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New applications of metal catalysis in natural product synthesis

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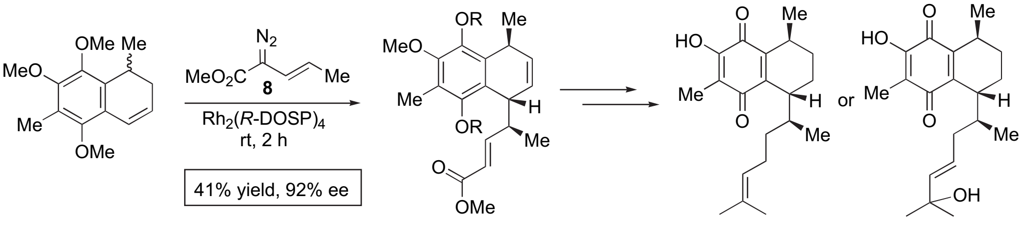

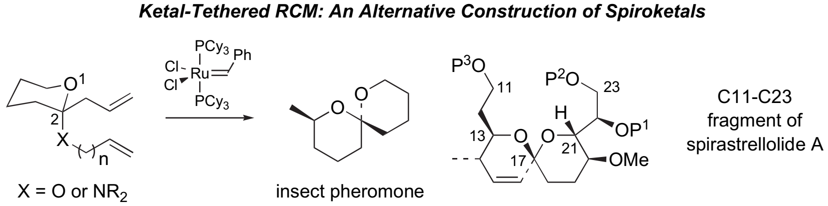

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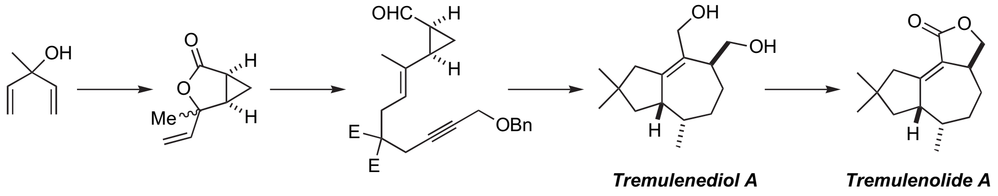
Brandon L. Ashfeld and Stephen F. Martin*



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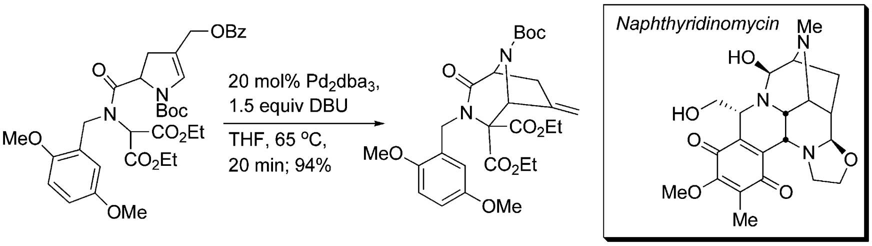

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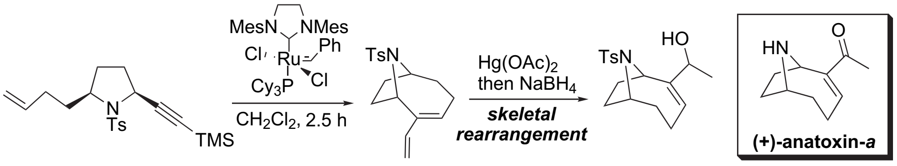

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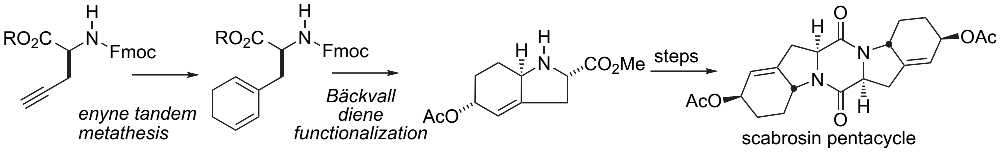
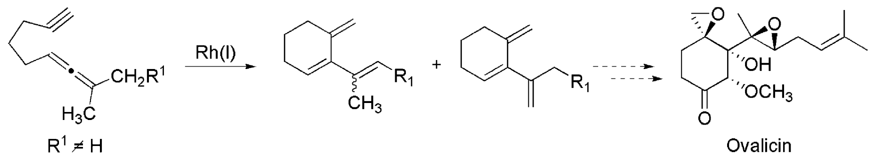
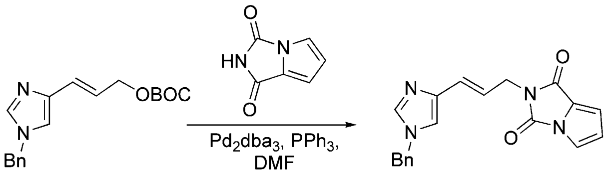
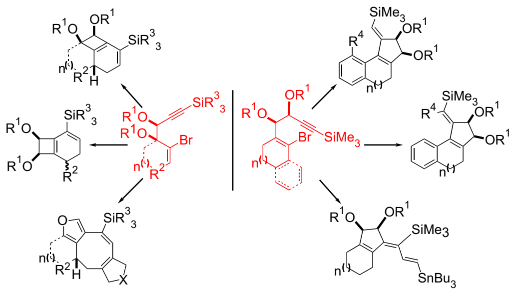
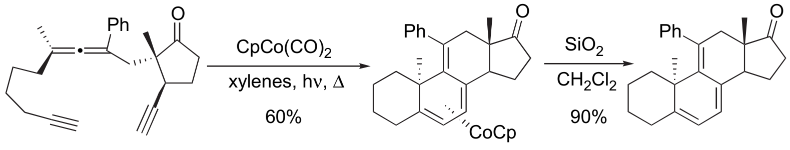
Mark D. Middleton, Brian P. Peppers and Steven T. Diver*

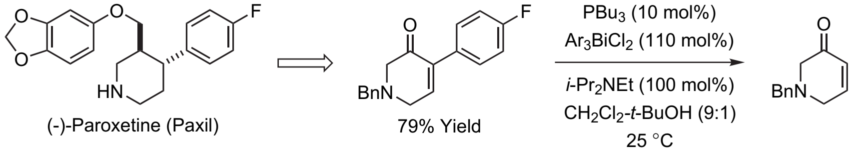



Rh(I)
CH2R1
H3C
R1 ≠ H
R1
CH3
+
R1
O
O
OH
OCH3
O
Ovalicin


N
N
Bn
OBOC
O
HN
N
O
Pd2dba3, PPh3,
DMF
N
N
Bn
O
N
O
N


R1O
OR1
SiR33
n
R2
H
SiR33
R1O
R1O
R2
R1O
R1O
SiR33
Br
n
R2
O
SiR33
n
R2
H
X
SiMe3
R4
OR1
OR1
n
OR1
R1O
Br
SiMe3
n
SiMe3
R4
OR1
OR1
n
R1O
OR1
SiMe3
n
SnBu3


Ph
O
CpCo(CO)2
xylenes, hν, Δ
60%
Ph
O
CoCp
SiO2
CH2Cl2
90%
Ph
O



Exposure of dihydropyridinone **1** to the arylbismuth(V) reagent (p-F-Ph)$_3$BiCl$_2$ in the presence of substoichiometric quantities of tributylphosphine results in aryl transfer to the transiently generated (β-phosphonio)enolate, ultimately providing the α-arylated enone **2**. This transformation, which represents a regiochemical complement to the Mizoroki–Heck arylation, is used strategically in concise formal and enantioselective total syntheses of the blockbuster antidepressant (−)-paroxetine (PAXIL).

OTHER CONTENT

*Corresponding author

ⓘ+ Supplementary data available via ScienceDirect

COVER

The cover graphic illustrates a projected synthesis of ovalicin which takes place through the cross-conjugated triene intermediate generated in excellent yield using a rhodium(I)-catalyzed allenic Alder-ene reaction discovered in our group.

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