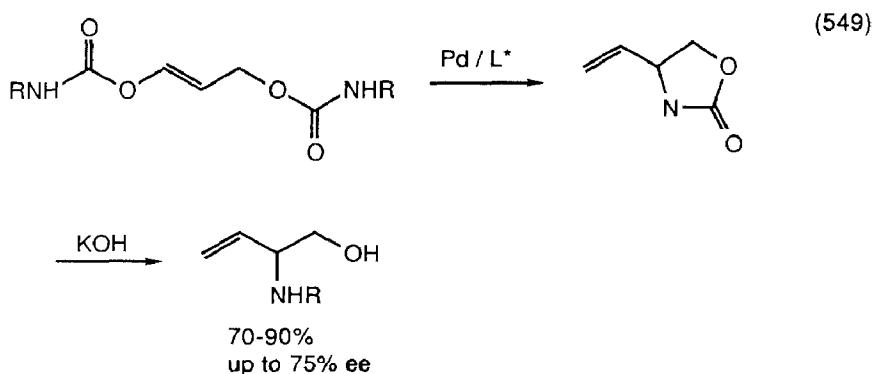
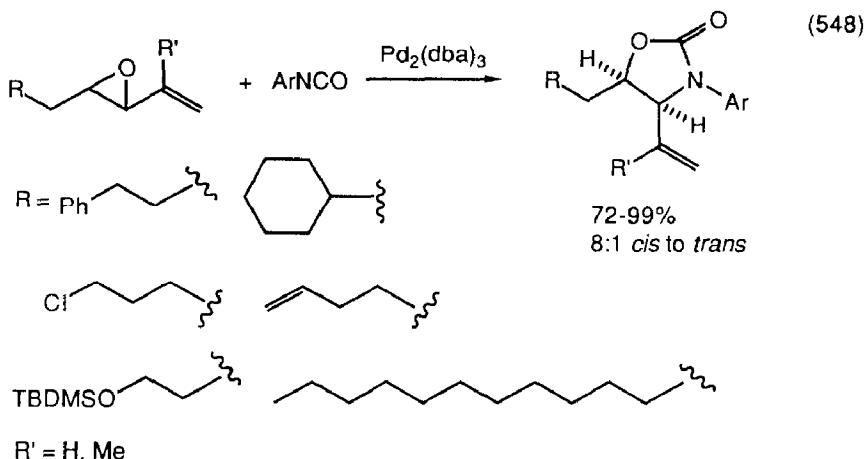
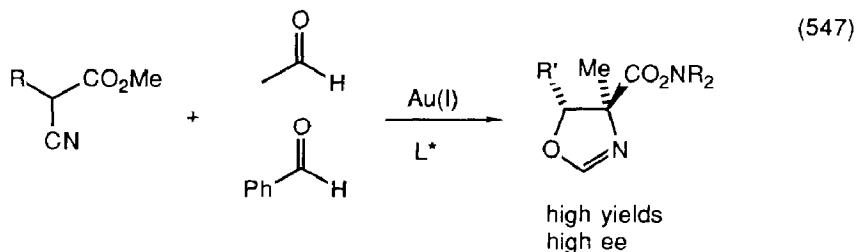
Palladium(0) catalyzed the formation of piperidines from aminoacetates (equation 541) [724]. A correlation of  $^{59}\text{Co}$  NMR spectra of cobalt catalysts with the reactivity for the coccyclotrimerization of alkynes and nitriles to give pyridines has been published [725]. 4-Aminopyridines were prepared by the reduction of the corresponding N-oxides by titanium(0) [726] and by the reduction of 4-nitropyridines with the same reagent [727]. Isoquinolinol derivatives were prepared by the nickel(0) catalyzed cyclization of o-iodobenzylphenacylamines (equation 542) [728]. Molybdenum hexacarbonyl catalyzed the isomerization shown in equation 543 [729].



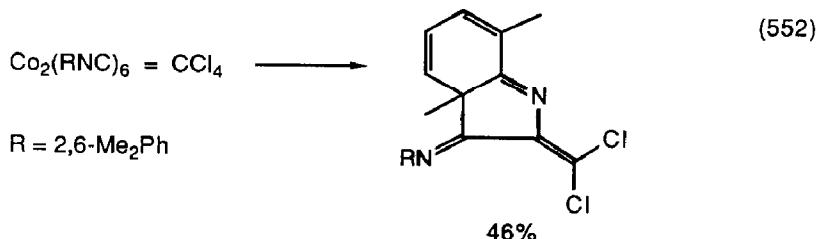
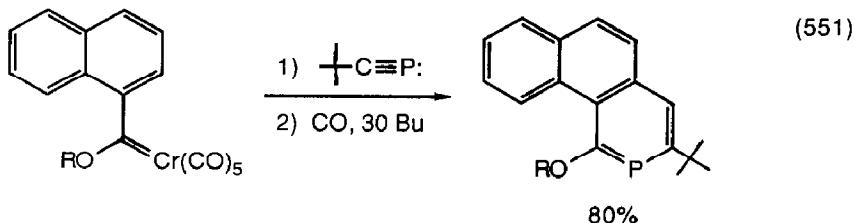
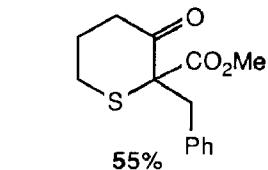
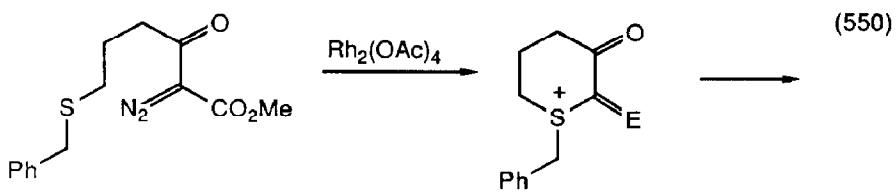
The condensation of isonitriles with aldehydes in the presence of chiral gold complexes has been utilized extensively to make chiral amino acids (equation 544) [730], (equation 545) [731], (equation 546) [732], (equation 547) [733]. Chiral carbamates were produced by the condensation of isocyanates with allyl epoxide (equation 548) [734] or allyl carbamates (equation 549) [735]. Nitriles cycloadded to azide ligands or a cobalt(III) template to produce tetrazines [736].



$\text{R}^1 = \text{CH}_3, \text{Et}, \text{iBu}, \text{Ph}, \text{pBnOPh}$   
 $\text{R}^2 = \text{Me, } (\text{CH}_2)_5$

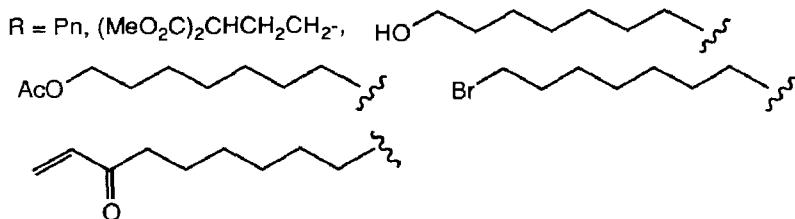
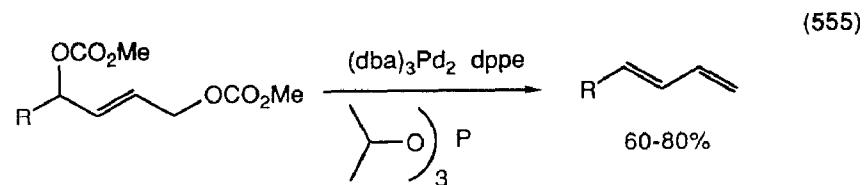
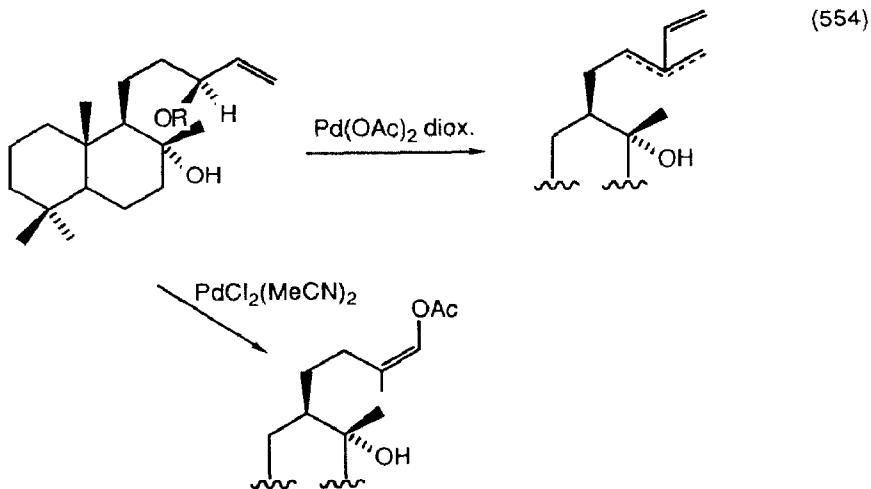
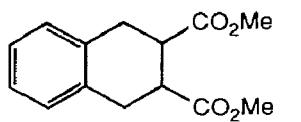
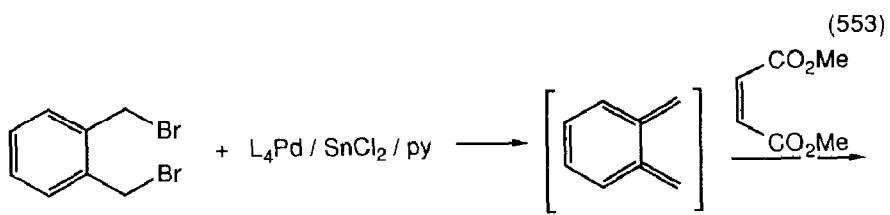


Synthesis of condensed heteroaromatic compounds using palladium catalyzed reactions has been reviewed (62 references) [737]. Sulfur heterocycles were made by rhodium(II) catalyzed decomposition of diazo compounds (equation 550) [738][739]. Phosphabzenes were made from chromium carbene complexes (equation 551) [740]. Carbon tetrachloride underwent reaction with cobalt isonitrile complexes (equation 552) [741].

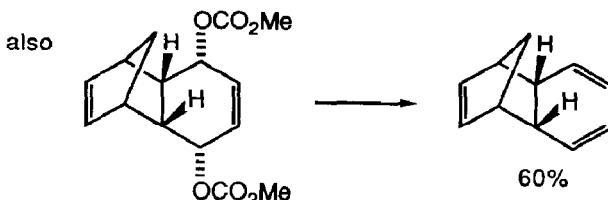


## 6. Alkenes, Alkanes

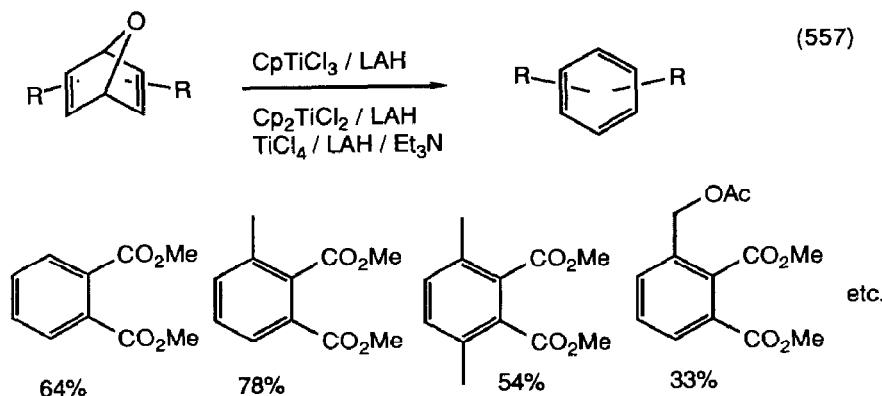
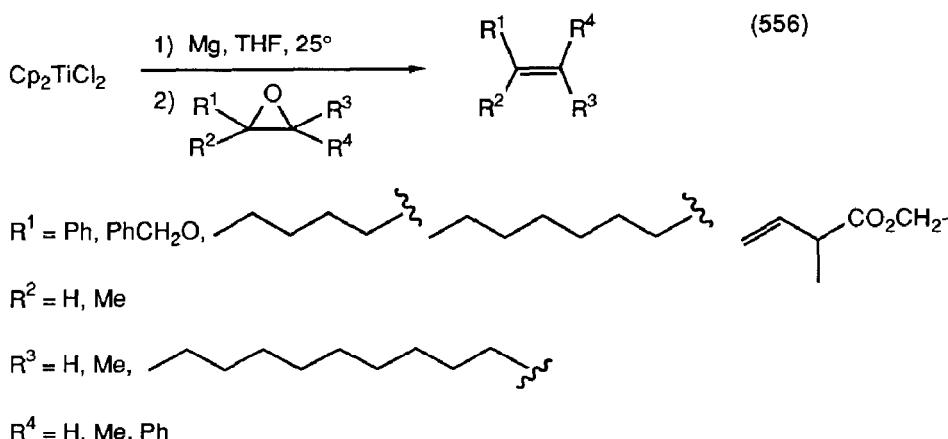
$\alpha$ -Quinodimethanes were synthesized by the palladium catalyzed reduction of  $\alpha,\alpha'$ -dibromo-oxylene (equation 553) [742]. Palladium acetate converted allylic acetates to dienes, while palladium chloride catalyzed allylic transposition (equation 554) [743]. 1,4-Dicarbonates were eliminated to dienes by palladium(0) complexes (equation 555) [744].

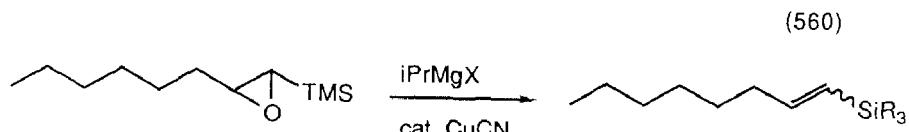
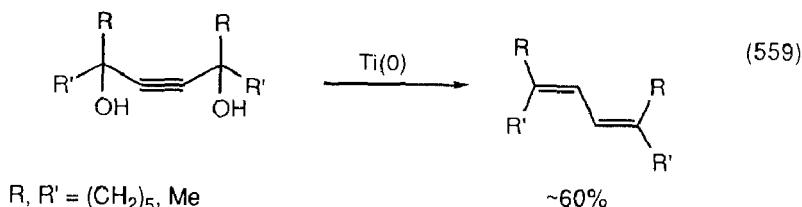
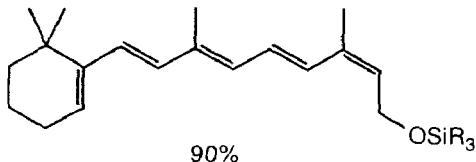
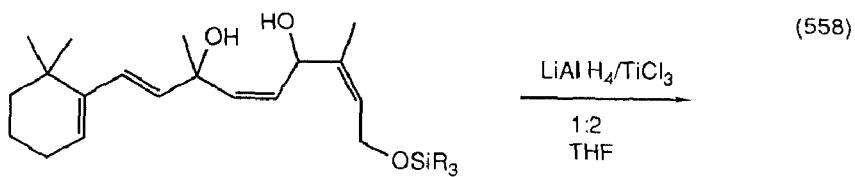


(equation 555 continued)

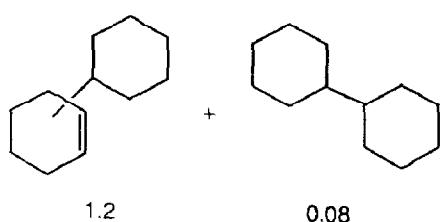
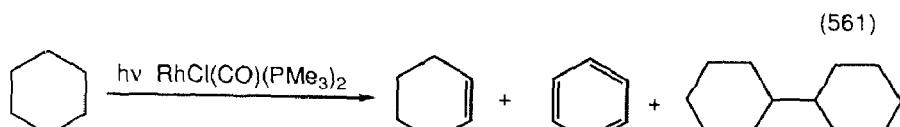


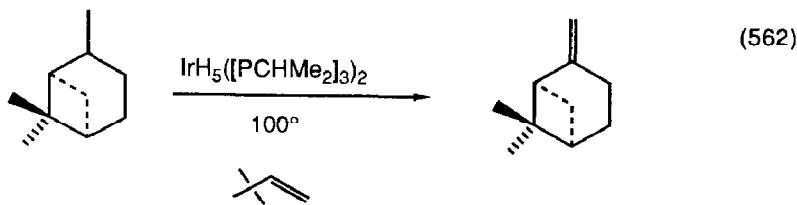
Reduced titanium species deoxygenated epoxides (equation 556) [745], 7-oxonorbornadiene species (equation 557) [746], allylic alcohols (equation 558) [747][748] and *bis*-propargyl alcohols (equation 559) [749]. Trimethylsilyl epoxides were converted to vinyl silanes by copper catalyzed Grignard reactions (equation 560) [750].



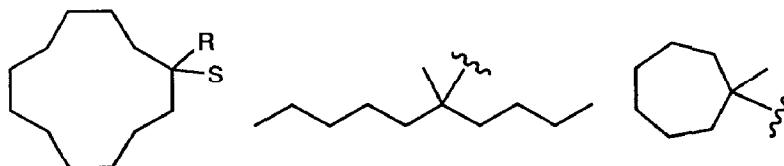
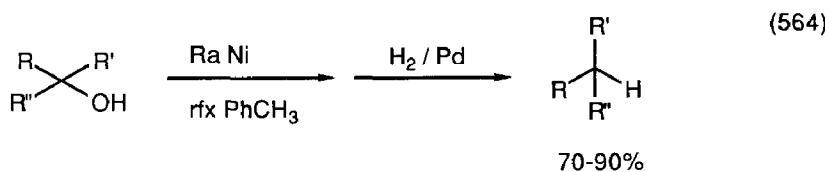
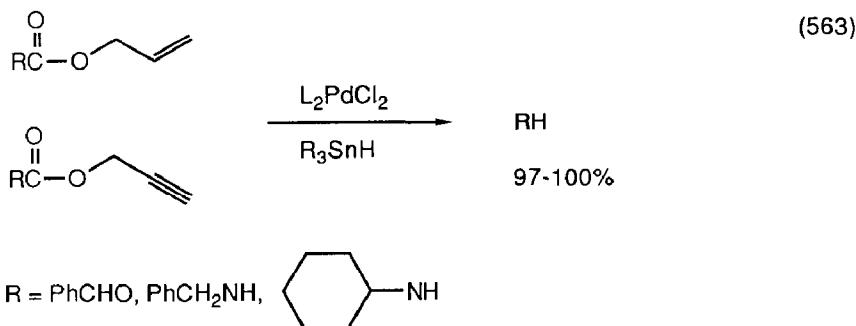


Hydrocarbon activation continues its slow development into a useful procedure to make olefins (equation 561) [751] [752], (equation 562) [753].

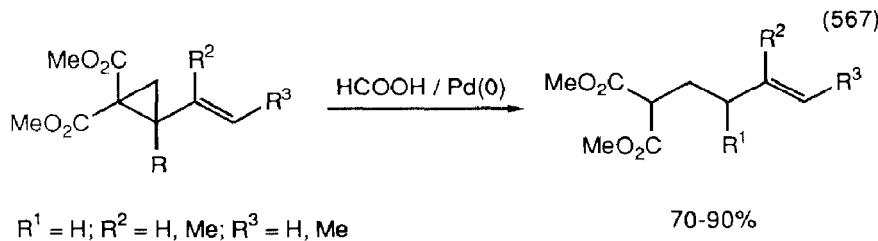
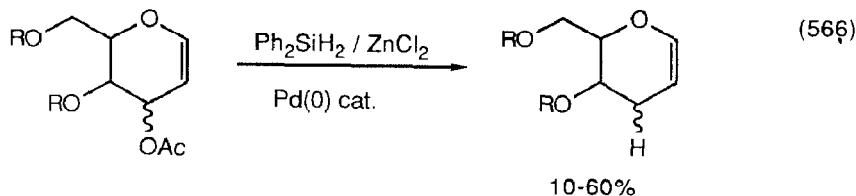
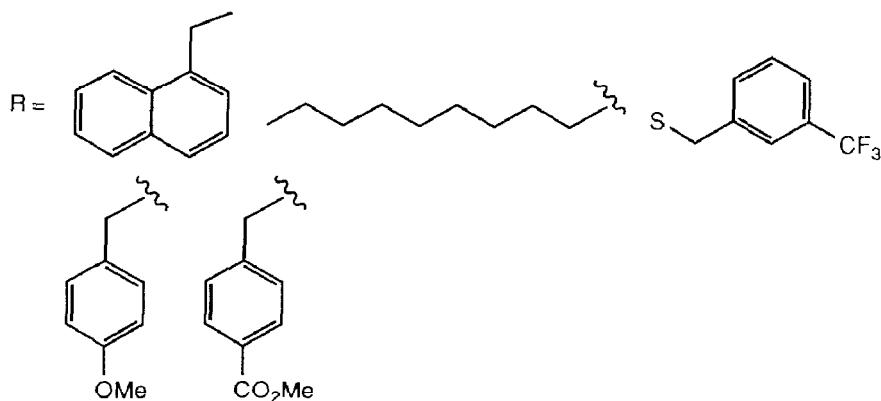
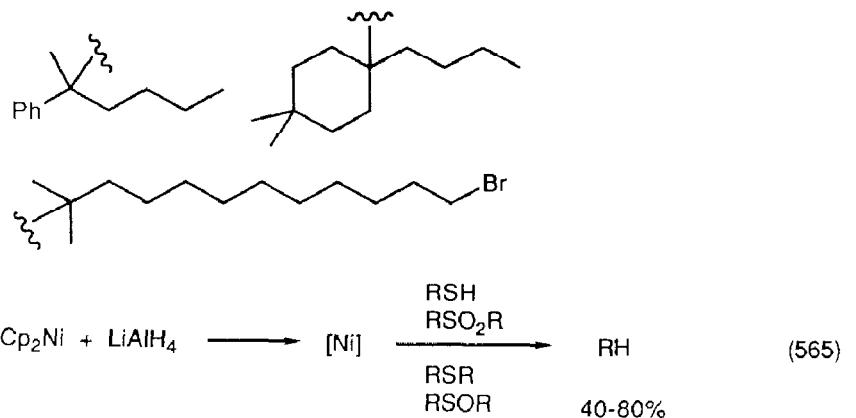


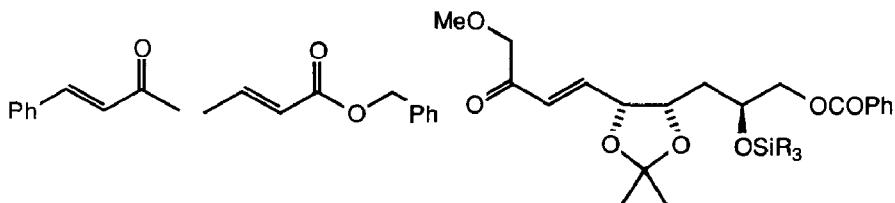
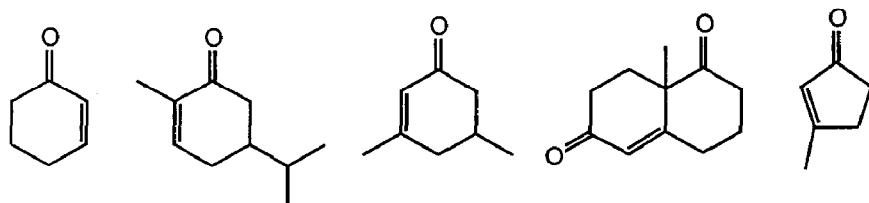
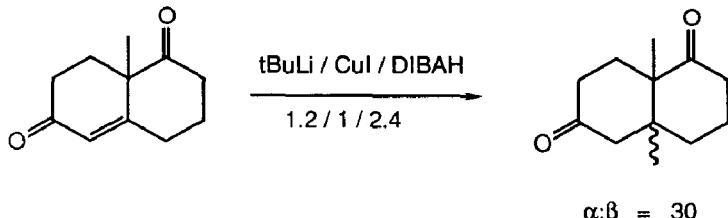
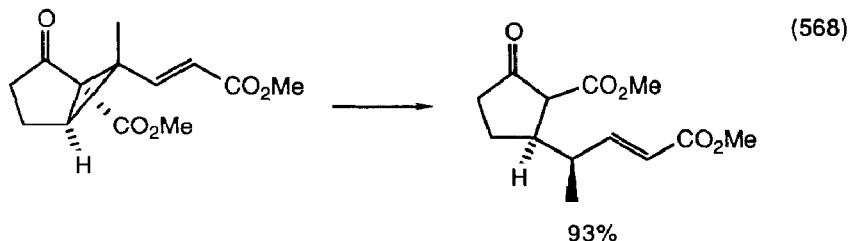


Allyl and propargyl esters were converted to hydrocarbons by treatment with palladium(II) chloride and tin hydrides (equation 563) [754]. Tertiary alcohols were reduced to hydrocarbons by sequential treatment with Raney nickel and hydrogen/palladium (equation 564) [755]. Reduction of nickelocene with lithium aluminum hydride produced a species which converted sulfur compounds to hydrocarbons (equation 565) [756]. Allyl acetates (equation 566) [757] and vinyl cyclopropanes (equation 567) [758] were converted to hydrocarbons with palladium(0) catalyst/reducing agent. The system t-butyllithium/copper(I) iodide/DIBAH effected 1,4-reduction of conjugated enones (equation 568) [759], as did the reagent  $[Ph_3PCuH]_6$  (equation 569) [760].

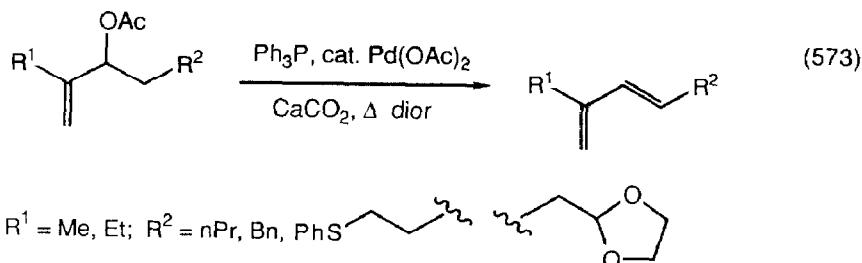
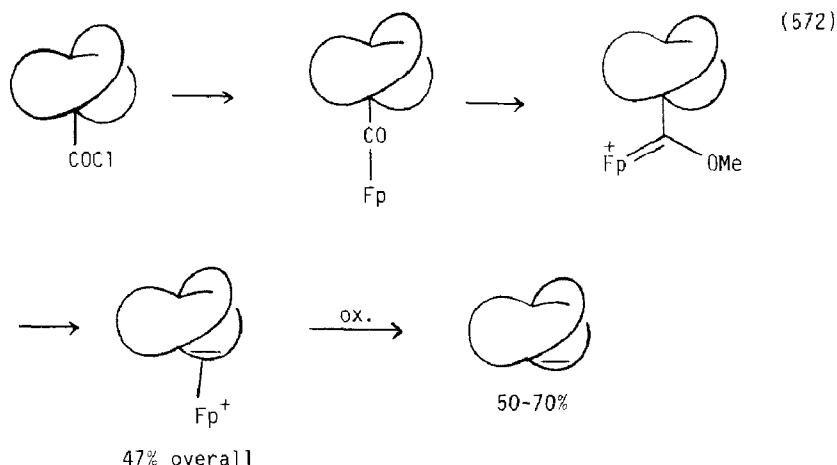
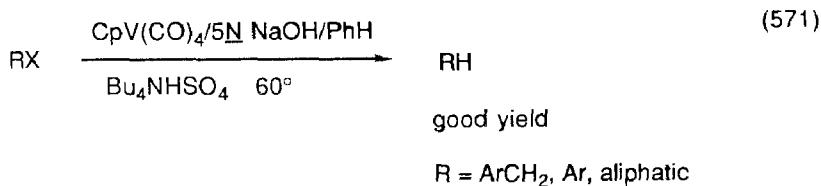
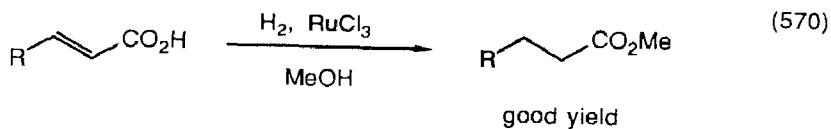


(equation 564 continued)



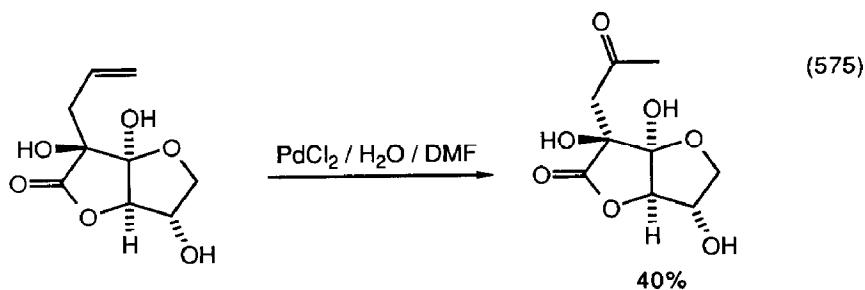
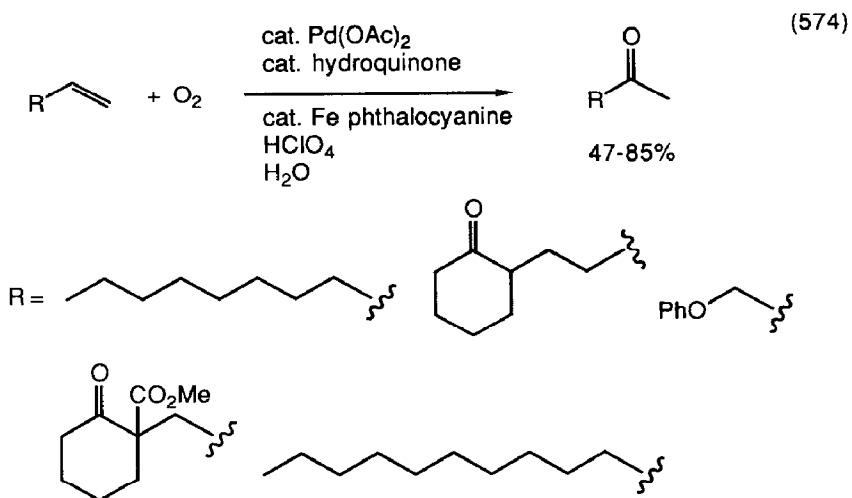


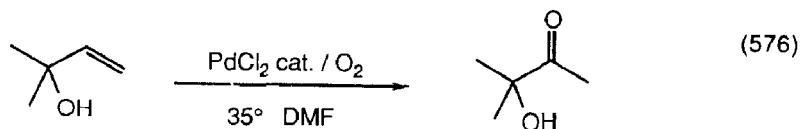
Ruthenium trichloride catalyzed the reduction of conjugated acids to saturated esters (equation 570) [761]. Cyclopentadienylvanadium complexes reduced halides to hydrocarbon (equation 571) [762]. Bridgehead olefins were prepared utilizing iron acyl, carbene, and olefin complex chemistry (equation 572) [763]. Allylic acetates were converted to dienes by palladium acetate based catalysts (equation 573) [764].



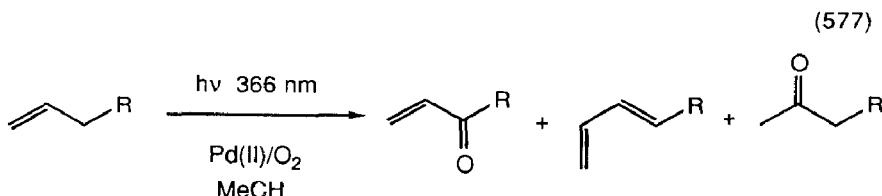
## 7. Ketones, Aldehydes

Wacker oxidation of olefins to ketones continues to be exploited. Aqueous ruthenium(III) chloride efficiently oxidized 1-hexene to 2-hexanone [765], while  $\text{PdCl}_2/\text{CuCl}_2/\text{O}_2$  systems were used to convert cyclopentane to cyclopentanone [766][767]. Palladium(II) nitro and nitrate complexes have been studied as oxidizing agents for olefins [768]. Alkenes were oxidized to ketones by  $\text{PdCl}_2/\text{BiCl}_3/\text{LiCl}$  systems [769]. A very efficient redox system for Wacker chemistry has been developed (equation 574) [770]. Palladium(II) oxidation of olefins to ketones has been successfully used in highly functionalized systems (equation 575) [771], (equation 576) [772]. Photolysis of olefins in the presence of palladium(II) salts resulted in allylic oxidation (equation 577) [773].

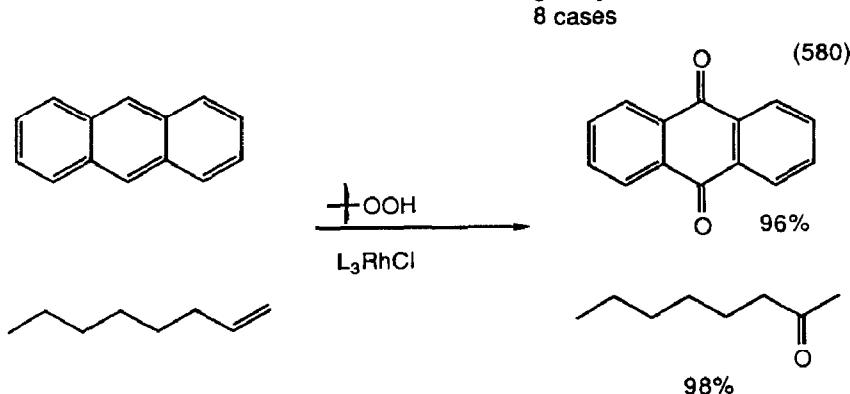
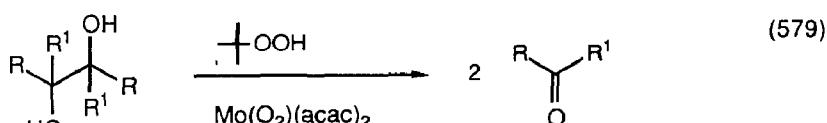
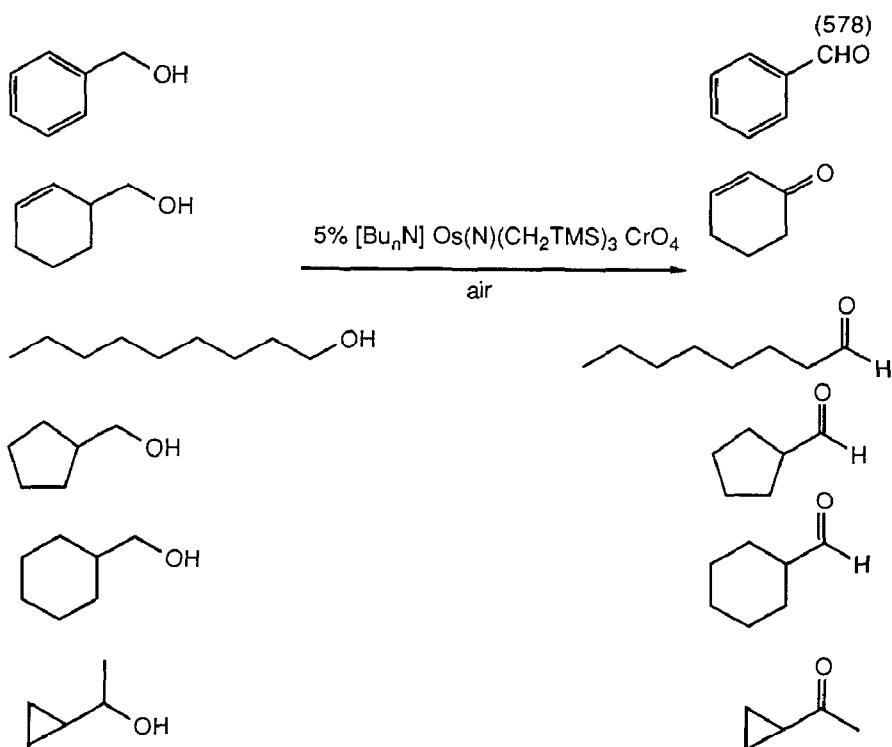


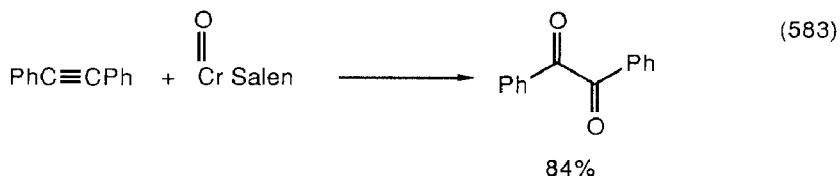
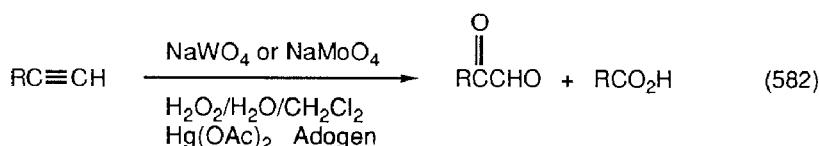
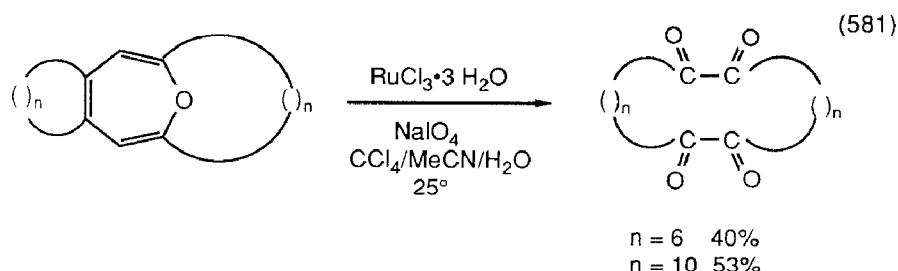


98% conversion  
90% selectivity

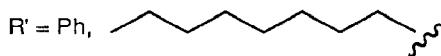
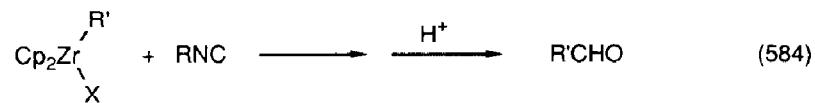


An osmium based reagent was a moderately active catalyst for the oxidation of alcohols to aldehydes or ketones (equation 578) [774]. Diols were oxidatively cleaved by t-butylhydroperoxide and a molybdenum oxo catalyst (equation 579) [775]. Rhodium(I) complexes catalyzed the oxidation of olefins to ketones by t-butylhydroperoxide (equation 580) [776]. Ruthenium(III) salts catalyzed the oxidation of unsaturated cyclic ethers (equation 581) [777]. Alkynes were oxidized to carbonyl compounds by hydrogen peroxide/tungstate or molybdate complex catalyst (equation 582) [778] and by high valent chromium complexes (equation 583) [779].

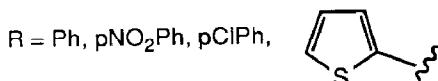
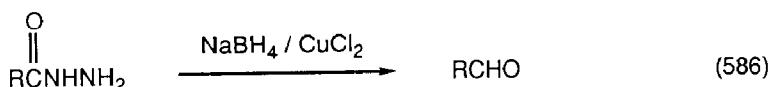
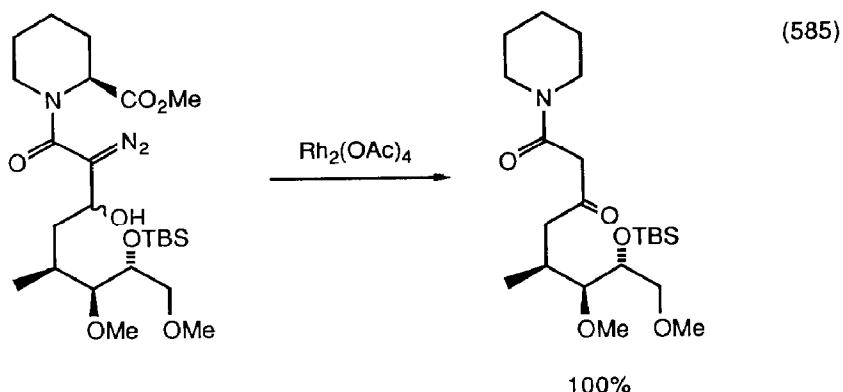
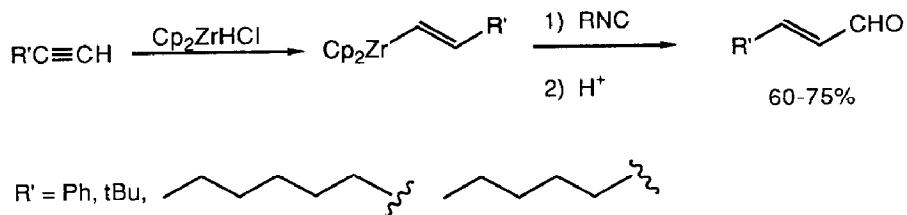




Benzyl bromide was inefficiently converted to the aldehyde by electrochemical reductions in the presence of iron pentacarbonyl followed by acid hydrolysis [780]. Alkylzirconium species were converted to aldehydes by reaction with isonitriles (equation 584) [781]. Miscellaneous ketone syntheses are shown in equation 585 [783] and equation 586 [784].

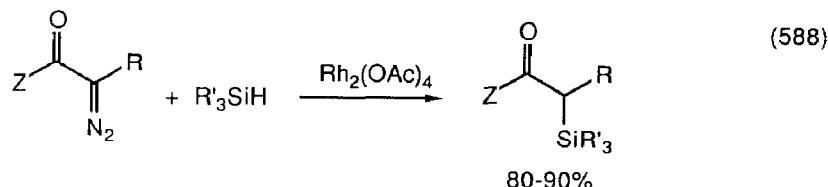
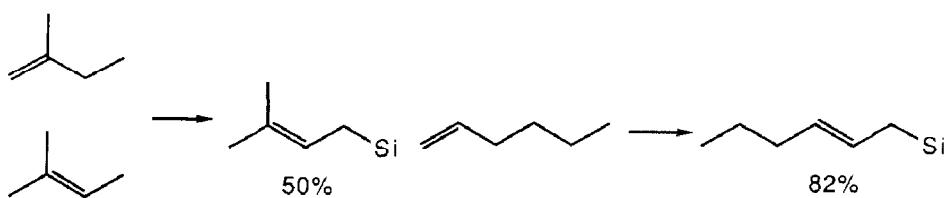
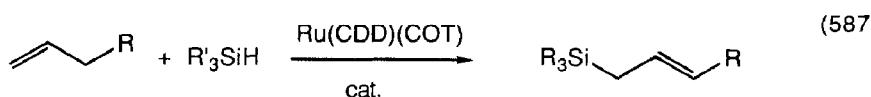


(equation 584 continued)

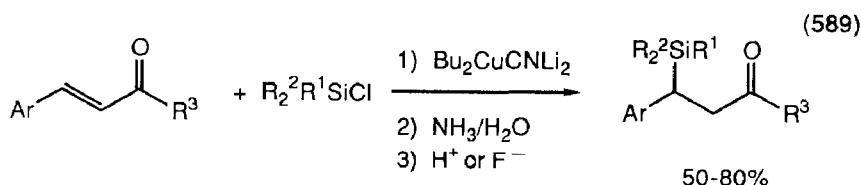


### 8. Organosilanes

Silicate supported rhodium(I) complexes catalyzed the hydrosilylation of alkenes and vinyl silanes [785]. Arenechromium tricarbonyl complexes catalyzed the hydrosilylation of 2,3-dimethyl-1,3-butadiene in excellent yield [786]. Supported rhodium complexes also catalyzed the hydrosilylation of acetylene [787]. Olefins were converted to allylsilanes using ruthenium catalysts (equation 587) [788]. Rhodium(II) catalyzed decomposition of diazo compounds in the presence of trialkylsilanes produced  $\alpha$ -silylketones and esters (equation 588) [789]. Conjugated enones were  $\beta$ -silylated using organocupper chemistry (equation 589) [790]. Silylcuprates added to allenes (equation 590) [791]. The full details of the chemistry depicted in equation 591 have been published [792][793].



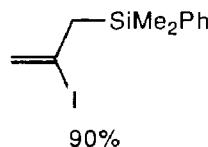
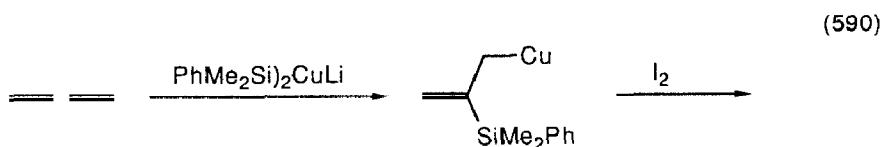
$\text{Z} = \text{EtO}_2\text{C}, \text{MeCO}, \text{PhCO}, \text{PhCH}_2\text{CO}$ ,  $\text{R} = \text{H, Me}$

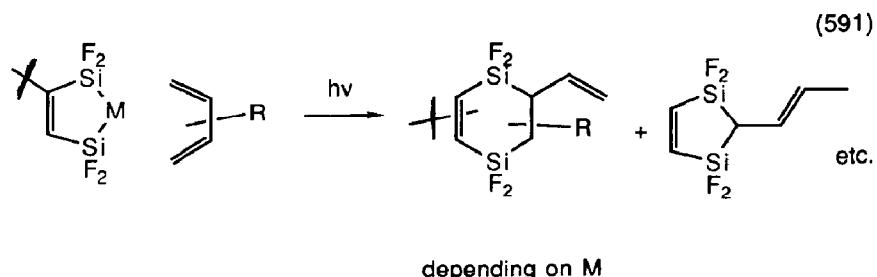


$\text{R}^1, \text{R}^2 = \text{Me, Et, tBu, Ph}$

$\text{R}^3 = \text{Me, Ph}$

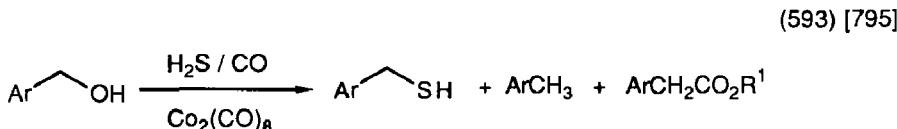
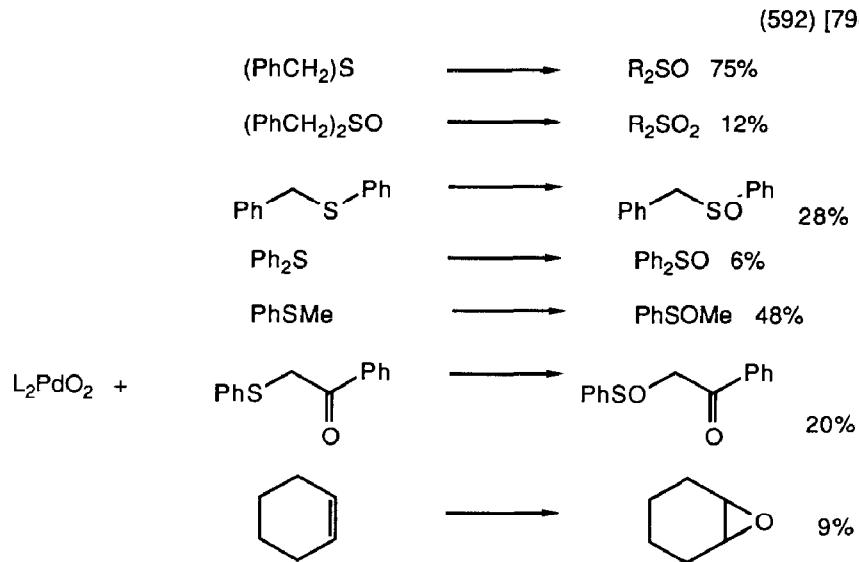
$\text{Ar} = \text{Ph, pMeOPh}$



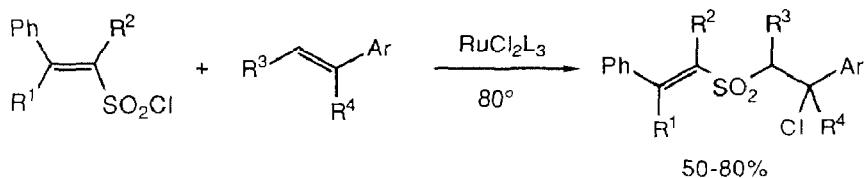


### 9. Miscellaneous

Miscellaneous processes producing sulfur and selenium compounds are presented in equations 592-598. The reference is found adjacent to the equation in brackets.

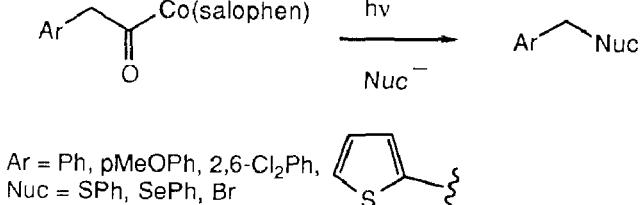


(594) [796]

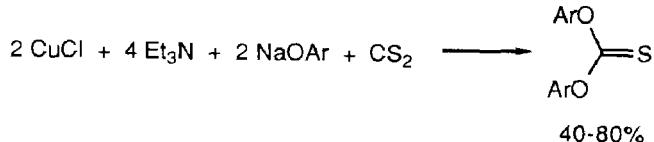


50-80%

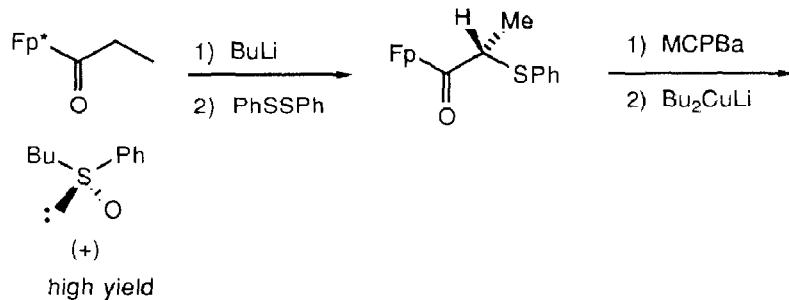
(595) [797]

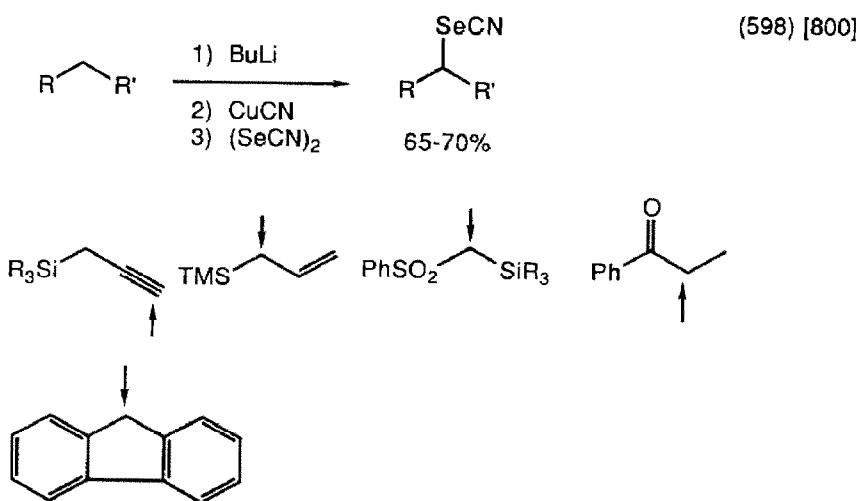


(596) [798]

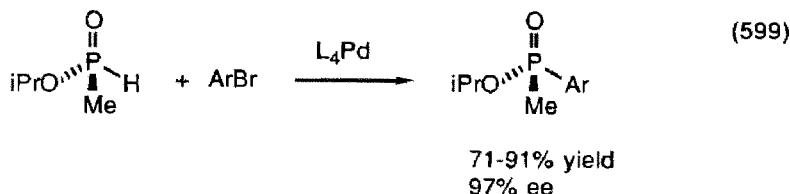


(597) [799]

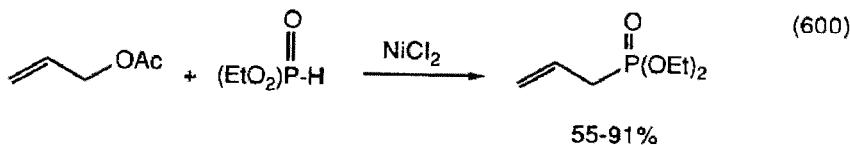




"Phosphaalkynes, syntheses, reactions, coordination behavior" was the subject of a review (183 references) [801]. Palladium (equation 599) [802] and nickel (equation 600) [803] catalysts were used in the synthesis of phosphorous compounds.

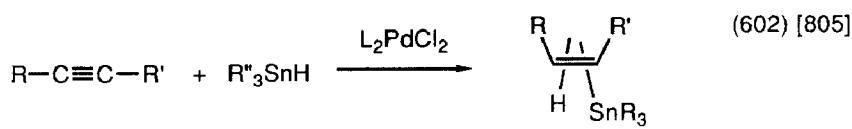
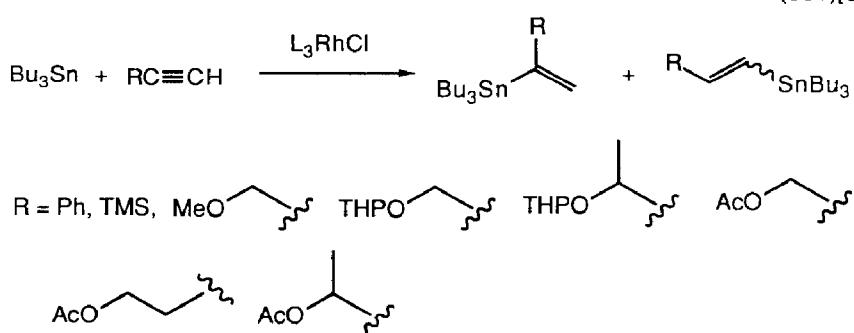


Ar = Ph, pMePh, pNO<sub>2</sub>Ph, β-napht, pPhPh



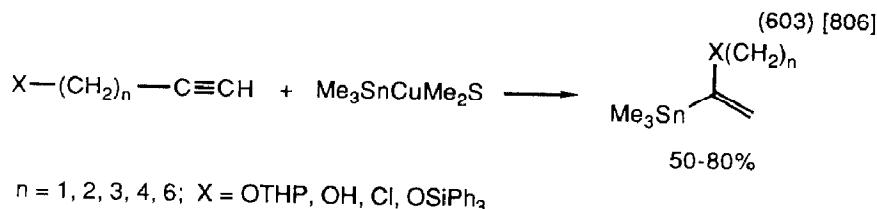
Reactions producing organotin compounds are shown in equations 601-605. Methyl groups on tertiary amines were oxidized by t-butyl hydroperoxide in the presence of a ruthenium catalyst (equation 606) [809].

(601)[804]

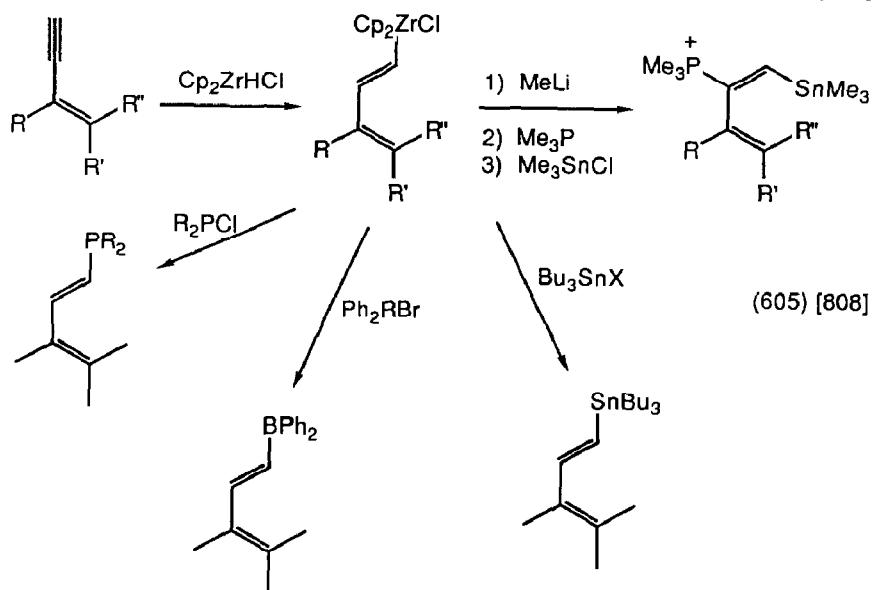


$\text{R} = \text{H}, \text{MeO}_2\text{C}, \text{Ph}$   
 $\text{R}' = \text{CH}_2\text{OH}, \text{CH}_2\text{OTHP}, \text{CO}_2\text{Me}, \text{Me}$

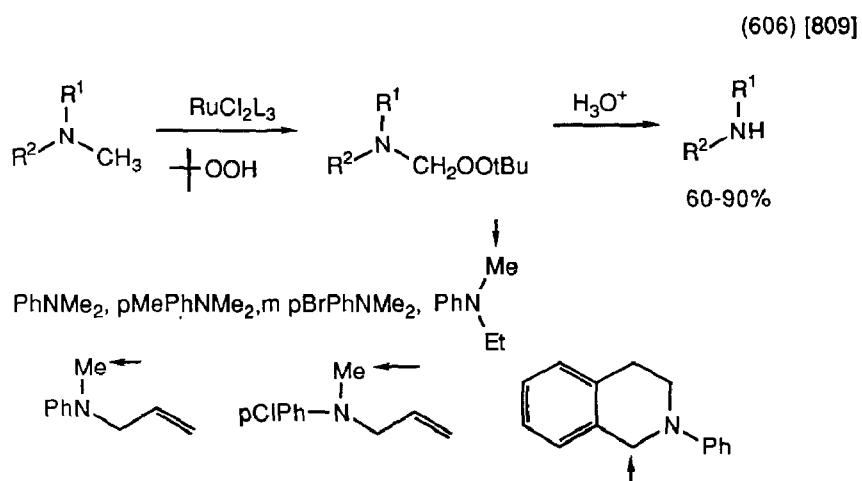
(603) [806]



(604) [807]



(605) [808]



#### IV. Reviews

The following reviews and dissertations have appeared.

- "Transition metals in organic synthesis", Annual Survey 1986 (843 references) [810].
- "Ammonium formate in organic synthesis" (30 references) [811].
- "Organozirconium compounds in organic synthesis" (178 references) [812].
- "Organic reactions of selected  $\pi$ -complexes", Annual Survey 1987 (383 references) [813].
- "Transition metals in organic synthesis, hydroformylation, reduction, oxidation" Annual Survey 1987 (673 references) [814].
- "Transition metals in organic synthesis, hydroformylation, reduction, oxidation" Annual Survey 1986 (672 references) [815].
- "Carbon-carbon bond formation through organometallic elimination reactions" (117 references) [816].
- "Organopalladium approaches to interphenylene prostaglandin analogs, heterocycles and carbocycles" (dissertation) [817].
- "Use of organoiron compounds in organic synthesis" (303 references) [818].
- "Transition metal-stabilized carbocations in organic synthesis" (150 references) [819].
- "Organometallic chemistry can simplify the synthesis of important biologically active natural products" (17 references) [820].
- "Organometallic methods in organic synthesis in industry" (7 references) [821].
- "Application of transition metals in natural products and heterocycles synthesis" (110 references) [822].
- "Some uses of metal carbonyl complexes in organic synthesis" (29 references) [823].
- "Organic synthesis using ruthenium compounds" (153 references) [824].
- "Organic synthesis using tin" (35 references) [825].
- "Fine organic synthesis and metals" (6 references) [826].
- "Organic synthesis with palladium catalysts. Intramolecular addition of OH, NH, COOH to acetylene moiety" (29 references) [827].
- "Application of palladium complexes in organic synthesis" (39 references) [828].
- "Organonickel chemistry" (77 references) [829].
- "Metal reagents in stereo- and regioselective organic synthesis" [830].
- "Novel methods of carbon-carbon bond formation via organometallic reagents" (dissertation) [831].
- "Transition metals in organic synthesis" (dissertation) [832].
- "Making pharmaceuticals via homogeneous catalysis" (43 references) [833].
- "Organochromium reagents for highly selective carbon-carbon bond formation" (31 references) [834].

- "Organic synthesis promoted by transition-metal complexes" (35 references) [835].
- "Use of organonickel compounds in organic synthesis" (363 references) [836].
- "Use of organorhodium compounds in organic synthesis" (551 references) [837].
- "Development of metalorganic assisted synthetic methods toward important organic molecules" (20 references) [838].
- "Carbon-carbon bond forming reaction of organotransition metal enolate complexes" (9 references) [839].
- "Asymmetric synthesis catalyzed by chiral ferrocenylphosphine-metal complexes" (25 references) [840].
- "Asymmetric synthesis via the iron chiral auxiliary  $[(\eta^5\text{-C}_5\text{H}_5)\text{Fe}(\text{CO})(\text{PPh}_3)]$ " (34 references) [841].
- "Metal mediated routes to 5-membered rings" (22 references) [842].
- "Metal catalyzed carbonylation and oxidation-reduction reactions" (26 references) [843].
- "Metal-directed stereoselective functionalization of alkenes in organic synthesis" (17 references) [844].
- "Chiral acetals in enantio- and diastereoselective substitution or elimination reactions" (26 references) [845].
- "Organocuprates addition to  $\alpha,\beta$ -unsaturated compounds: synthetic and mechanistic aspects" (32 references) [846].
- "New organometallic reagents for olefin synthesis" (14 references) [847].
- "Stereospecific synthesis of olefins through sequential cross-coupling reactions" (23 references) [848].
- "Total synthesis of polyprenoid natural products via Pd(0)-catalyzed oligomerizations" (31 references) [849].
- "Cyclopropanations and cycloadditions of transition metal carbene complexes" (24 references) [850].
- "Modern asymmetric synthesis of  $\alpha$ -amino acids" (299 references) [851].
- "Asymmetric catalysis in organic synthesis with industrial perspectives" (88 references) [852].
- "Enantioselective syntheses with optically active transition-metal catalysts" (157 references) [853].
- "Platinum metal complexes in asymmetric catalysis. Continuing developments will increase potential" (12 references) [854].
- "Preparation of chiral ligands from amino acids, applications to catalytic asymmetric synthesis: a review" (43 references) [855].
- "Controlling metal-catalyzed organic syntheses. (Part 7): The concept of order factors in aspects of asymmetric synthesis" (34 references) [856].
- "Metal mediated routes to 5-membered rings" (94 references) [857].

- "Metal reagents in stereo- and regioselective functionalization of conjugated dienes" (29 references) [858].
- "Homogeneous catalysis by transition-metal complexes" (378 references) [859].
- "Novel routes to heterocyclic compounds" (19 references) [860].
- "Reactions and catalytic applications of ( $\eta^6$ -arene)nickel silyl complexes in their high nickel oxidation states" (dissertation) [861].
- "Role of the organometallic complexes in organic reactions and studies on the kinetic behavior of the complexes by NMR" [862].
- "Palladium-catalyzed reaction of tributyltin hydride. Regiospecifically controlled formation of  $\alpha$ -bromo- $\alpha,\alpha$ -disubstituted ketones from tributyltin  $\beta$ -ketocarbosylates" [863].
- "Palladium clusters - catalysts for reactions of unsaturated compounds" (43 references) [864].
- "Synthesis and reactivity of titanocene methylene complexes and bis( $\eta^5$ -cyclopentadienyl)titanacyclobutenes" (dissertation) [865].
- "Utilization of dioxygen for the specific oxidation of organic substrates with cobalt(II) catalysts" (29 references) [866].
- "Oxygenative cleavage of aromatic rings catalyzed by metal complexes" (41 references) [867].
- "Vinylcyclopropane rearrangements" (152 references) [868].
- "The influence of silyl groups on the reactivity and formation of metal-carbon double bonds" (32 references) [869].
- "The coordination chemistry of electron-rich alkenes (enonetetramines)" (94 references) [870].
- "Nickela-heterocyclen als Intermediate der präparativen Isocyanatchemie" (26 references) [871].
- "Nucleophilic additions reactions with cationic iron  $\pi$ -alkyne and related complexes" (46 references) [872].
- "Group(IV) metal complexes of benzenes, cycloalkynes, acyclic alkynes and alkenes" (59 references) [873].
- "General principles and characteristics of cyclopalladation reactions" (232 references) [874].
- " $\eta^6$ -Arene- $\eta^5$ -cyclopentadienylruthenium complexes and related systems" (41 references) [875].
- "Cleavage and formation of carbon-carbon and carbon-oxygen bonds in reactions involving dimolybdenum and ditungsten hexaalkoxides" (49 references) [876].
- "Application of the principles of homogeneous catalysis to the synthesis and mechanistic study of heterogeneous catalysts for olefin conversion" (22 references) [877].

- "Concerted and stepwise mechanisms in heterogeneous acid catalysis" (15 references) [878].
- "Catalytic activation of small molecules by transition metal complexes" (dissertation) [879].
- "Reductions promoted by low valent transition metal complexes in organic synthesis" (301 references) [880].
- " $\eta^2$ -Olefin complexes and their application in organic synthesis" (17 references) [881].
- "Group(VIII) metal-catalyzed organic syntheses via metallacyclic intermediates" (41 references) [882].
- "Contributions to organonickel chemistry" (77 references) [883].
- "Controlled carbometallation as a new tool for carbon-carbon bond formation and its application to cyclization" (101 references) [884].
- "Transition metal promoted reactions of polyhedral boranes and carboranes" (32 references) [885].
- "Theoretical elucidation of reaction mechanisms of transition metal complex catalysts using the molecular orbital methods" (24 references) [886].
- "Reactions involving alkynes and tungsten-tungsten triple bonds supported by alkoxide ligands" (39 references) [887].
- "Carbonyl compounds and organometallic carbonyl synthons: syntheses, stabilities, and new transformations" (dissertation) [888].
- "Chemistry of transition metal ketene complexes effective for utilization of carbon monoxide" (56 references) [889].
- "The activation and functionalization of C-H bond in saturated hydrocarbons by using soluble transition metal systems" (37 references) [890].
- "Activation of molecules by homogeneous catalysts. Activation of alkanes and model of the reaction intermediate" (19 references) [891].
- "Transition metal catalyzed reductions of organic molecules by molecular hydrogen and hydrides, an overview" (51 references) [892].
- "Diene-, alkyne-, alkene-, and alkyl complexes of earlier transition metals: structures and synthetic applications in organic chemistry and polymer chemistry" (131 references) [893].
- "Transition metal hydrides as reagents in organic and organometallic synthesis" (dissertation) [894].
- "Copper complexes and their role in light-induced oxidations" (8 references) [895].
- "New carbon-carbon bond-forming reactions of diiron complexes" (18 references) [896].
- "Metal complex catalysis in synthesis of polycyclic hydrocarbons" [897].

- "Transformations of unsaturated compounds in the presence of heterogenized complexes of transition metals. Synthesis of heterogenized water-soluble metal-complex catalysts" [898].
- "Transition metal species supported on silica gel. A versatile catalyst of organic reactions in liquid or vapor phase" [899].
- "Progress of polymer-supported catalysts" (56 references) [900].
- "Carbon-hydrogen activation by metal complexes. Direct and regioselective functionalization of hydrocarbons" (13 references) [901].
- "The role of organometallic intermediates in the catalysis of hydrocarbon transformations" [902].
- "Carbon-carbon bond activation by organometallic compounds" (dissertation) [903].
- "Metal-mediated making and breaking of carbon-carbon bonds in aromatic hydrocarbons" [904].
- "Carbon-carbon bond activation in hydrocarbons with organometallic complexes" (dissertation) [905].
- "Reactivity of coordinated hydrocarbons" (55 references) [906].
- "Reactions of transition metal ions with cycloalkanes and metal carbonyls" [907].

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