

Transition metals in organic synthesis highlights for the year 1994

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Keywords: Transition metals; Organic synthesis

1. General comments

The series of annual surveys covering transition metals in organic synthesis began in 1974, when it was decided that this was an emerging area of potential but largely unrealized importance to organic synthetic chemists. This first survey had 243 references. By 1982 it had grown to 1045 references, and became unmanageable for this reviewer. Thus the topics of oxidation, reduction, and hydroformylation were split off into a separate review, reducing the 1983 review back down to 755 references. As the use of transition metals in organic synthesis gained acceptance, the number of citations grew to 1033 for 1993 and many of these were to fairly routine applications of well-established methodology. Because of this continued growth and routine use, beginning with 1994, the literature will be highlighted rather than covered in depth, and equations and detail will only be provided for unusual and significant examples, while others will be referenced but not explicitly discussed. In this manner, this review can continue to be a rather complete reference source to the current literature in the area, albeit slightly less convenient for the user, while not becoming an inordinate burden to write, type, print and read.

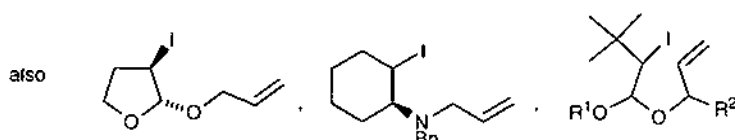
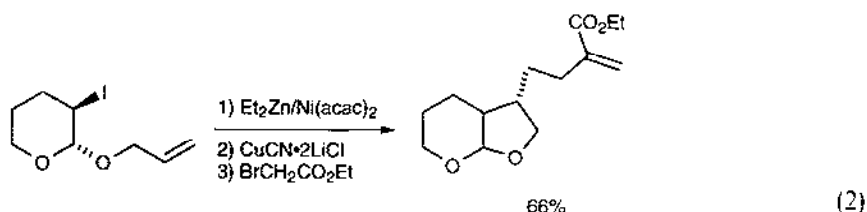
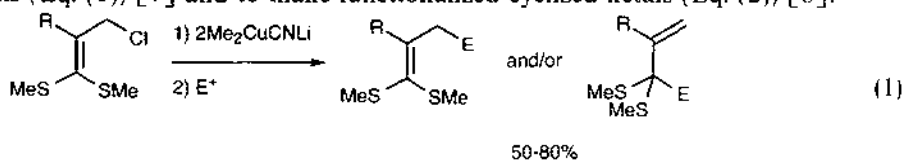
2. Carbon–carbon bond-forming reactions

2.1. Alkylations

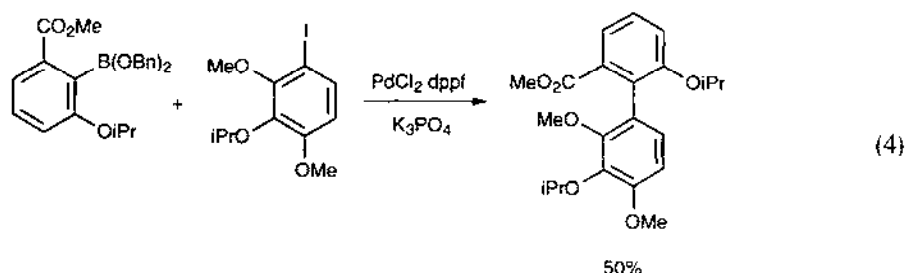
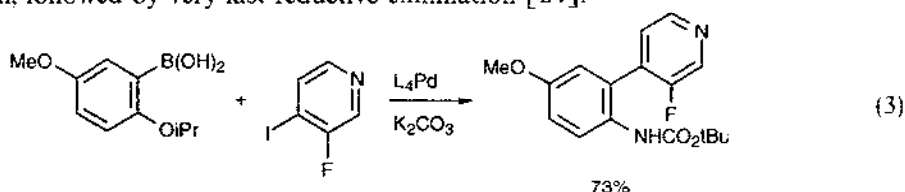
2.1.1. Alkylation of organic halides, tosylates, triflates, acetates and epoxides

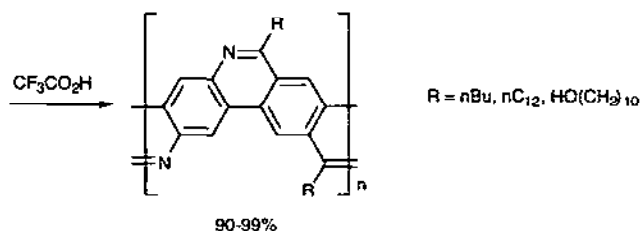
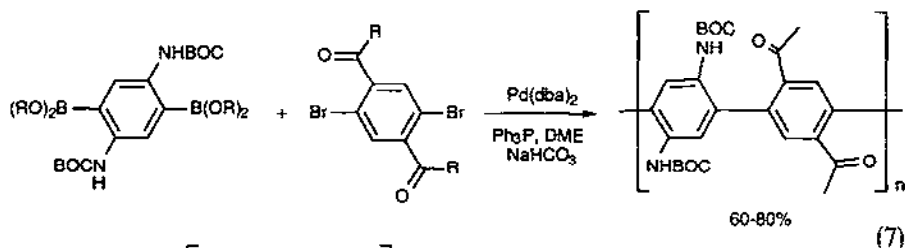
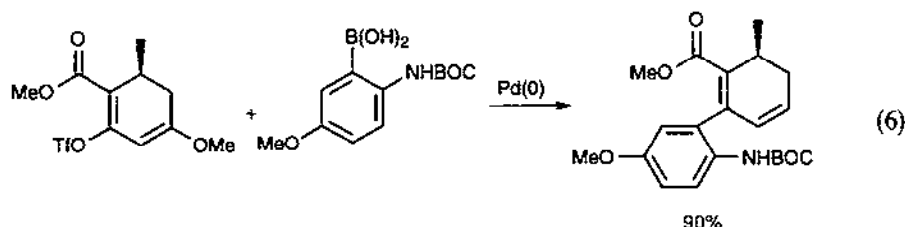
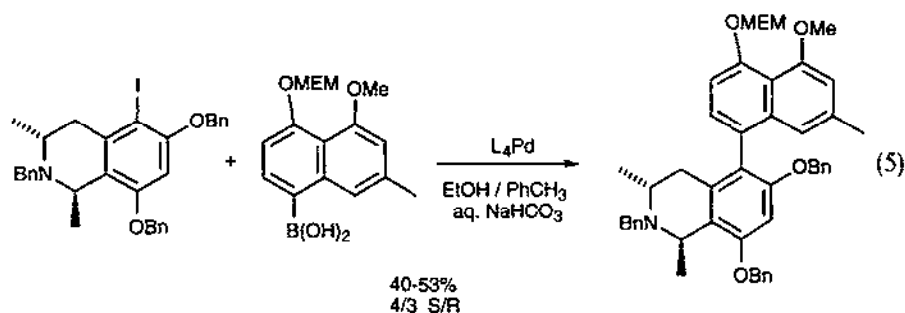
Nickel(II) complexes catalyzed the conversion of aryl dithianes to dienes by cyclopropyl Grignard reagents [1], and the alkylation of aryl sulfones by Grignards

[2]. Copper catalyzed the ring opening of aziridines by Grignard reagents [3], while palladium catalyzed the alkylation of γ -hydroxy vinyl halides [4] and α -alkoxyvinyl halides [5] by Grignard reagents. The location of CN^- in higher order cuprates has been proposed [6]. Higher order cuprates were used to functionalize ketene thioacetals (Eq. (1)) [7] and to make functionalized cyclized ketals (Eq. (2)) [8].



Palladium-catalyzed oxidative addition/transmetalation from boron (Suzuki coupling) has been extensively exploited, particularly for aryl-aryl couplings [9–13], including indoles [14,15], furans [16] and β -bromoporphyrins [17]. Complex examples are shown in Eq. (3) [18], Eq. (4) [19], Eq. (5) [20], Eq. (6) [21], and Eq. (7) [22]. The combined *ortho*-metallation/cross-coupling process for natural product synthesis has been reviewed [23]. From a kinetic study of the Suzuki coupling it was shown that the oxidative addition was the slow step, followed by fast transmetalation, followed by very fast reductive elimination [24].





Vinyl boranes [25–28] and vinyl halides [29–31] also undergo Suzuki coupling.

Palladium-catalyzed oxidative addition/fluoride-induced transmetalation from silicon (Hiyama coupling) has been used to make unsymmetrical biaryls [32] and alkylated aryls [33], and has been reviewed (19 references) [34]. Nickel or palladium catalyzed the alkylation of vinyl selenides with trimethylsilyl magnesium chloride [35].

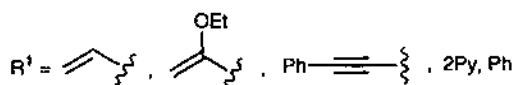
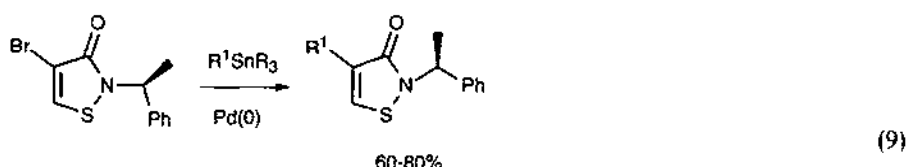
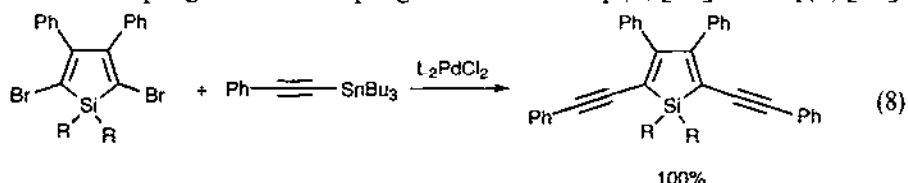
Palladium-catalyzed oxidative addition/tin transmetalation (Stille coupling) has been widely exploited in synthesis. Stille coupling is accelerated by copper(I) salts, which tie up hard phosphine ligands but not soft arsines [36]. Polymer-bound tin reagents react, leaving the tin on the polymer [37], and polymer-bound aryl iodides coupled [38]. Mono- [39] and di-stannyl alkynes [40] coupled to aryl iodides, as did allenic tins [41,42]. The production of homocoupled biaryls in attempted Stille cross coupling resulted from transmetalation to produce ArM [43].

β -Stannyl acrylates [44], β -stannyl cyclobutenones [45] and α -stannyl acrylates [46] underwent clean Stille coupling to a range of organic halides, although there

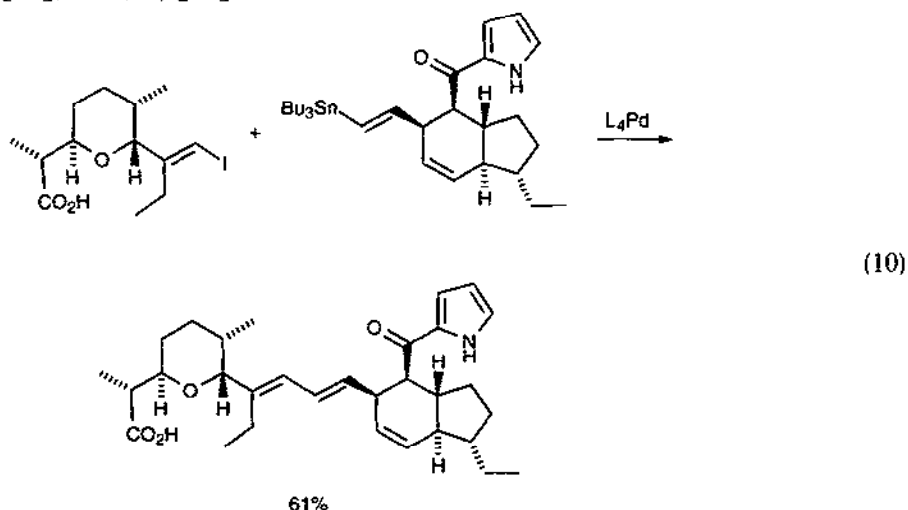
is a report that an α -stannyl acrylate resulted in β -coupling [47]. α -Ethoxystannanes also coupled effectively [48,49]. Vinyl stannanes [50,51], 1,2-bis-stannyl ethylene [52], stannyl cyclopropenes bearing β -silyl groups [53] and γ -stannyl allyl alcohol all underwent effective Stille coupling [54].

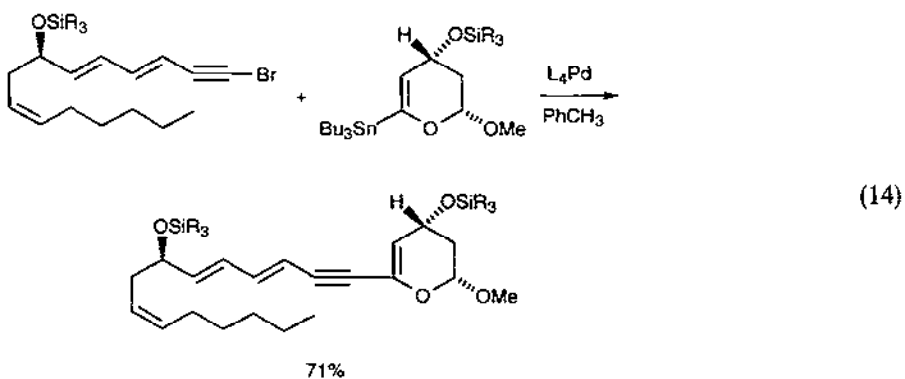
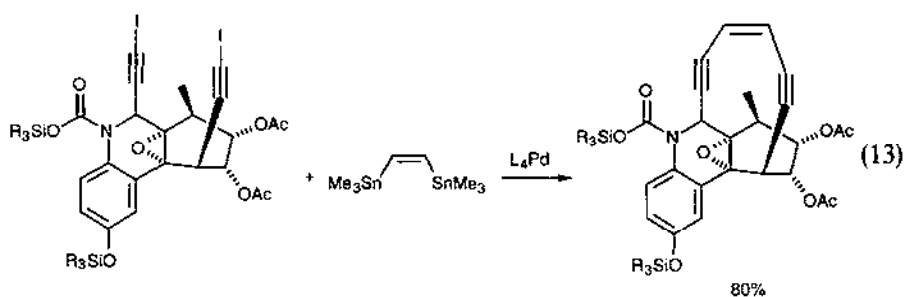
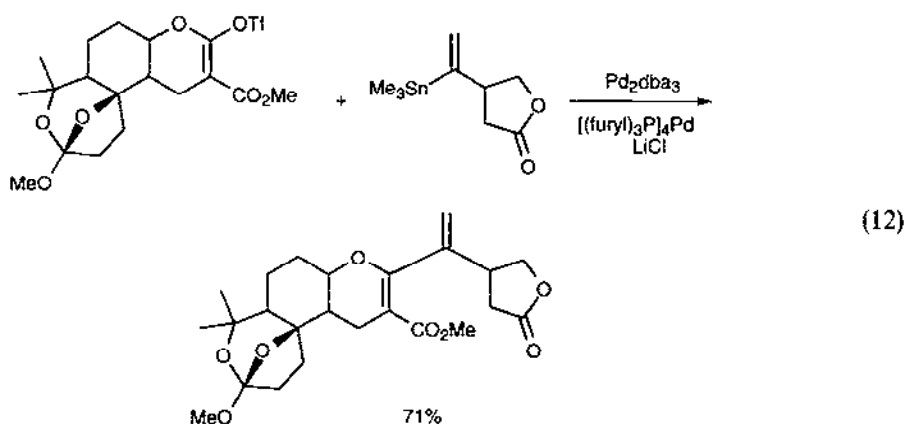
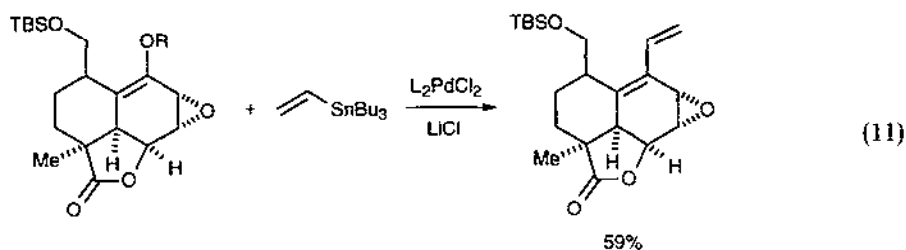
Palladium catalyzed the coupling of vinyl triflates with $\text{Bu}_4\text{NPh}_3\text{SnF}_2$ [55], and α -bromonaphthoquinones were efficiently alkylated by functionalized stannanes [56]. Amino acid esters having pendent vinyl iodides were efficiently alkylated under Stille coupling conditions [57], as were β -iodoacyl silanes [58].

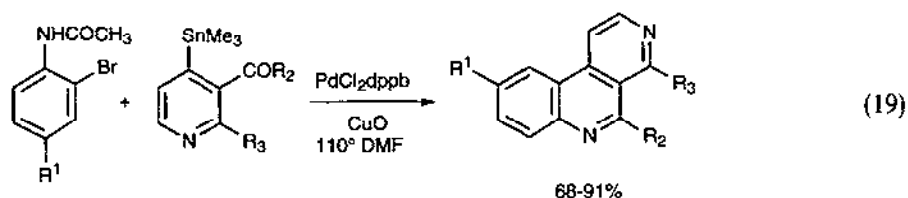
Indoles stannylated in the 2-position [59], 3-position [60] and 7-position [61] efficiently coupled to aryl- and vinyl-halides and triflates. 3-Stannylfurans [62], 5-stannyl- [63] and 4-stannyl-isoxazoles [64] and 2-stannylthiophenes [65] all underwent efficient Stille coupling. Unusual couplings are shown in Eq. (8) [66] and Eq.(9) [67].



Synthetically significant applications of Stille coupling are presented in Eqs. (10) [68], (11) [69], (12) [70], (13) [71,72], (14) [73], (15) [74], (16) [75], (17) [76,77], (18) [78], and (19) [79].

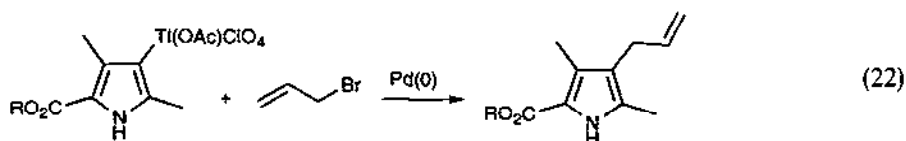
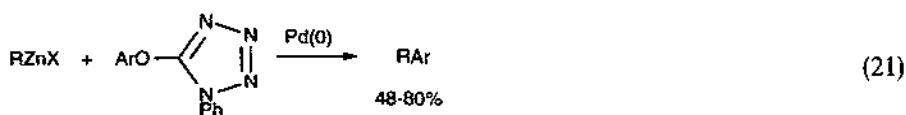
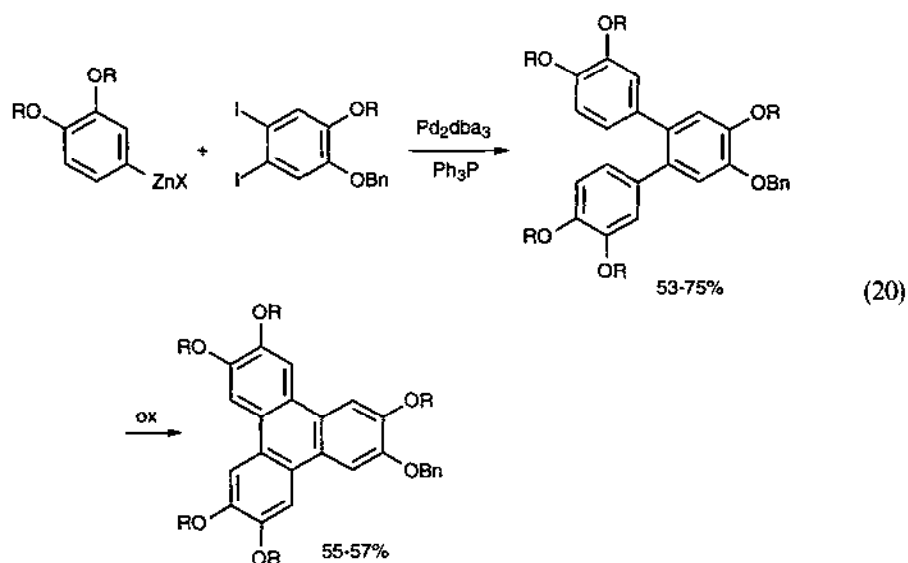






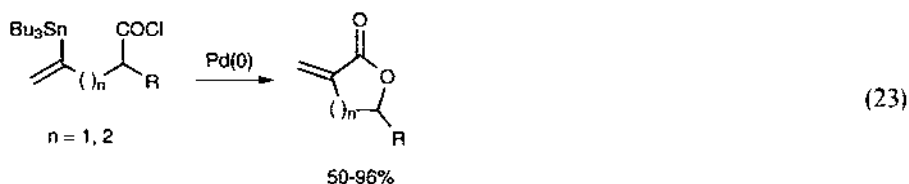
Palladium(0) also catalyzed the coupling between organozinc reagents and organic halides and triflates [80–82]. Substrates included β -halovinyl boranes [83,84], chloropyrimidines [85], and iodoenynes to make ene-dienes [86]. 3-Zincated indoles coupled efficiently to aryl and heteroaryl halides [87,88]. Functionalized phenyl alanines [89,90] and homophenyl alanines [91] were made by palladium-catalyzed coupling of aryl halides with β -zincated protected alanine.

α,β -Dibromoacrylates coupled with functionalized arylzinc reagents exclusively at the β -position [92], as did iodoallylic carboxylic acids [93]. Other interesting coupling reactions of organozincs are shown in Eqs. (20) [94] and (21) [95]. Organothallium reagents also coupled (Eq. (22)) [96].



2.1.2. Alkylation of carboxylic acid derivatives

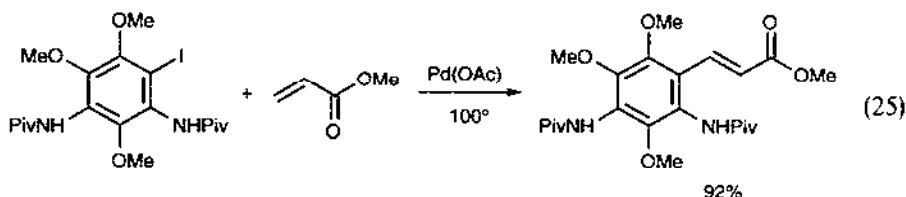
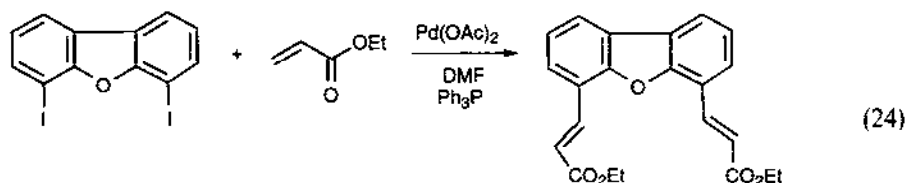
Palladium catalyzed the alkylation of acid halides by α -alkoxy stannanes [97], α -trifluoromethyl vinyl stannanes [98], β -zincated alanine esters [99], 1,2-bis(tri-butyltin)ethylene [100] and β -stannyl- α,β -unsaturated ketones [101]. α -Methylene lactones were made by an intramolecular version of this process (Eq. (23)) [102].



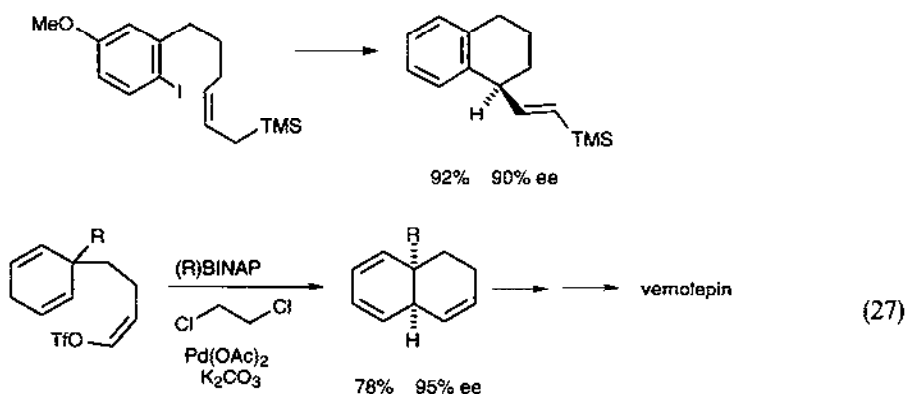
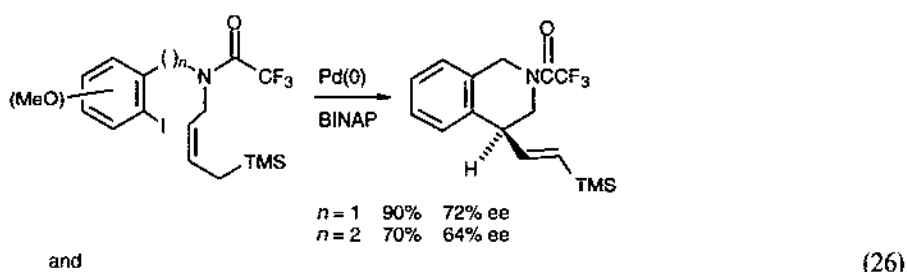
2.1.3. Alkylation of olefins

Palladium-catalyzed oxidative addition/olefin insertion (the Heck reaction) continues to be the premier method for the alkylation of olefins. Reviews entitled 'The Heck Reaction in Modern Garb' (300 references) [103] and 'Applications of Intramolecular Heck Reactions for Forming Congested Quaternary Carbon Centers in Complex Molecule Total Synthesis' (25 references) [104] have appeared. Polymer-bound phosphine palladium catalysts for the Heck reaction had long lives because the polymer prevented aggregation [105]. The Heck reaction proceeded readily in water [106], and proceeded faster and at lower temperature under high pressure (10 kbar) [107]. The origin of the chelate effect in the Heck reaction was studied [108]. Polymer-bound styrenes underwent clean Heck arylation [109]. Triaryl antimony served as an aryl source in the Heck reaction [110].

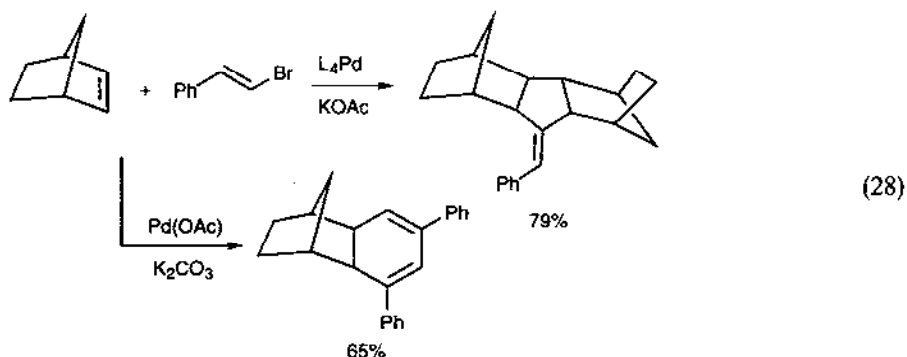
Anilines were converted to styrenes by palladium-catalyzed reaction with ethylene and butyl nitrite [111]. Bromotoluenes were converted to methyl styrenes by Heck reaction with ethylene on a 10-gallon scale [112]. Vinyl ferrocenes [113], 4-vinyl- β -lactams [114], 4-vinyl pyridine [115] and vinyl boranes [116] were arylated under Heck conditions, as were acrylic esters [117], *o*-allylcarbamates [118], 7-aza-[2.2.1]-bicycloheptenes [119] and silylketene acetals [120]. Two interesting simple Heck reactions are shown in Eqs. (24) [121] and (25) [122].

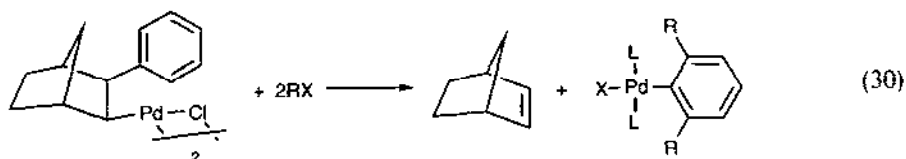
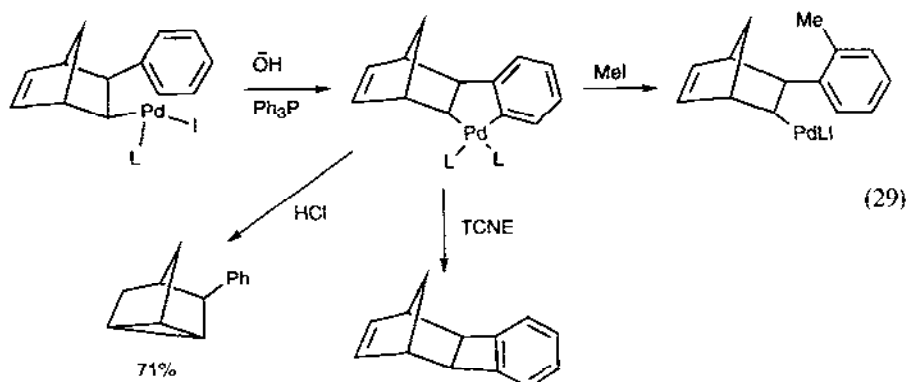


A review entitled 'Asymmetric Heck Reaction: Catalytic Asymmetric Synthesis of Bioactive Molecules' (36 references) [123] has appeared. Norbornene ring systems, including 7-oxa- and 7-aza-analogs [124] underwent asymmetric Heck arylation in up to 70% ee, utilizing BINAP [124] or aminophosphines derived from valine [125]. BINAP was the chiral ligand of choice for the asymmetric arylation of but-2-ene-1, 4-diol ketals (up to 77% ee) [126] and dihydrofuran (78% ee) [127]. Interesting asymmetric intramolecular Heck reactions are shown in Eqs. (26) [128] and (27) [129].

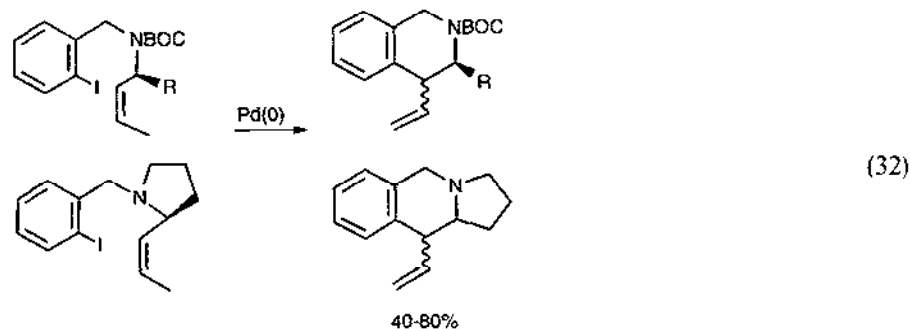
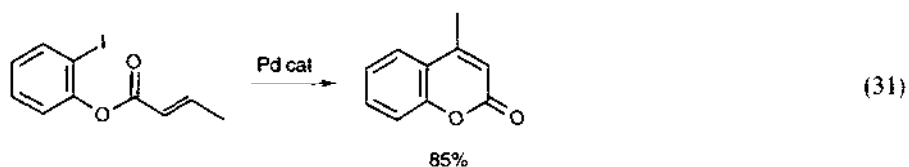


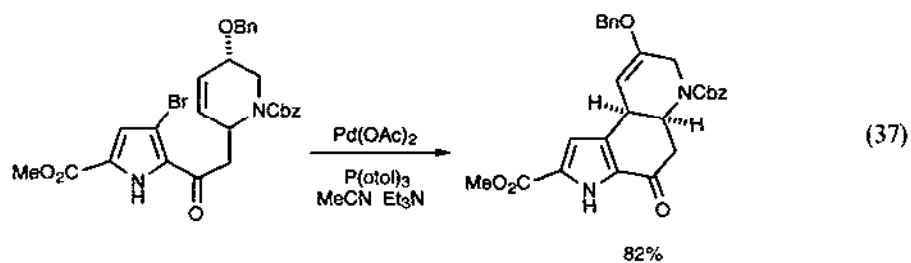
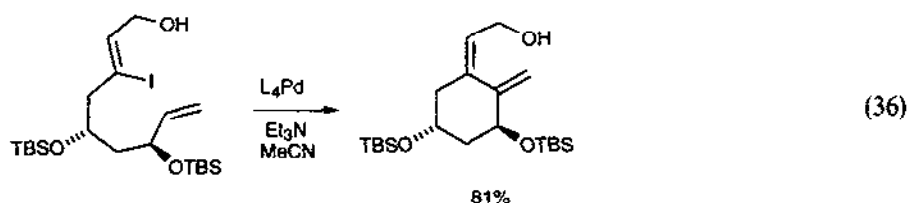
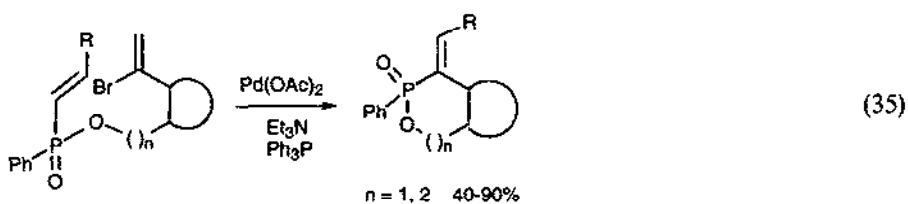
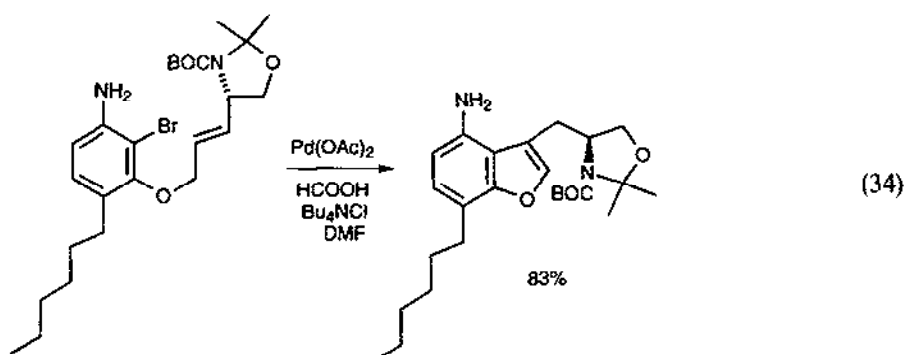
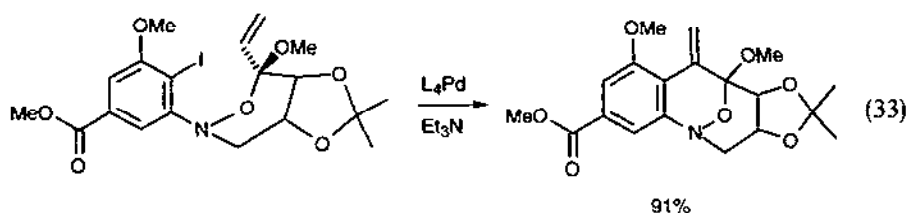
Norbornyl systems behaved strangely (Eqs. (28)) [130], (29) [131], and (30) [132].

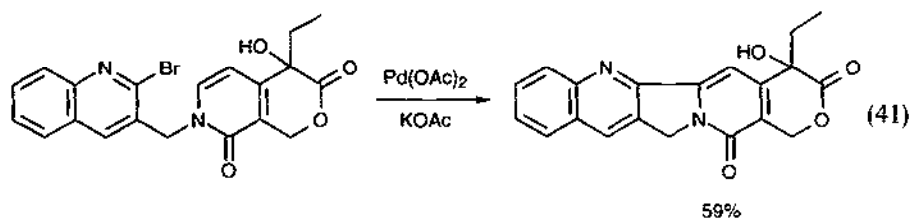
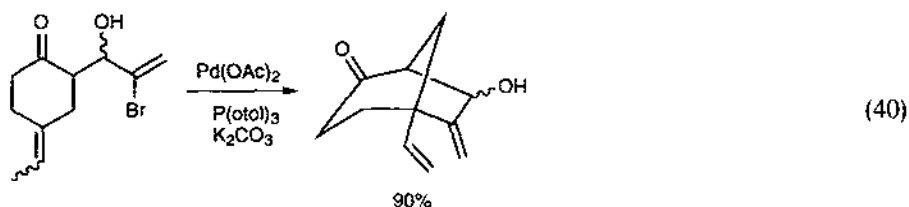
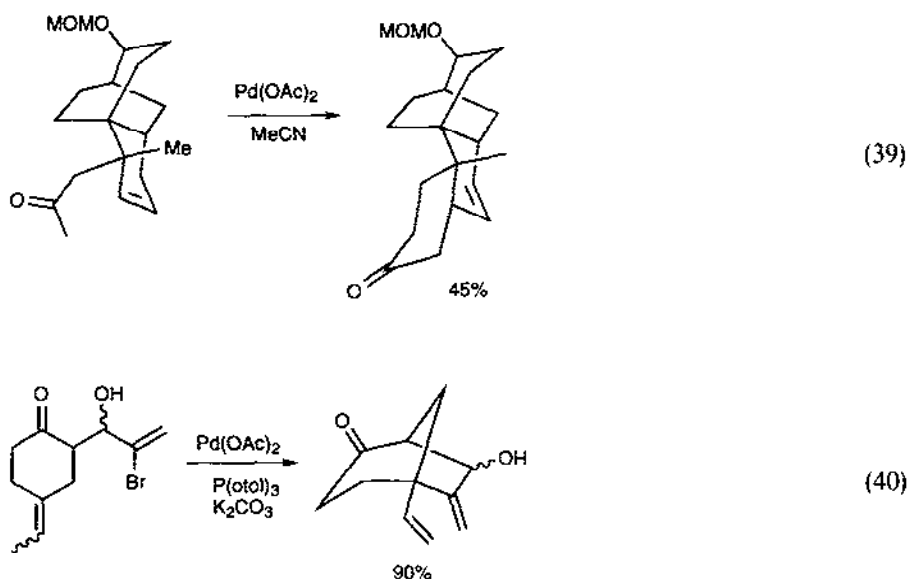
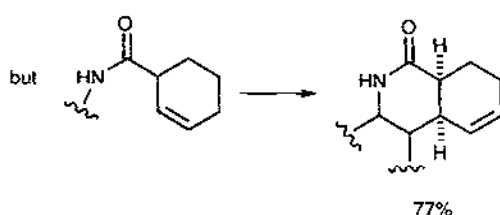
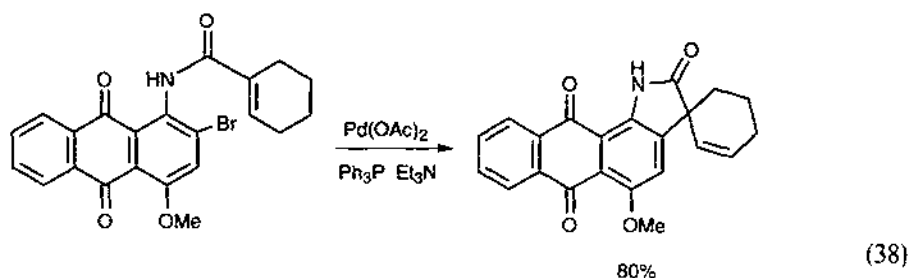


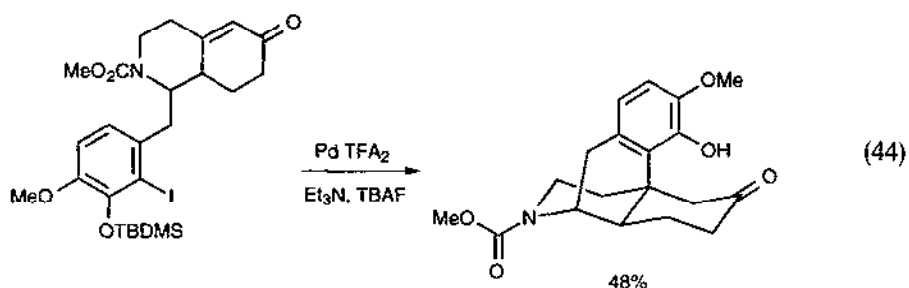
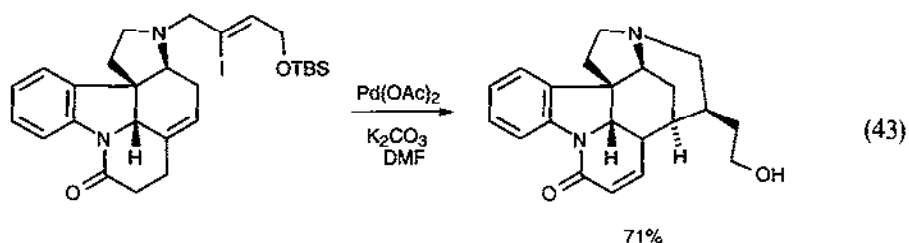
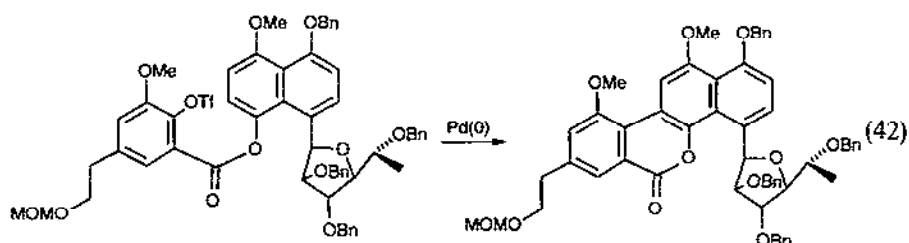


Intramolecular Heck reactions have become central to the formation of rings in the total synthesis of quite complex systems (Eqs. (31) [133], (32) [134], (33) [135,136], 34 [137], (35) [138], (36) [139], (37) [140], (38) [141], (39) [142], (40) [143,144], (41) [145], (42) [146–148], (43) [149], and (44) [150])

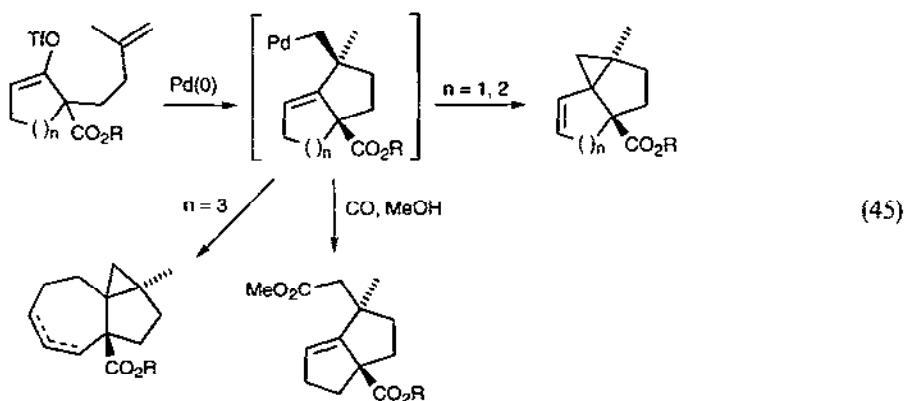


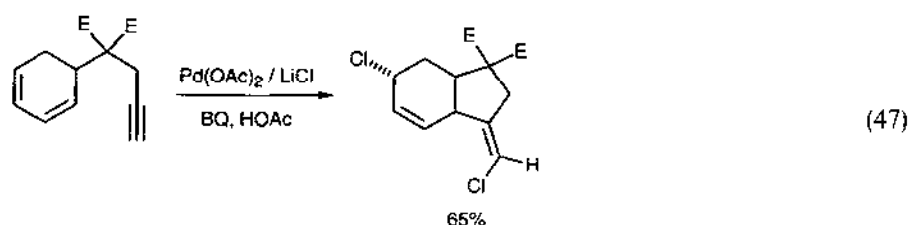
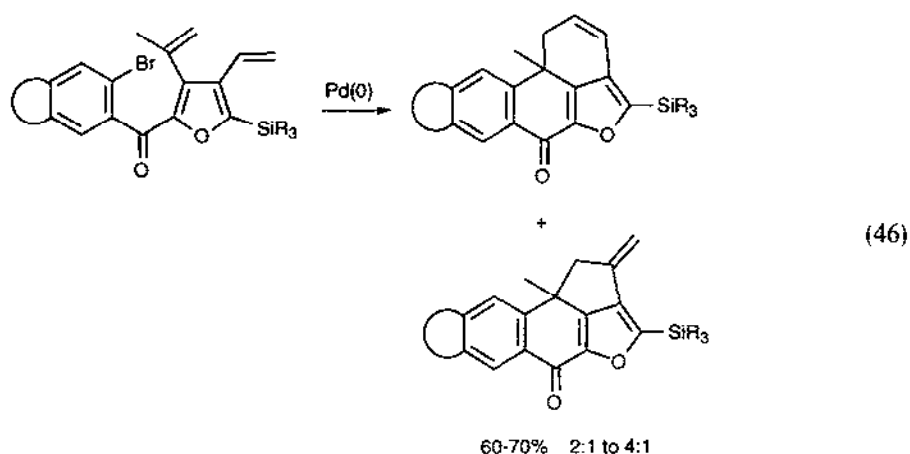




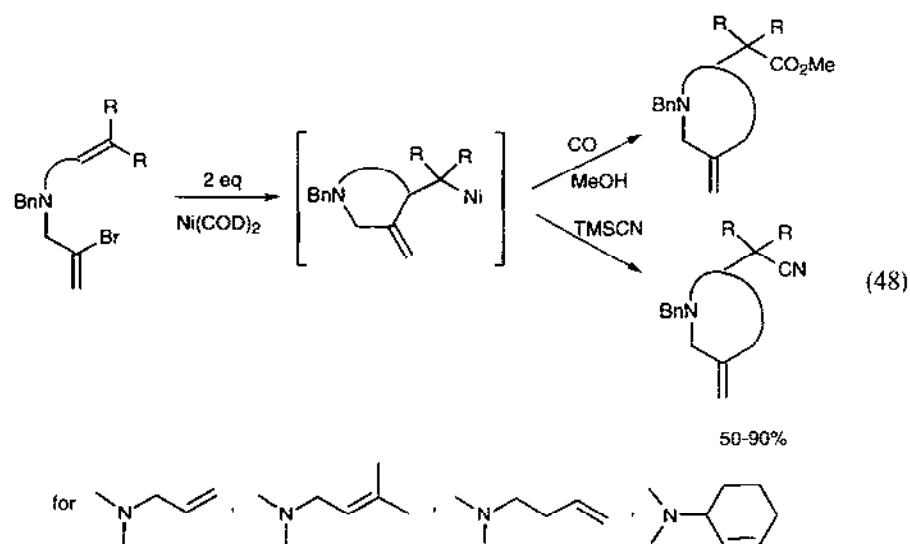


Heck type oxidative addition/olefin insertion processes need not be terminated by β -elimination. Rather, the σ -alkylpalladium complex can be trapped by nucleophilic attack by stabilized carbanions [151,152], by alkoxides [153,154], by transmetalation from tin [155,156] and by CO insertion [156]. Polyinsertion cascades have also been developed (Eq. (45)) [157] and (Eq. (46)) [158]. A slightly different cascade is shown in Eq. (47) [159].





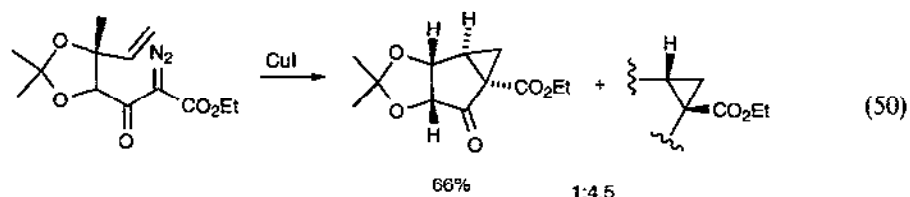
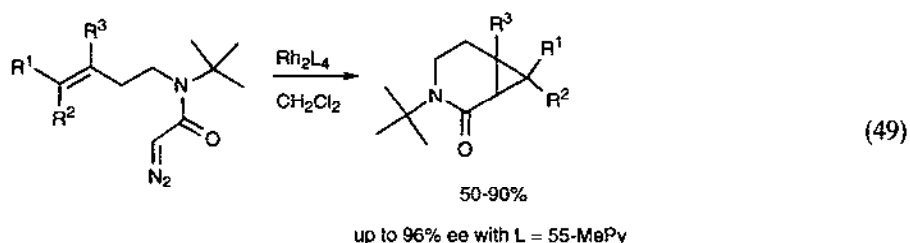
Nickel(0)-catalyzed intramolecular oxidative addition/insertion processes (Eq. (48)) [160] and the multiple reaction of conjugated enones, terminal alkynes and alkynyl stannanes to produce δ -alkynyl- γ,δ -unsaturated enones [161]. Optically active dihydropyrans were kinetically resolved by alkylations with ethyl Grignard reagents using chiral zirconocenes as catalysts [162].



2.1.4. Decomposition of diazoalkanes and other cyclopropanations

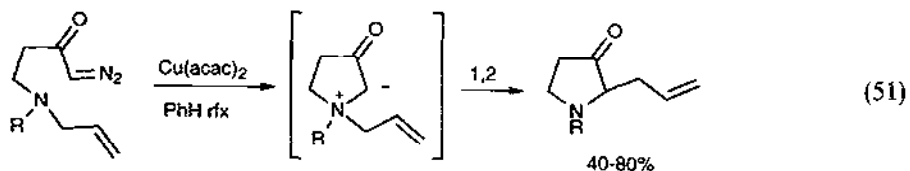
Reviews entitled 'Reactions of Diazoalkanes with Transition Metal Complexes' (145 references) [163]; 'Electron Effects in Rhodium(II) Carboxylate Linear Free Energy Relationships in Catalyzed Decomposition of Diazo Compounds' [164] and 'Ligand Effects on the Chemoselectivity of Transition Metal Catalyzed Reactions of α -Diazo Carbonyl Compounds' (177 references) [165] have appeared. Asymmetric cyclopropanations of alkenes using chiral ruthenium pyridine-bis-oxazoline (PYBOX) ligand [166], chiral sultam amide substrates [167] and chiral rhodium catalysts [168] have been achieved.

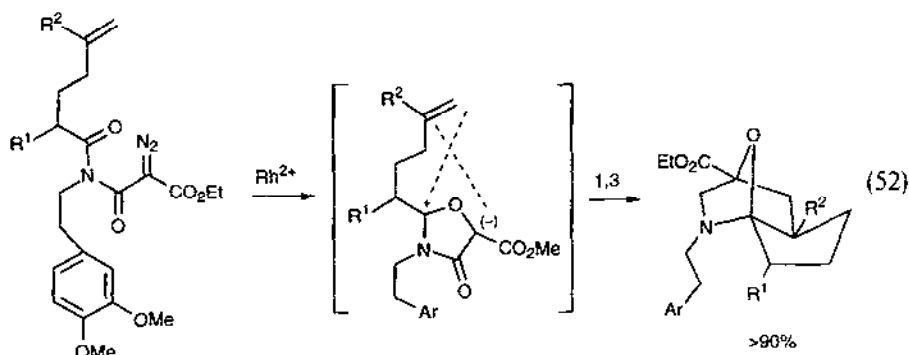
Intramolecular cyclopropanations have also been developed (Eqs. (49) [169] and (50) [170]).



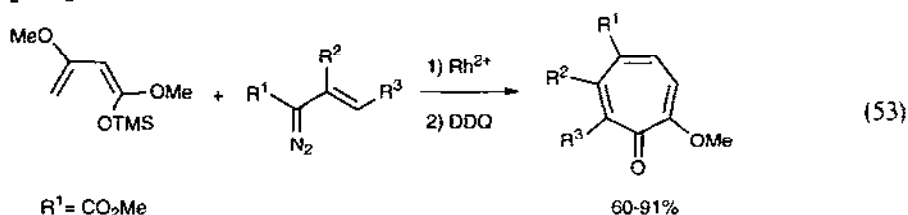
Rhodium(II) complexes catalyzed the cyclopropanation of alkynes in conjugated enynes [171], and simple alkynes (50%–98% ee with MEPEY ligand) [172] and the cyclopropanation of allenes to give methylenecyclopropanes [173]. The cycloaddition of α -diazo- β,γ -unsaturated ketones and esters to alkenes to give 2+1-cyclopropanation or 2+3-cyclopentene formation could be controlled by the nature of the R group [174]. Phenyl diazomethane cyclopropanated alkenes in the presence of $\text{CpFe}(\text{CO})_2^+$ [175].

Rhodium(II) catalyzed diazoalkane CH insertion into furans [176] and benzyl CH bonds to make optically active cyclopentanones [177,178]. Copper(II) [179] and rhodium(II) complexes [180,181] catalyzed the diazo decomposition/nitrogen or sulfur ylid formation/1,2-rearrangement process (e.g. Eq. (51) [179]). Oxonium ylid/intermolecular cycloaddition [182–184] and intramolecular cycloaddition to alkenes [185] is an efficient process. A complex example is shown in Eq. (52) [186,187].



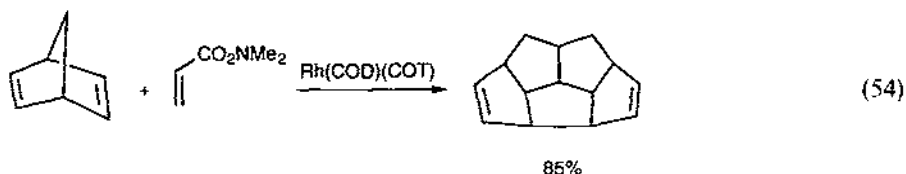


'Catalytic Decomposition of Diazomethane as a General Method for the Methylenation of Chemical Compounds' is the title of a review (284 references) [188]. Acrylic esters were made by the MeReO_3 -catalyzed decomposition of α -diazoesters in the presence of aldehydes [189]. Rhodium(II) carboxylates catalyzed the [4 + 3] cycloaddition of dienes with α -diazo- β,γ -unsaturated systems [190] (Eq. (53)) [191].

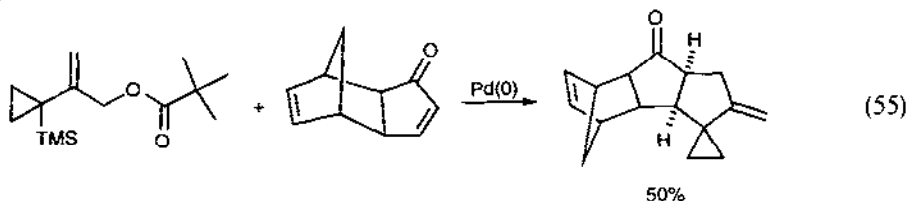


2.1.5. Cycloaddition reactions

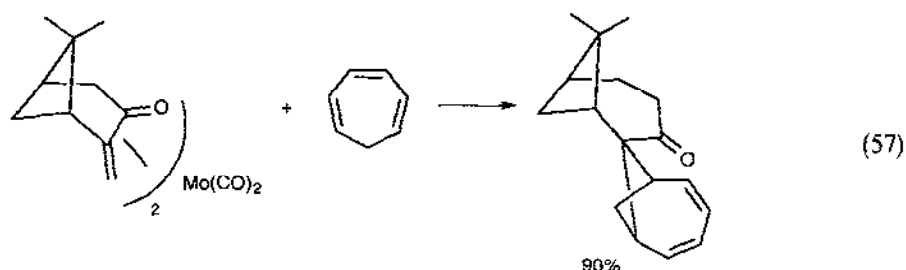
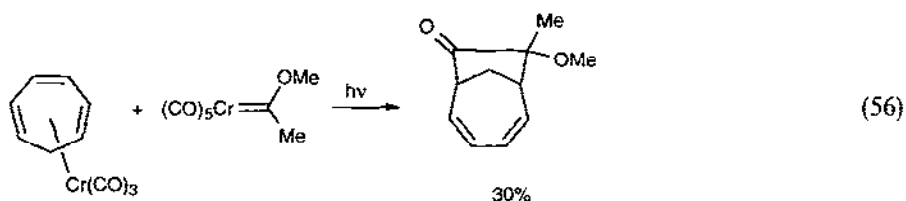
The affects of 7-position functionality on nickel(0)-catalyzed 4 + 2 cycloadditions of alkenes to norbornadienes have been studied [192]. With rhodium catalysts the strange reaction shown in Eq. (54) occurred [193].



Palladium(0)-catalyzed 'trimethylene methane' cycloadditions continued to be developed (Eq. 55) [194,195]. Aldehydes cycloadded to 2-trimethylstannylmethyl- π -allylpalladium halides to give methylene tetrahydrofurans [196].

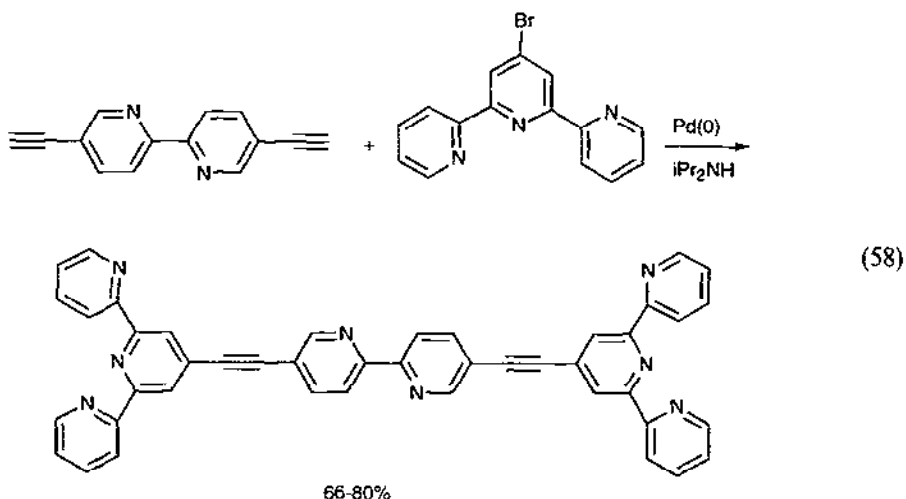


Higher order cycloaddition reactions in natural products synthesis has been reviewed (9 references) [197]. Examples are shown in Eqs. (56) [198] and (57) [199]. Iridium(III) alkyne-metallacyclobutane complexes underwent an oxidatively driven reductive elimination to produce cyclopentenones [200].

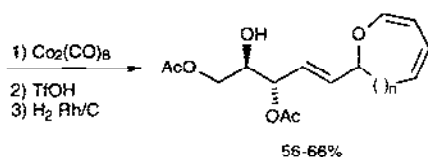
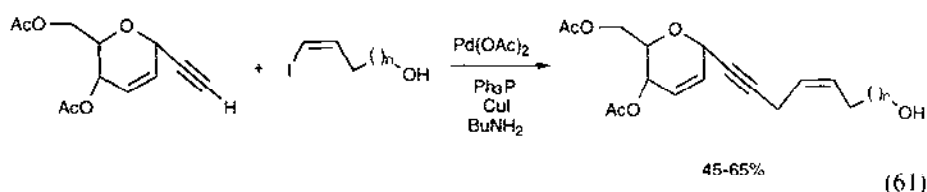
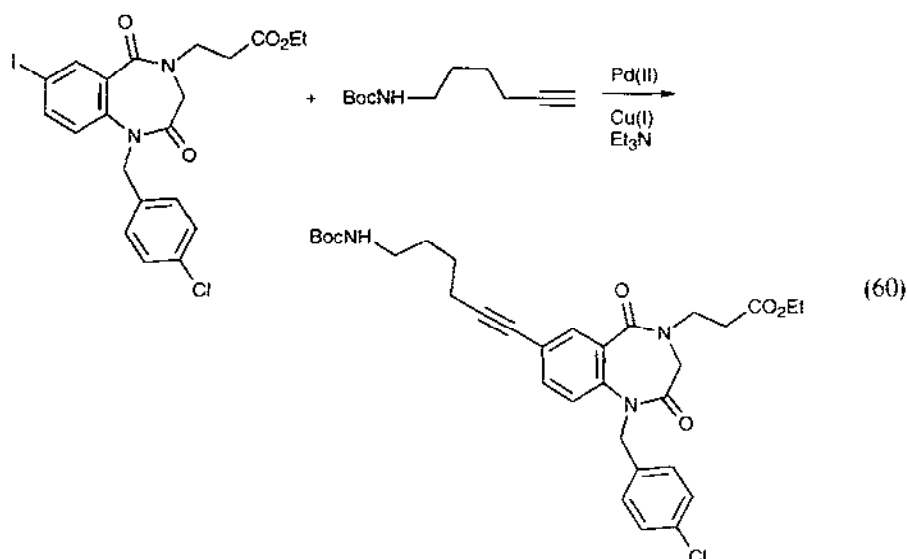
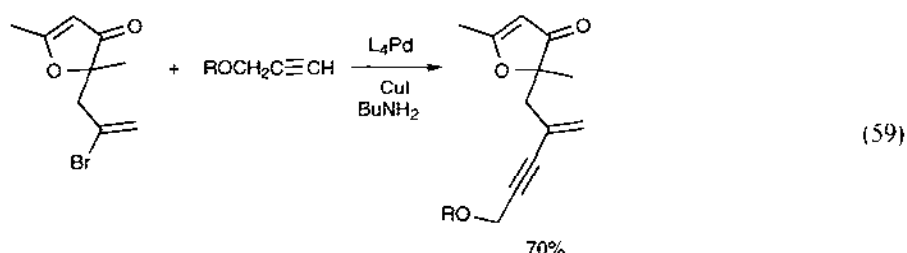


2.1.6. Alkylation of alkynes

The palladium/copper-catalyzed coupling of terminal alkynes with aryl and vinyl halides (the Castro–Stevens reaction) has been used in many contexts including the alkylation of functionalized aryl halides [201–203], the use of cyclopropyl alkynes [204], and alkylation of porphyrin aryl triflates [205], α -bromostyrenes [206], and triaryl ethers [207]. Complex acetylenic polypyridines were made this way (Eq. (58)) [208], and triflates of 2-hydroxypyridines, quinolines and isoquinolines [209],

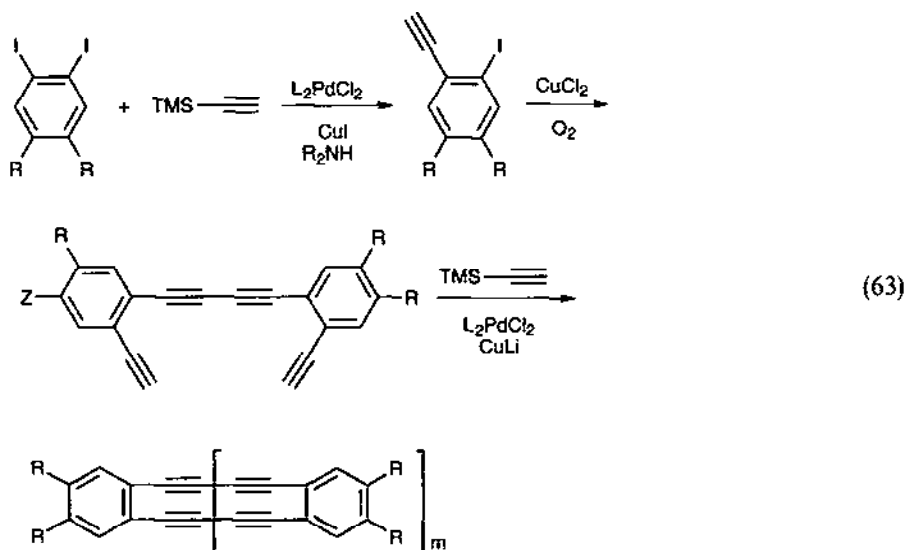
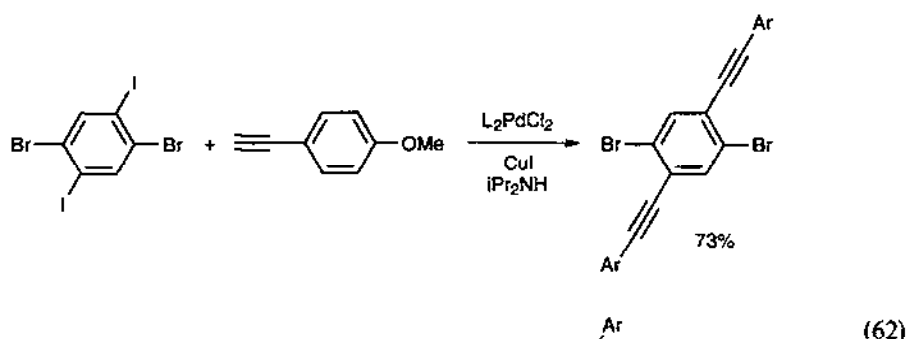


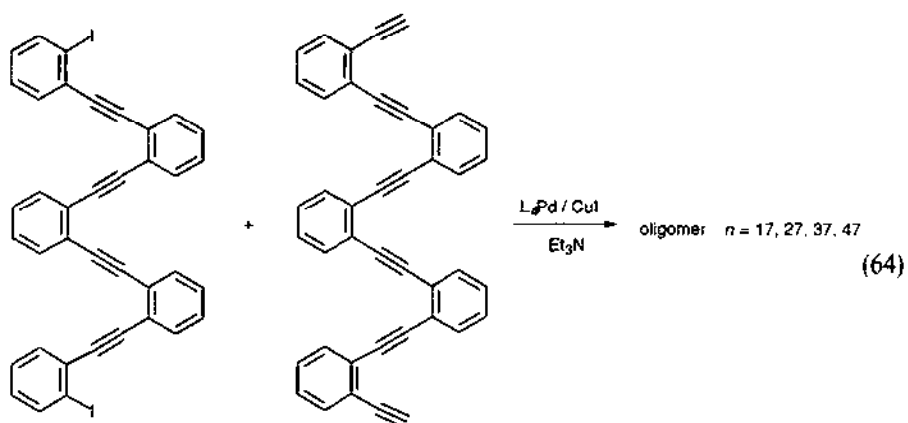
3-bromopyridines [210] and 3-iodothiophenes [211] were all alkynylated by this procedure. As seen in Eqs. (59) [212], (60) [213] and (61) [214], it works well in complex systems.



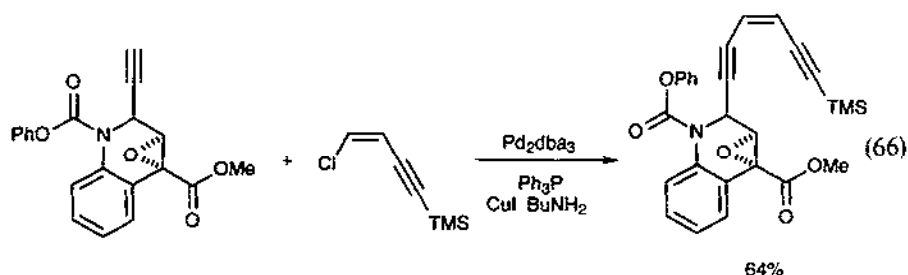
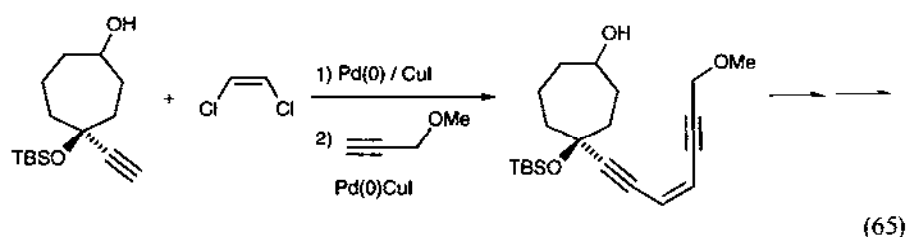
This process was particularly used for the synthesis of polyacetylenic systems by alkynylating *o*-diiodobenzene with acetylenic alcohols [215], *p*-diiodobenzene with polyalkynes [216], bis- β -bromo-*p*-divinyl benzene with trimethylsilyl acetylene fol-

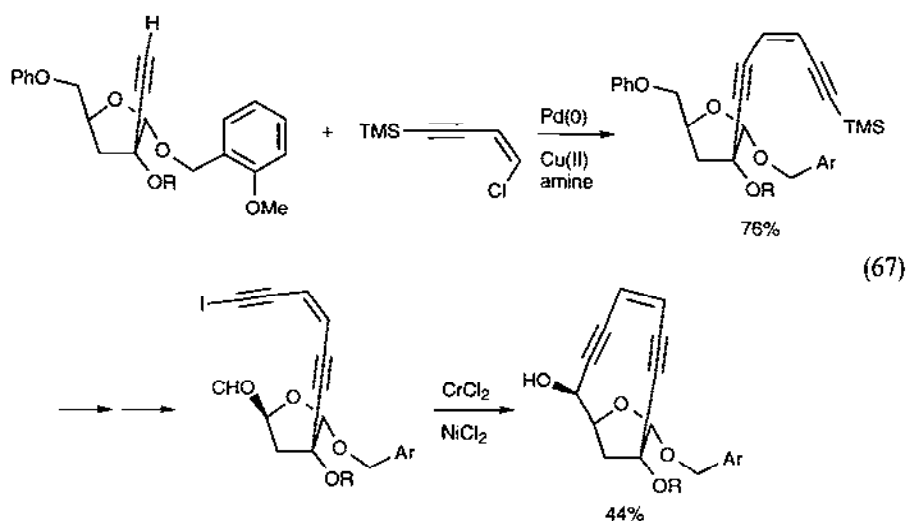
lowed by coupling [217], 9,10-diiodoanthracene with alkynes and diynes [218], geminal vinyl bromides with alkynes to produce tetraalkynylalkenes [219], dibromobinaphthols with trimethylsilyl acetylene [220], 1,4-bis-(2-iodophenyl)butadiyne with trimethylsilyl acetylenes [221] and polystannylidienes with enynes and diynes [222]. Many of these were of interest in the context of polymers and materials (Eqs. (62) [223], (63) [224] and (64) [225].



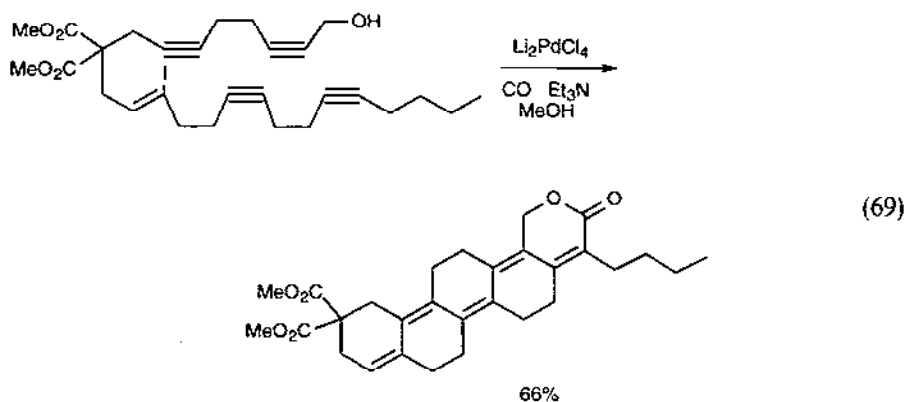
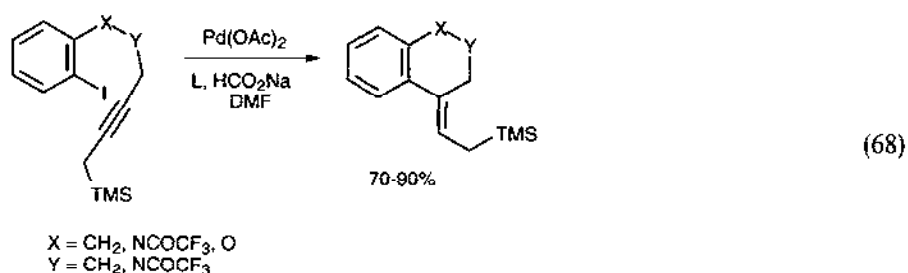


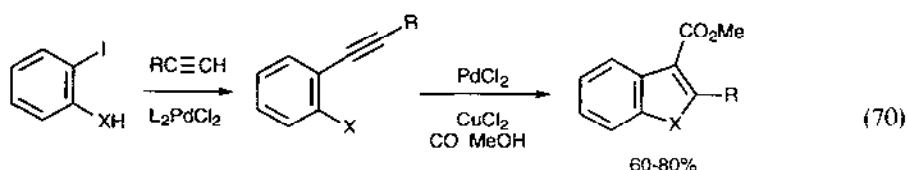
The Castro–Stevens coupling was central to the synthesis of ene-diynes [226–231] some of which were used to make polymers by thermolysis [232], and others to feed into various ene-diyne antitumor agent syntheses (Eqs. (65) [233], (66) [234,235] and (67) [236].



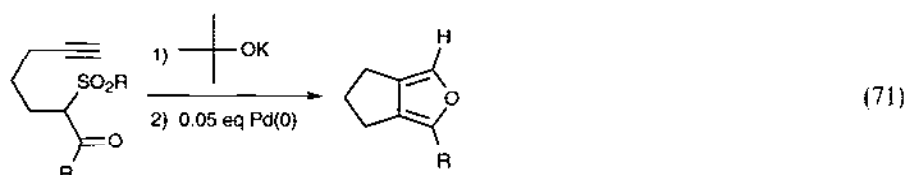


Alkynes were alkylated via palladium-catalyzed oxidative addition/alkyne insertions (Eqs. (68) [237], (69) [238], (70) [239] and (71) [240]). Palladium(II) catalyzed the addition of aroyl cyanides to alkynes [241].



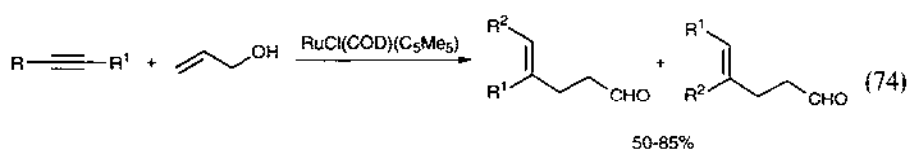
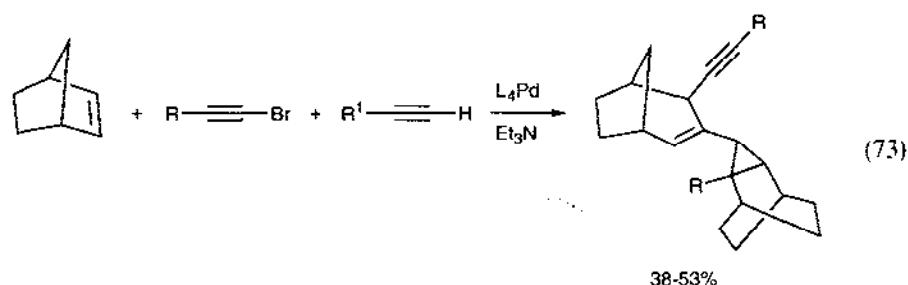
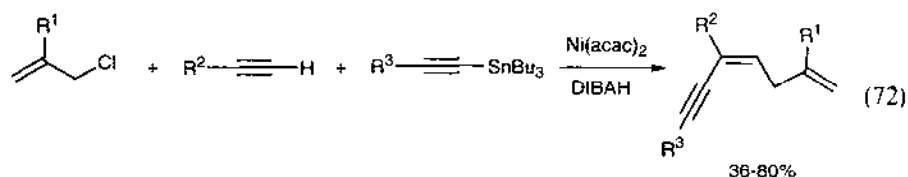


X = O, NH, NSO₂Me



CpCo(CO)₂-catalyzed the alkylation of alkynes by stabilized carbanions [242,243]. Mixtures of TaCl₅ and zinc formed tantalacycloprenes which coupled to alkenes [244] and alkylated imines [245]. 1,6-Hexadiyne was converted to 1,9-dihydroxy-3,7-dimethylnona-2,7-diene [246].

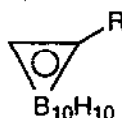
Miscellaneous alkyne alkylations are shown in Eqs. (72) [247], (73) [248] and (74) [249].



2.1.7. Alkylation of allyl, propargyl and allenyl systems

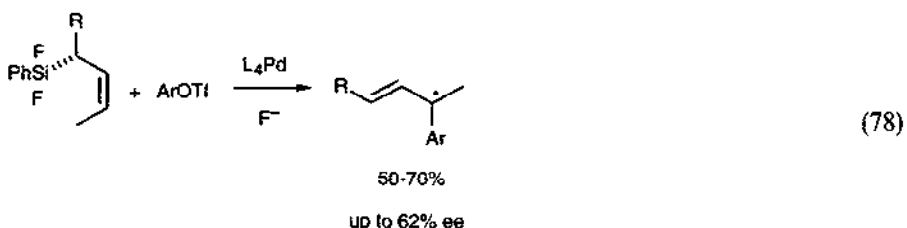
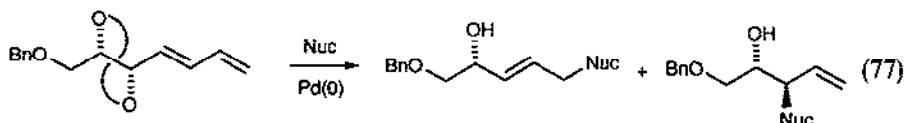
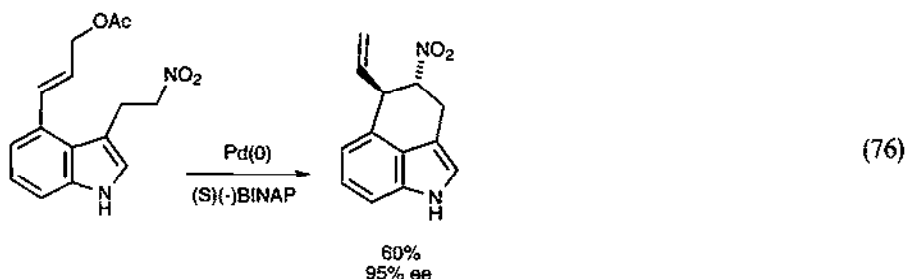
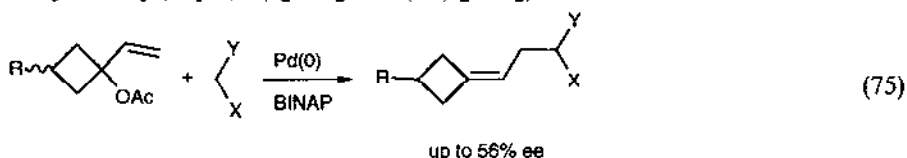
Palladium-catalyzed alkylation of allyl system continues to be developed. By using sterically encumbered phenanthrolines as ligands the stereochemistry of the alkene in the product could be controlled [250]. Palladium(0)-catalyzed the allylic alkyla-

tion of vinyl epoxides by sulfone stabilized carbanions of polyisoprenes [251], allyl acetates by dimethylfulvene [252], allyl carbonates by

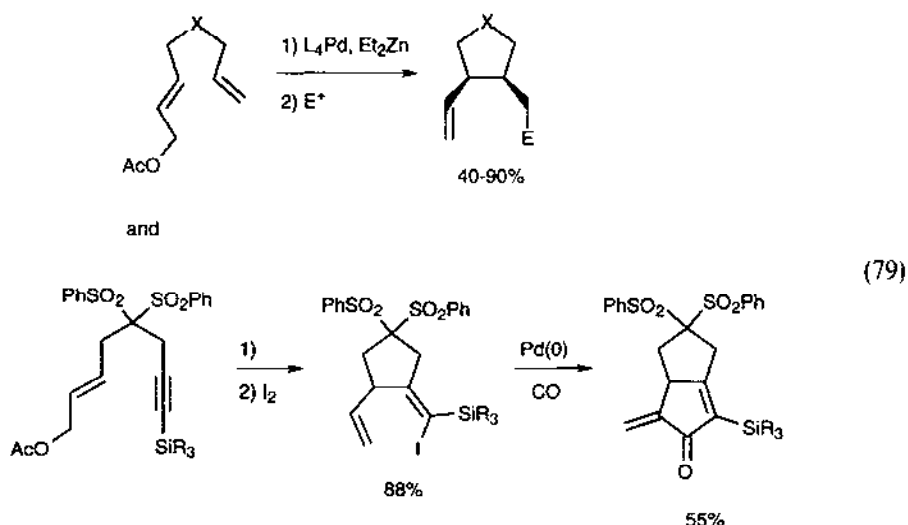


anions [253] and alkyl boronates [254], 2-tributylstannyl-allyl acetates by aryl stannanes [255] and 3-phenyl propenol by stabilized carbanions [256]. Palladium-catalyzed allylic alkylation was studied by NMR spectroscopy [257].

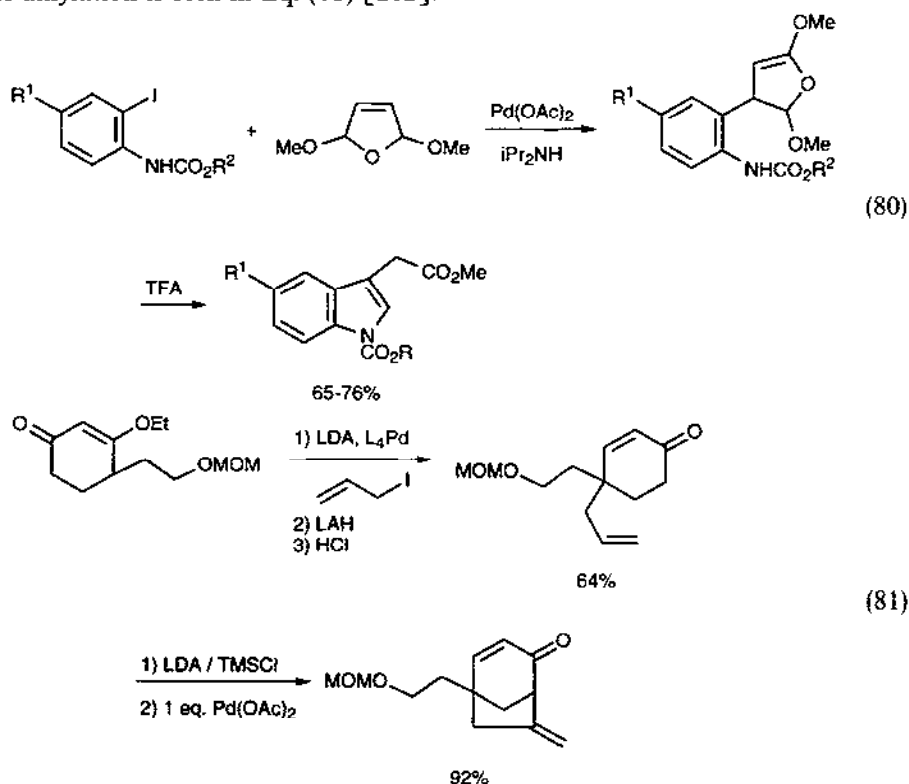
Palladium-catalyzed asymmetric allylic alkylation has been popular this year [258], and many new ligands for the asymmetric alkylation of racemic 1,3-diphenylprop-1-ene-3-ol acetates have been developed. These include aryl phosphines containing one *ortho* chiral oxazoline [259,260], diaryl sulfides with the same *ortho* chiral oxazoline [261], chiral 1,2-diamines [263] and aminophosphines [264–266]. Interesting applications are seen in Eq. (75) [267] and Eq. (76) [268]. Cyclic chiral enamines alkylated allylic acetates with fair to good ee [269], and chiral substrates also were used to induce asymmetry (Eqs. (77) [270] and (78) [271]).



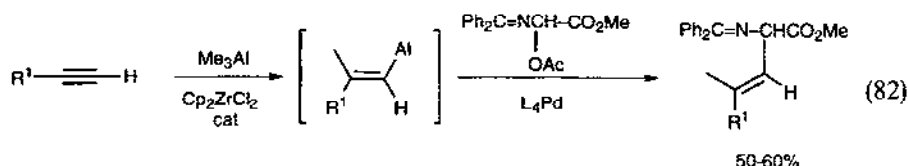
Intramolecular alkene insertion into π -allylpalladium complexes formed by oxidative addition of allyl sulfones [272], acetates [273,274] and carbonates [275] has been used to make interesting cyclic systems (for example, Eq. (79)) [276].



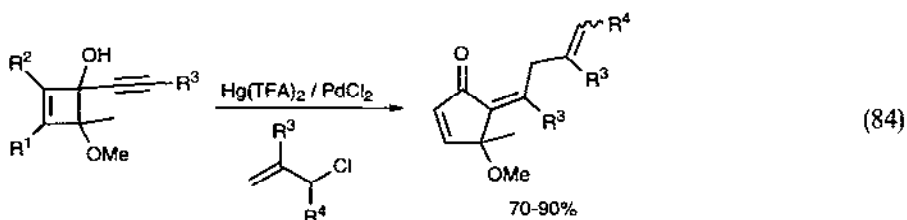
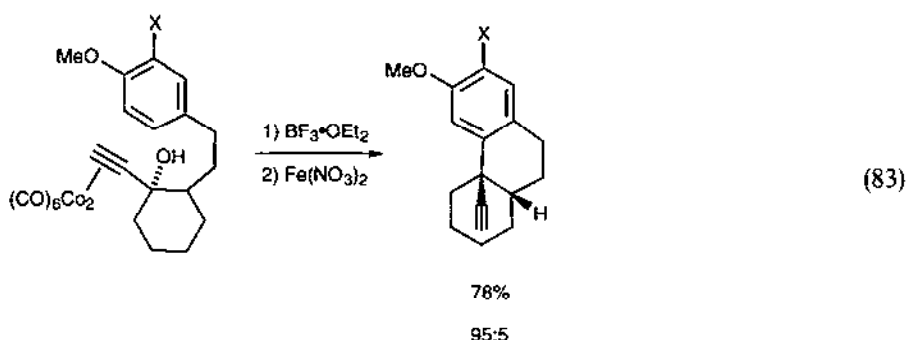
Allylic alcohols were alkylated by palladium(0)-catalyzed oxidative addition/olefin insertion processes. Secondary alcohols gave ketones [277] while tertiary alcohols incorporated as allyl alcohols [278,279]. Allyl ethers gave enol ethers (Eq. (80) [280]), while allyl trifluorosilanes gave alkenes [281]. An interesting application of allylic alkylation is seen in Eq. (81) [282].



The regiochemistry of arylation of allylic chlorides by aryl Grignard reagents was a function of rate of addition, temperature and copper concentration [283]. Optically active nickel–phosphine complexes catalyzed the asymmetric alkylation of allyl phenyl ethers by Grignards with good ee [284]. Vinyl glycines were made by a palladium-catalyzed coupling process (Eq. 82) [285].

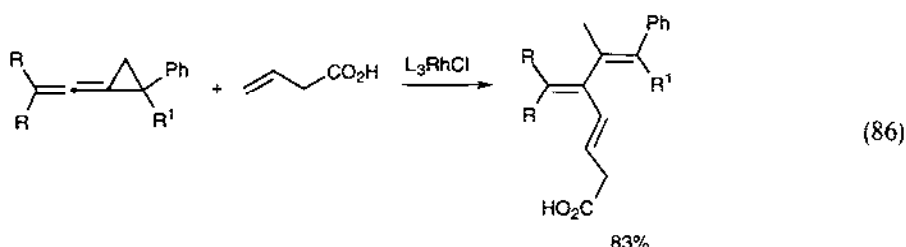
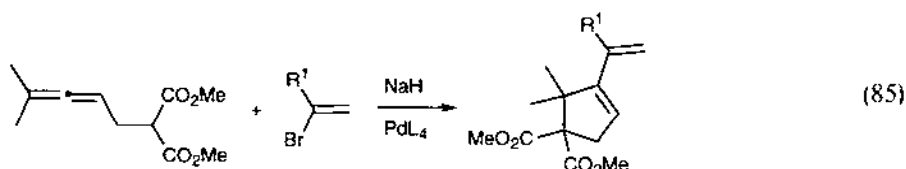


A review entitled 'A General Strategy Using $\eta^2\text{Co}_2(\text{CO})_6$ Acetylene Complexes for the Synthesis of Eneidyne Antitumor Agents' (19 references) has appeared [286]. Electron-rich arenes alkylated cobalt-stabilized propargyl cations with high stereo-selectivity [287] (Eq. (83)) [288]. Enamines alkylated molybdenum stabilized propargyl cations [289]. Cobalt complexed propargyl aldehydes underwent alkylation by silylketene thioacetals with high *syn* selectivity [290]. Palladium(0) catalyzed the alkylation of propargyl carbonates by alkyl boranes [291] and alkynes [292] to give allenes. A more complex system is seen in Eq. (84) [293].



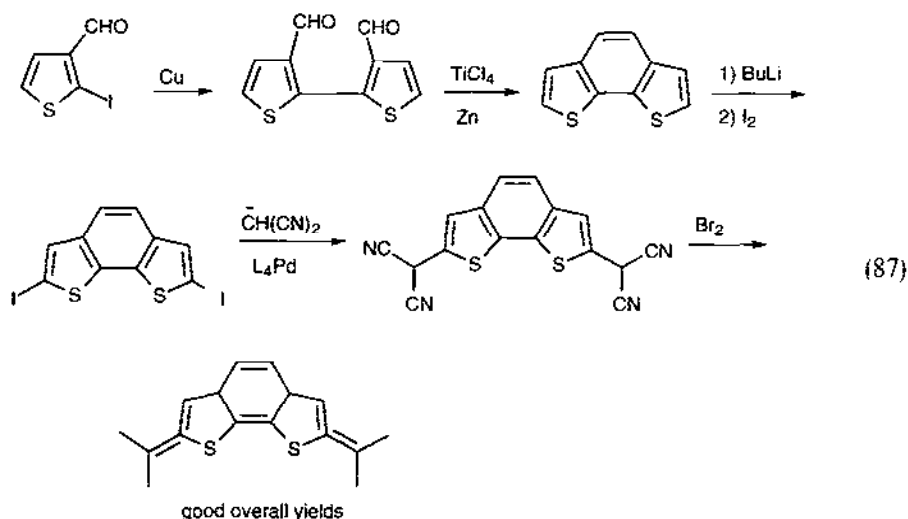
Palladium(0) catalyzed the alkylation of 2,3-dienic (allenic) carbonates by alkyl boranes [294], allenes by nitrile stabilized anions [295] and allenes by vinyl halides via oxidative addition/alkene insertion [296]. These intermediates could be intercepted by carbanions both intramolecularly (Eq. (85)) [297] and intermolecu-

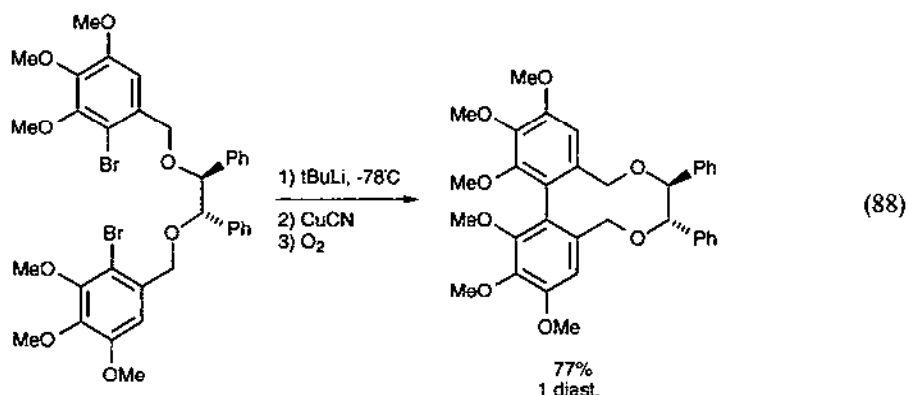
larly [298]. Rhodium(I) catalyzed the alkylation of allenes by alkynes to give enynes [299], as well as the strange reaction shown in Eq. (86) [300].



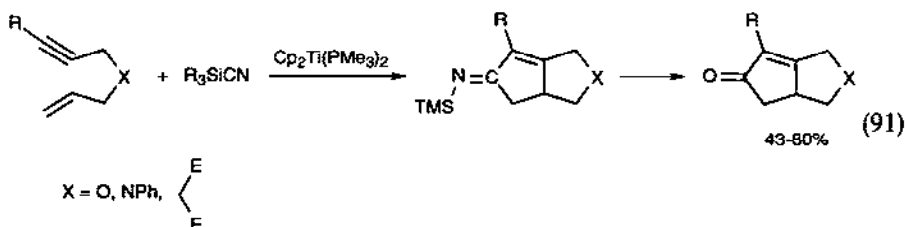
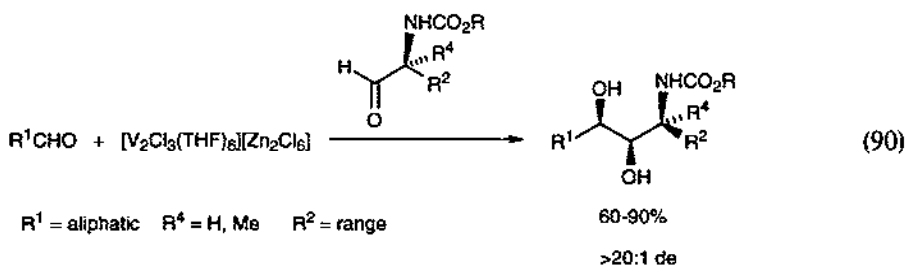
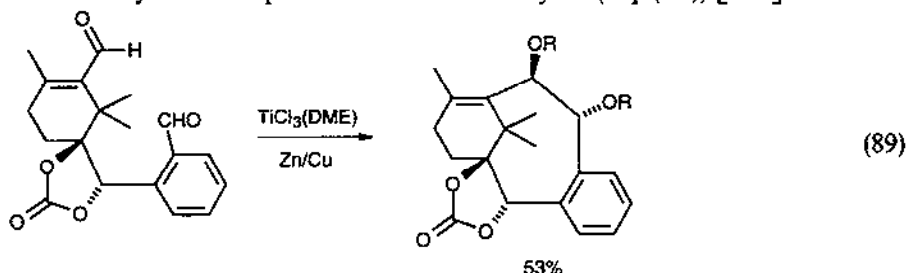
2.1.8. Coupling reactions

Aryl bromides were selectively cross-coupled to aryl chlorides using nickel(II) phosphine complexes in pyridine [301]. Copper metal was used to couple 2-iodo-3-formyl thiophene at the 2-position. Treatment with reduced titanium coupled the aldehydes (Eq. (87)) [302]. Biaryls linked at the *o*-positions were synthesized by low temperature lithiation/transmetalation to copper/oxidation [303] (Eq. (88)) [304,305]. Optically active biaryls were produced by copper powder coupling of chiral *o*-bromophenyl oxazolines [306,307]. Highly functionalized optically active binaphthyls were synthesized from naphthalenes having optically active side chains using the low temperature oxidation of cuprates [308].



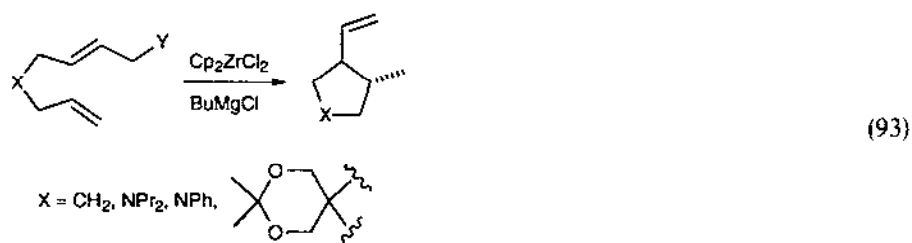
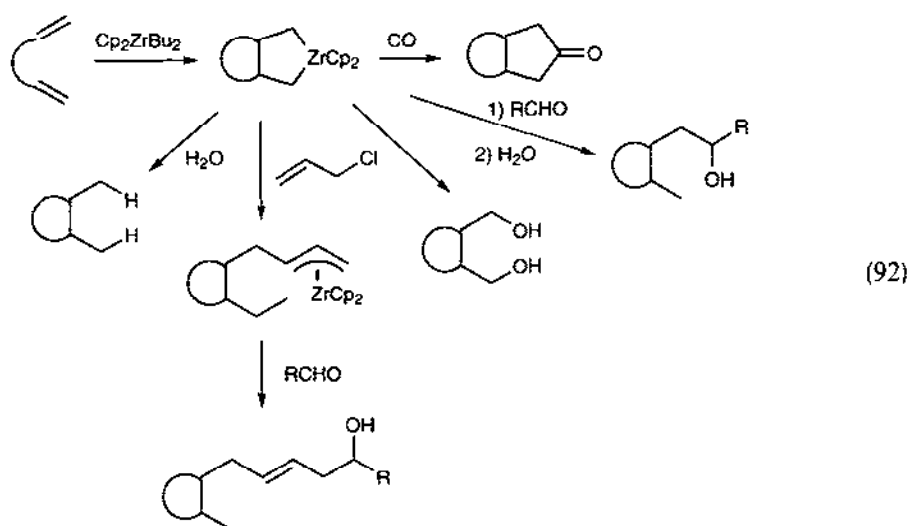


The coupling of carbonyl compounds by reduced titanium species continues to be improved. Long chain polyisoprenyl keto esters were cyclized to conjugated ketones [309,310], long chain dialdehydes (36 carbons) were cyclocoupled to the cyclic alkene [311] (poly-ether backbone), [312], strained cyclic diketones coupled to form polyolefins [313], and *o*-benzoyl acetanilides coupled to form indoles [314]. Under modified conditions, the reductive coupling could be stopped at the diol stage, forming tetrahydrothiophene-3,4-diols from the appropriate diketone [315], and ten-member cyclic diols from keto aldehydes [316]. This coupling has been used in the context of taxane synthesis (Eq. (89)) [317,318]. Reduced vanadium species stoichiometrically cross-coupled two different aldehydes (Eq. (90)) [319].

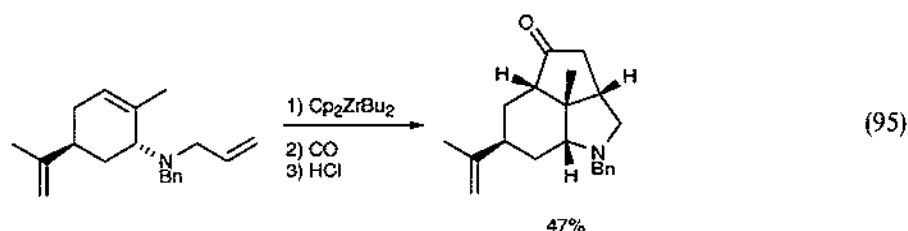
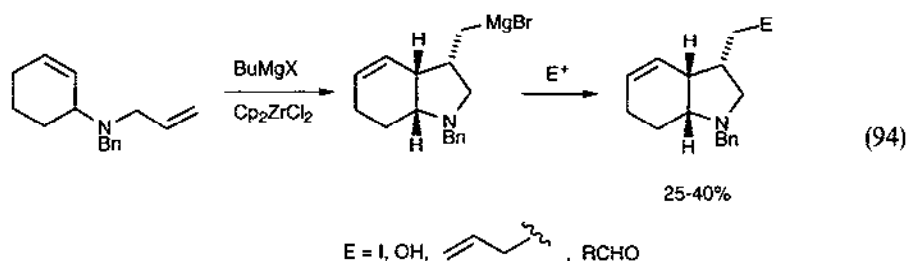


Reduced titanocene species dimerized alkynes with formaldehyde to give 3-hydroxypent-1,4-dienols [320], and cyclocoupled enynes with trimethylsilyl cyanide (Eq. (91)) [321]. Appropriately situated trienes were cascade-cyclized to decalins by $\text{Et}_2\text{AlCl/Ti}(\text{OiPr})_4$ [322].

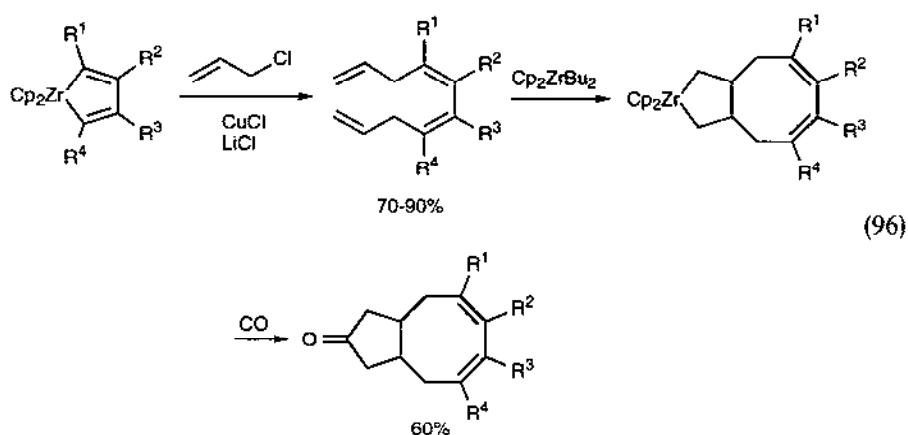
Reduced zirconium species have been extensively used for cyclocoupling. A review entitled 'Patterns of Stoichiometric and Catalytic Reactions of Organozirconium and Related Complexes' (58 references) has appeared [323]. Dienes underwent cyclocoupling with Cp_2ZrBu_2 to produce zirconacyclopentanes (Eq. (92)). These inserted allylic halides to give η^3 -allyl zirconium complexes [324,325], aldehydes to give alcohols after hydrolysis [326], isonitriles [327], carbon monoxide to give cyclopentanones [328]; they also undergo oxidation to give diols (O_2) or dibromides (Br_2) [329], and hydrolysis to give hydrocarbons [330,331]. In a related process, zirconocene dichloride catalyzed the cyclodimerization of alkenes with alkyl aluminum [332] or magnesium reagents [333,334] (Eq. (93)). The mechanism of this reaction was studied [335].



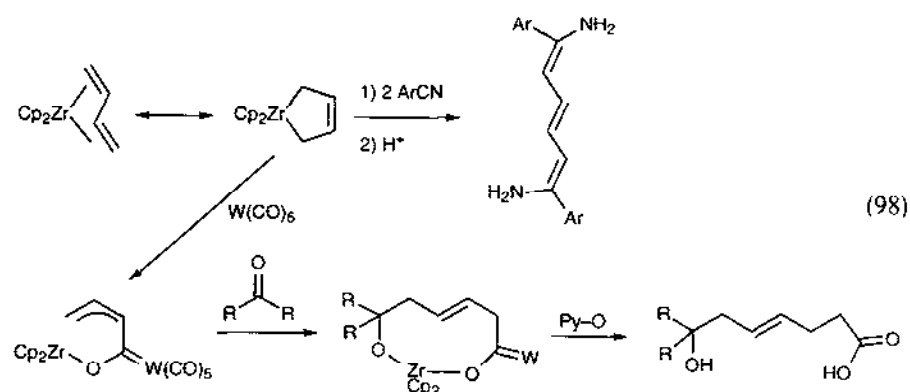
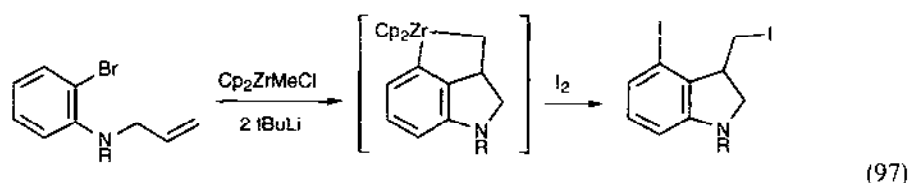
Reduced zirconocenes ring opened vinyl cyclopropanes to give η^1 alkyl- η^3 -allyl species (see Eq. (93)) which underwent further reaction with electrophiles [336,337]. Diallyl amines underwent zirconocene dichloride-catalyzed cyclization with butyl Grignard reagents (Eq. (94)) [338], as well as cyclocoupling/carbonylation in the context of the synthesis of (–)-dendrobine [339] (Eq. (95)) [340].



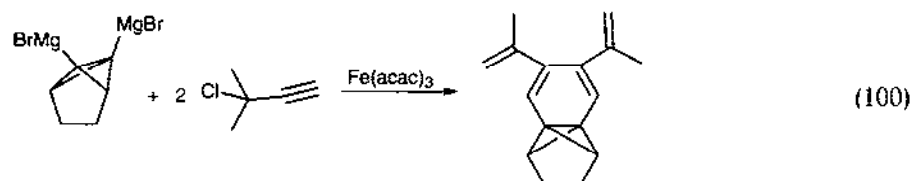
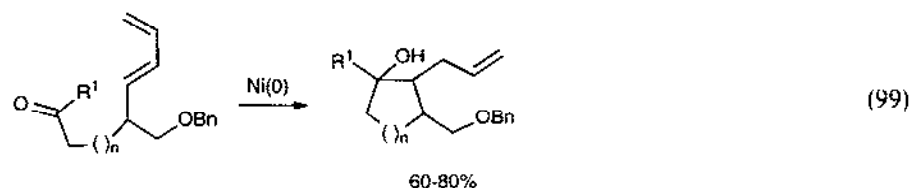
Reduced zirconocenes coupled alkynes with *N*-anions of amines to give allylic amines [341], while diethylzirconium coupled to alkynes to give zirconacyclopentenes which underwent further coupling reactions with allylic halides [342]. Zirconacyclopentanes alkylated aldehydes [343]. The full paper on the reductive cyclization of alkynes to zirconacyclopentadienes, followed by cleavage with trihalides of P, As, Sb, Bi, Si, Sn, Se and S to give the aromatic heterocycles has appeared [344]. An interesting use of zirconacyclopentadienes is presented in Eq. (96) [345].

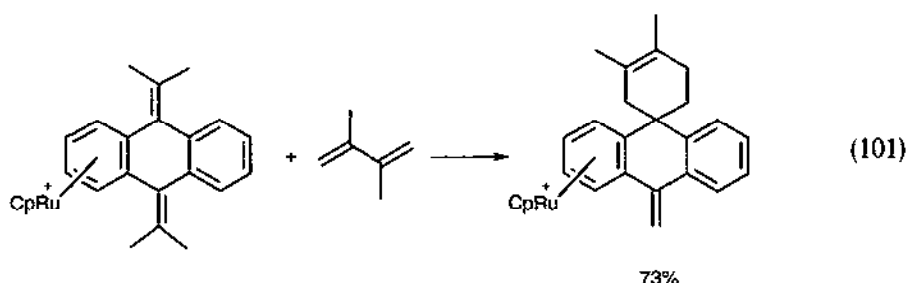


Indoles were produced by the insertions of alkenes into zirconobenzynes [346–348] (Eq. (97)) [348]. Zirconocene diene complexes coupled to nitriles to give highly unsaturated enamines (Eq. (98)) [349,350], and with ketones followed by nitriles to give dieneamine allylic alcohols [351]. Reaction with tungsten carbonyl followed by ketones resulted in homologation (Eq. (98)) [352].

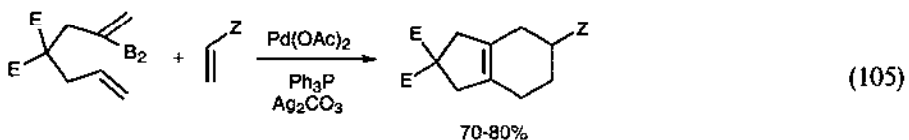
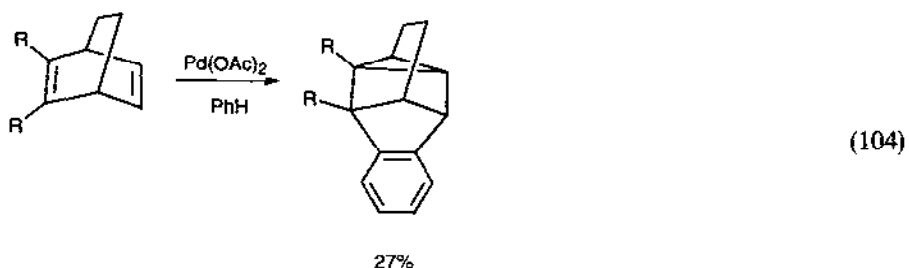
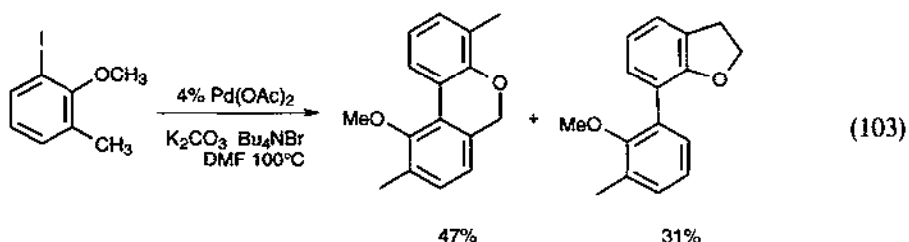
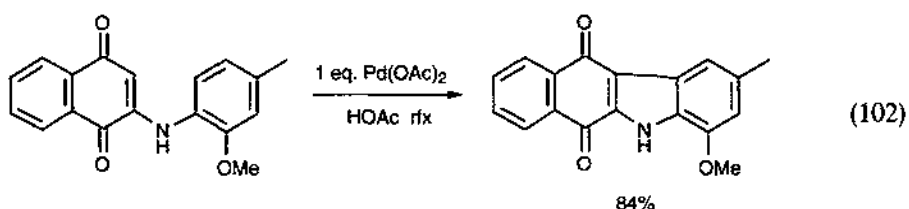


A wide range of heterocyclic aromatic halides was symmetrically coupled by nickel complex reducing agents (NiCRA) [353]. Nickel(II) phosphines catalyzed the coupling of lithium acetylides [359], while nickel(0) complexes catalyzed the cyclocoupling of 1,6-diynes with isonitriles [355], and the reductive coupling of dienes to ketones (Eq. 99) [356]. Alkyl triflates were carbonylatively coupled by Collman's reagent, $\text{Na}_2\text{Fe}(\text{CO})_4$ [357]. Two unusual couplings are presented in Eqs. (100) [358] and (101) [359].



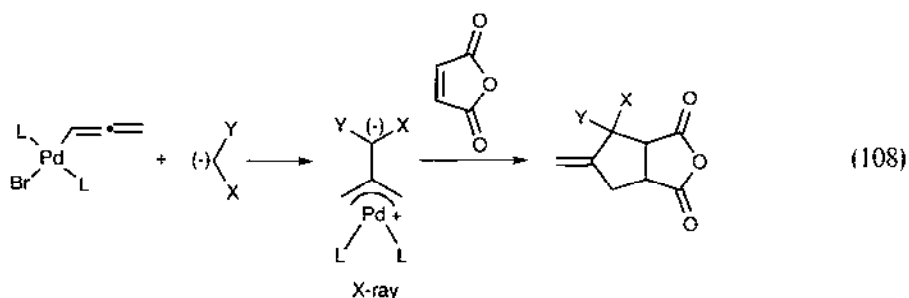
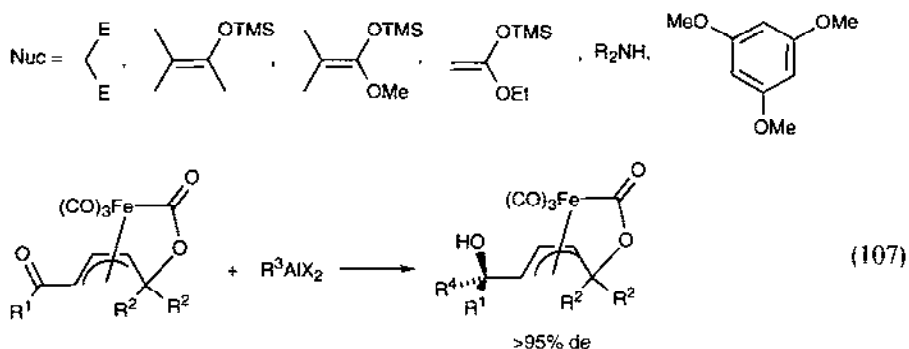
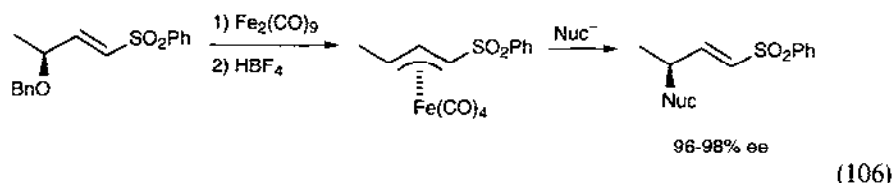


Palladium complexes catalyzed a number of coupling processes which involved formal CH activation, including the dimerization of *N*-trityl imidazoles [360] and other arene-CH activation processes (Eqs. (102) [361], (103) [362] and (104) [363]). Palladium(0) complexes cyclocoupled dienes containing a vinyl halide with electron poor alkenes (Eq. (105)) [364].



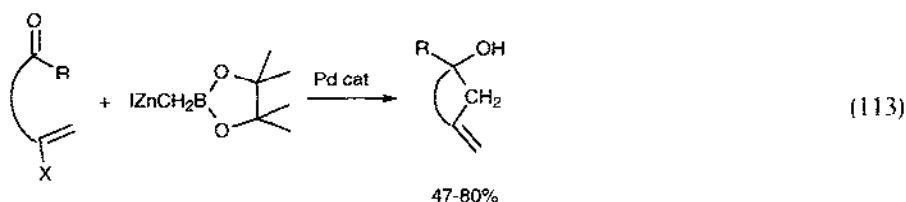
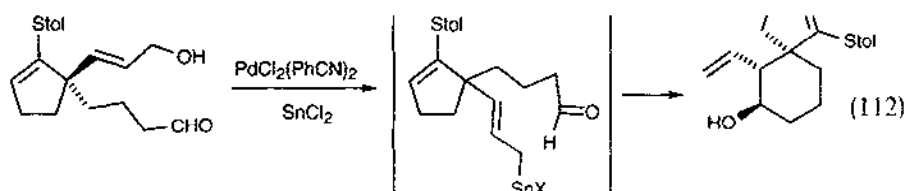
2.1.9. Alkylation of π -allyl complexes

π -Allyl-iron complexes, generated by treatment of iron carbonyl-complexed allyl ethers with Lewis acids, underwent stereo- and regio-selective alkylation by a range of anions [365] (Eq. (106)) [366,367]. π -Allyliron-nitrosyl complexes alkylated enones in a 1,4-addition process [368]. 1-Acyl- η^3 -allyl-iron complexes underwent alkylation with high diastereoselectivity (Eq. (107)) [369]. η^3 -Allylmolybdenum complexes of cyclopentenone underwent clean γ -alkylation by carbanions [370]. The regiochemistry of reductive elimination from η^1 -aryl- η^3 -allylpalladium complexes depended on the geometry of the complex, with arylation always occurring with the allyl terminus *cis* to the aryl group [371]. The formal intermediate in Pd-catalyzed 3+2 cycloadditions via trimethylene methane complexes was characterized (Eq. (108)) [372].



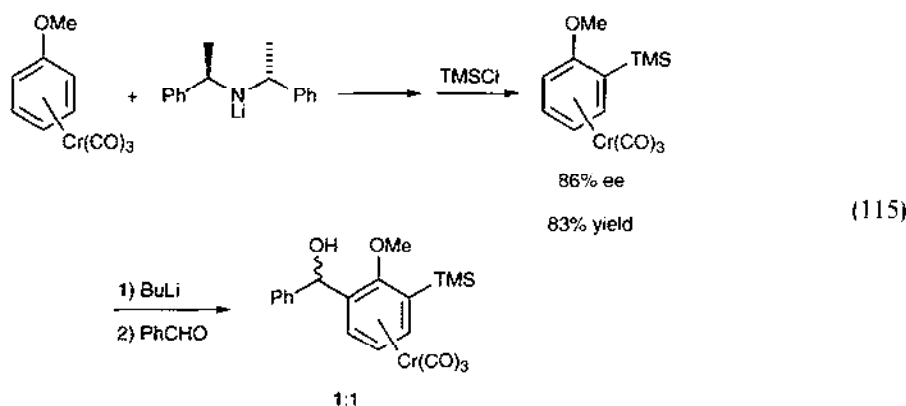
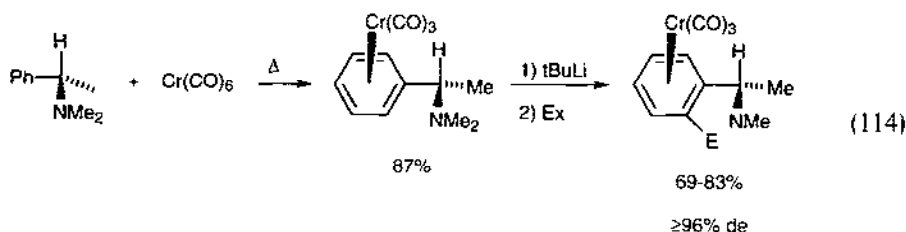
2.1.10. Alkylation of carbonyl compounds

Cp_2TiMe_2 (Petasis' Reagent) cleanly dimethylenated cyclic acid anhydrides and sulfones [373] while Tebbe's reagent (Cp_2TiCH_2) methylenated α -alkoxylactones [374]. Tebbe's reagent methylenated aryl esters, while Takai's reagent converted



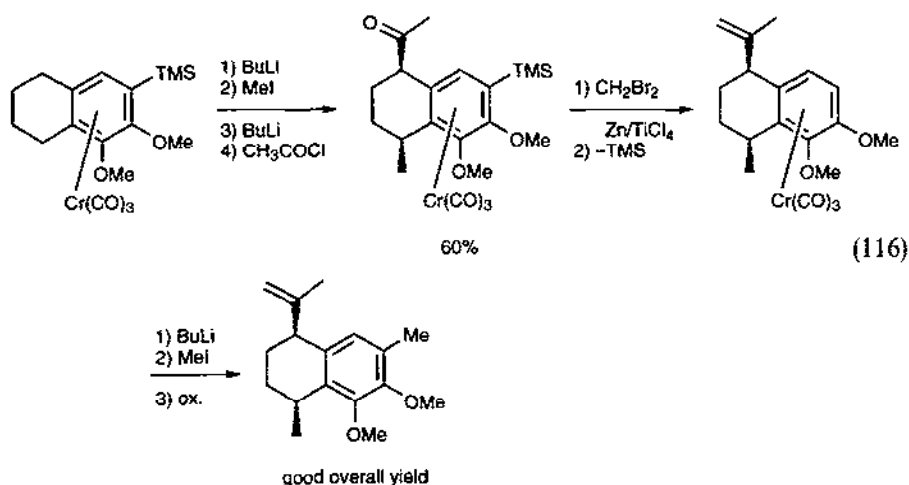
2.1.11. Alkylation of arenes

Chromium tricarbonyl complexed thiophene underwent lithiation in the two position, followed by alkylation with aldehydes [396]. Optically active α -phenethylamine underwent stereospecific complexation to the chromium tricarbonyl fragment followed by stereospecific *ortho* lithiation/alkylation (Eq. 114) [397]. Chromium tricarbonyl complexes of anisole [398] and benzaldehyde ketal [399,400] underwent stereoselective *ortho* lithiation/silylation using optically active C2 symmetric amide anions (Eq. (115)).

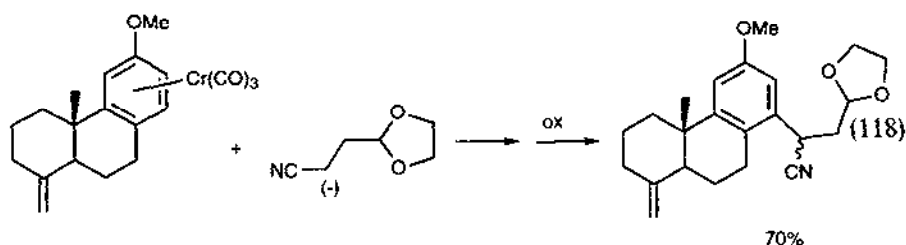
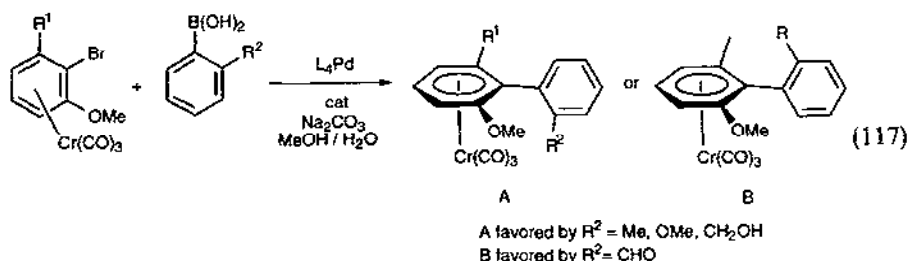


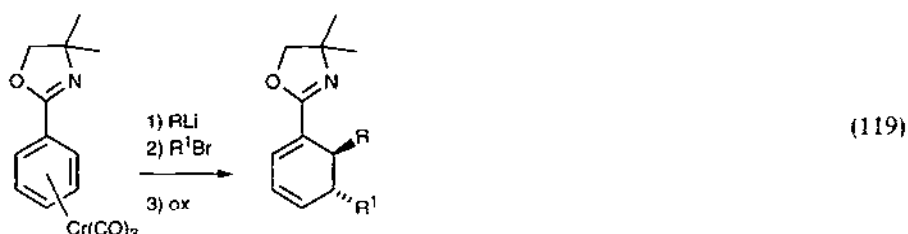
Benzylic protons of metal complexed arenes are acidic, and can be removed by base, and the resulting anions alkylated. The cationic cyclopentadienyl iron hexamethyl-

benzene underwent deprotonation with *t*-butoxide, and *hexa*-benzylation with benzyl bromide in 86% yield [401]. The relative rates of deprotonation of various benzylic positions in chromium-complexed arenes was probed [402]. This benzylic alkylation was useful in synthesis (Eq. (116)) [403].



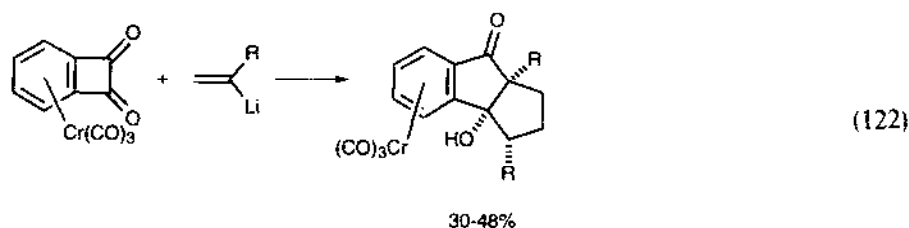
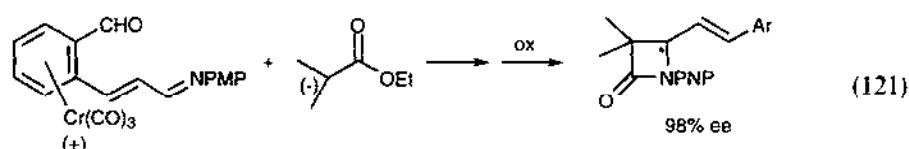
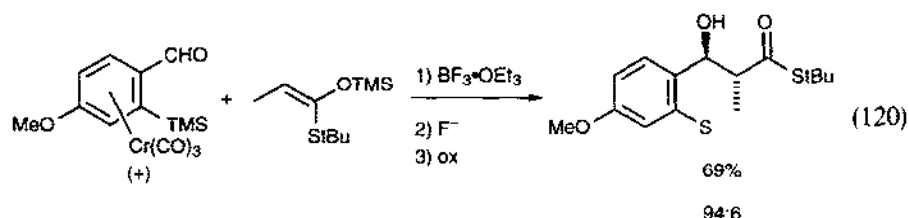
Chromium tricarbonyl-complexed *o*-chlorotoluene, *m*-chlorotoluene and *p*-chlorotoluene all underwent alkylation (displacement of Cl^- by sulfone-stabilized carbanions) to each give a mixture of *o*-, *m*- and *p*-alkylation, indicating that *ipso*-, *cine*- and *tele*-substitution was occurring [404]. Chromium tricarbonyl-complexed aryl chlorides underwent palladium-catalyzed alkylation by aryl boronates, stannanes and zinc reagents [405], and by vinyl and aryl boronates in the presence of chiral phosphines with modest ee [406]. This process was used to make biaryls (Eq. (117)) [407].



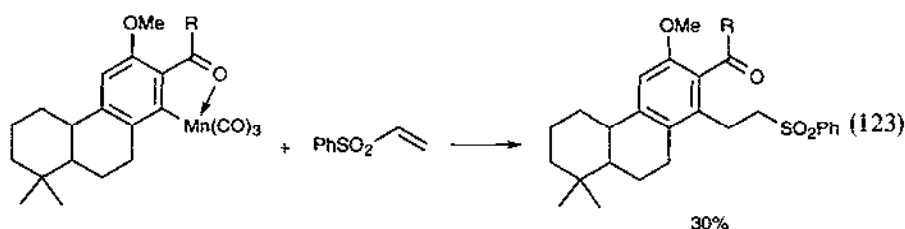


Chromium tricarbonyl-complexed triisopropylsilylbenzene underwent clean *para*-alkylation by carbanions [408]. Chromium tricarbonyl-complexed tricyclic arenes underwent clean alkylation (Eq. (118)) [409,410]. Chromium-complexed aryl oxazolines underwent bis-alkylation (Eq. (119)) [411]. Cationic benzene manganese tricarbonyl complexes were alkylated by α -carbanions of Fischer carbene complexes [412]. Rhodium(I) complexes catalyzed the *ortho*-alkylation of 2-phenylpyridine by alkenes [413]. Quinoline complexed through nitrogen to chiral cationic cyclopentadienyl rhenium species underwent α -alkylation by organolithium reagents from a single face [414].

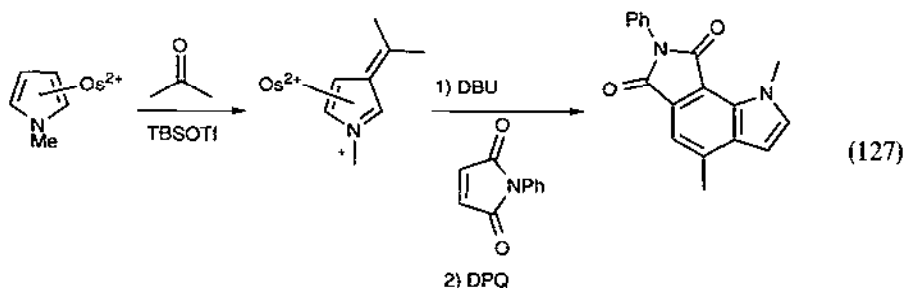
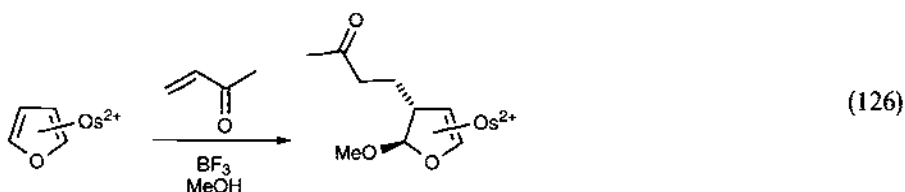
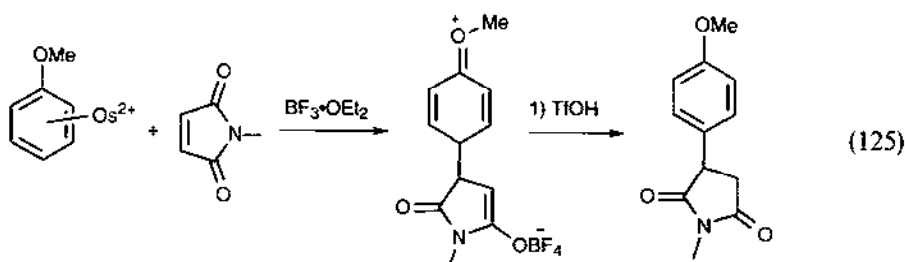
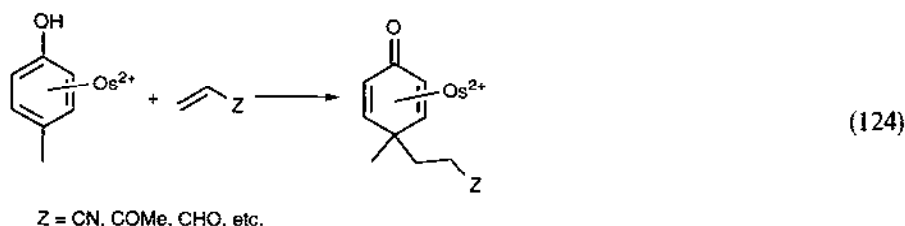
Chromium tricarbonyl complexed imines [415], aldehydes (Eq. 120) [416] and conjugated imines (Eq. 121) [417] underwent alkylation with high stereoselectivity. Benzocyclobutane dione complexes underwent the strange reaction in Eq. 122 [418].



Ortho-manganated arenes inserted alkenes [419] (Eq. (123)) [420].

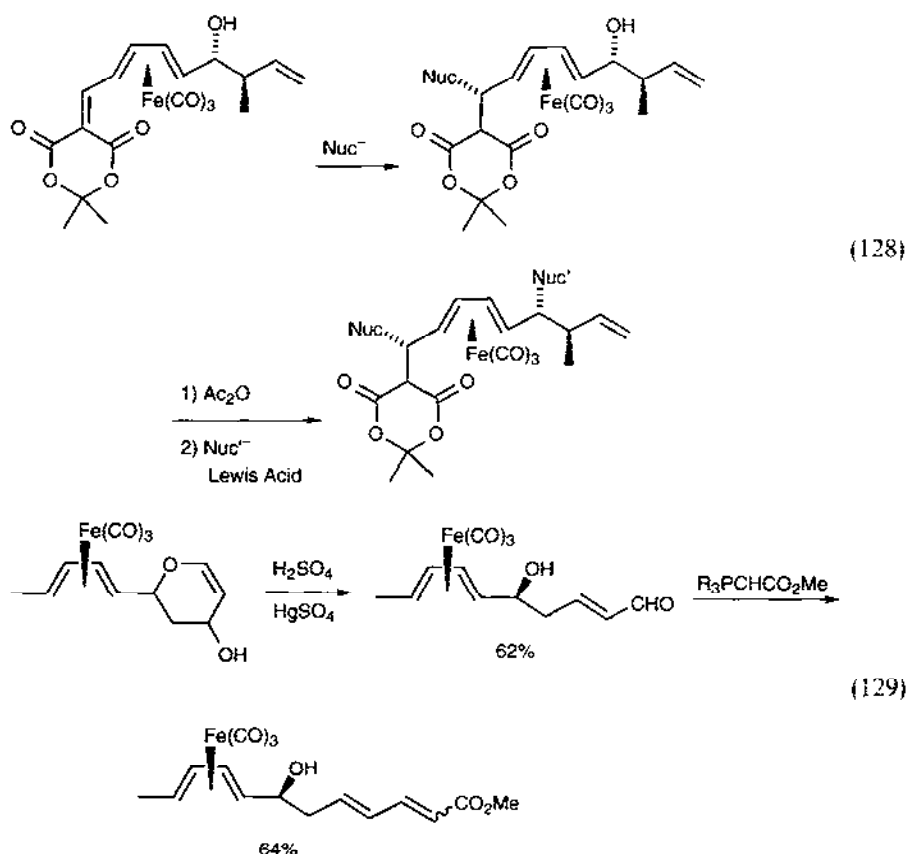


Osmium(II) complexes selectively complex to a single double bond of aromatic systems, 'deconjugating' them to allow very unusual aromatic chemistry (Eqs. (124) [421], (125) [422], (126) [423] and (127) [424]).



2.1.12. Alkylation of dienyl and diene complexes

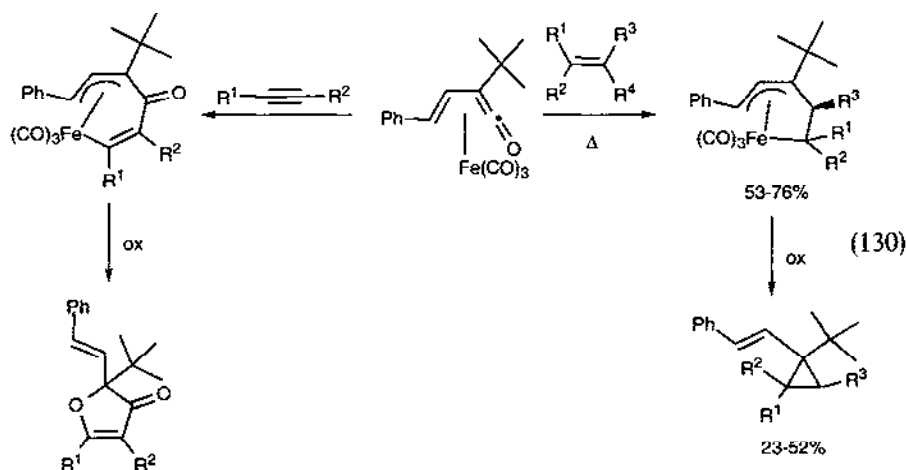
A full paper on the resolution of optically active 1,4-disubstituted 1,3-dieneiron-tricarbonyl complexes has appeared [425]. A main use for these complexes is to stabilize, by complexation of the diene, $\alpha,\beta,\gamma,\delta$ -unsaturated aldehydes and ketones to permit clean reactions at the carbonyl. Complexed polyisoprenoid ketones were alkylated by stabilized carbanions [426]. Complexed dienes containing an ester group at the 1-position and an aldehyde at the 4-position were cleanly alkylated at the aldehyde by organolithium reagents [427]. Complexed acid chlorides were alkylated by allyl silanes [428], while the complexed optically active 1,4-dialdehyde was monoalkylated by chiral crotyl boronates in high yield and ee [429]. Hydroxy and acetoxy groups α to the complexed diene underwent clean nucleophilic displacement from the face opposite the metal [430], while 1,4-addition to an α -vinyl malonate was highly stereoselective (Eq. (128)) [431]. A synthetic application is presented in Eq. (129) [432].



Iron complexed butadienes underwent Friedel–Crafts acylation at the terminal position [433], while iron complexed cycloheptatrienone underwent monoalkylation by cuprates at the 3-position [434]. Cationic cyclohexadienyliron tricarbonyl was alkyl-

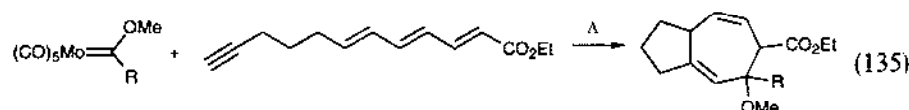
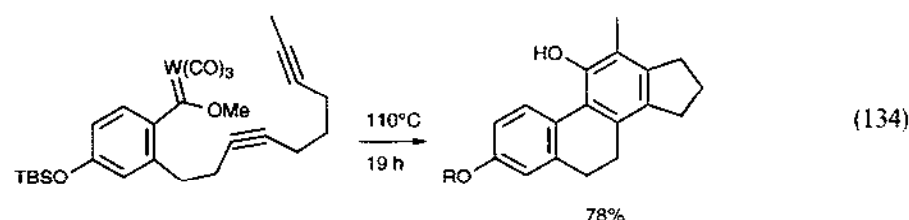
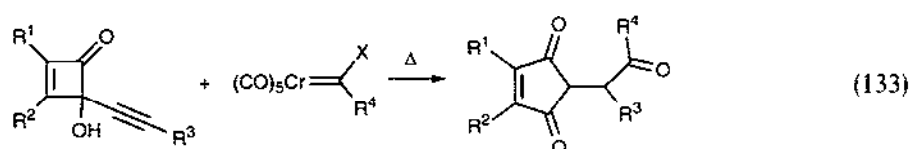
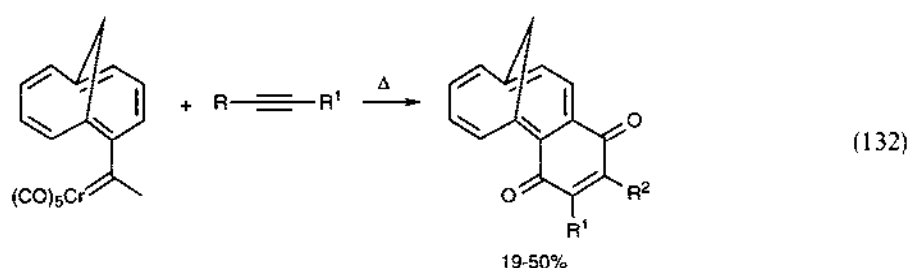
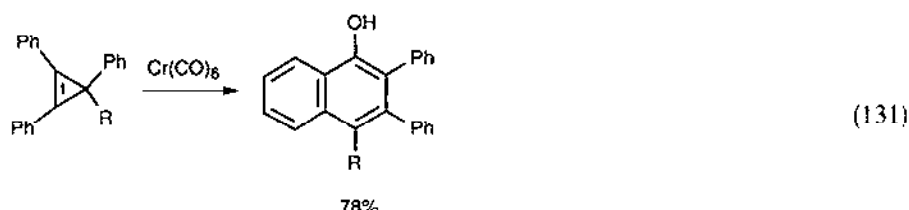
ated by ester enolates and ultimately converted to complexed dihydroindolines [435]. The alkylation of complexed cyclohexadienyl ligand by the α -carbanion of glycine ester diphenylimine went in fair to moderate yield [436]. Cyclohexadienyl-iron complexes containing a menthyl ether group were alkylated by malonic ester in poor yield but high ee [437]. The 4-cyanation of 1-methoxy-4-methyl cyclohexadienyl-iron complexes was improved by using trimethylsilyl cyanide [438]. The regiochemistry of alkylation of cationic molybdenum diene complexes depended on the solvent [439].

Iron vinylketene complexes inserted alkenes [440] and alkynes [441] (Eq. (130)). Butadiene containing a cobalt(DMG)₂ group at the 2-position was generally reactive towards Diels–Alder cycloaddition [442].

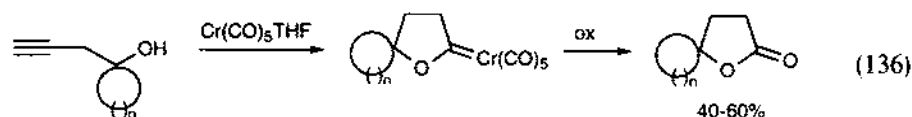


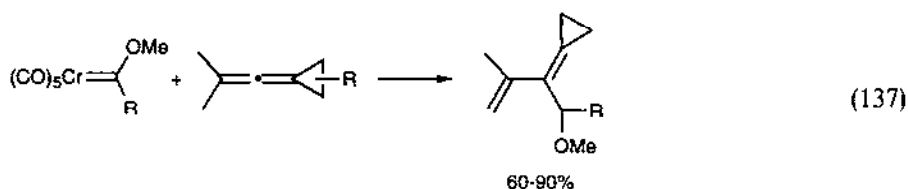
2.1.13. Metal carbene reactions

Chromium carbene complexes in organic synthesis has been reviewed (17 references) [443]. A full paper on the effects of the metal on the outcome of the Dötz reaction (phenol vs. indane) has appeared [444]. The intermediate from reactions of amino carbenes with alkynes that leads to indenenes (alkyne insertion product) has been characterized by X-ray crystallography [445]. The β -silyl vinyl carbene complex of chromium was equivalent to the ethylene carbene complex in Dötz reactions [446]. Tributylstannyl acetylene underwent the Dötz reaction with regioselectivity opposite that of other terminal alkynes [447]. The regiochemistry of the Dötz reaction was controlled by tethering the alkyne to the oxygen of the carbene with a removable silyl linker [448]. Trapping the Dötz reaction of vinyl carbene complexes and alkynes with triflic anhydride produced aryl triflates directly used in Stille couplings [449]. The Dötz reaction of vinyl carbenes with optically active propargyl ethers went with high diastereoselectivity [450]. Aryl cyclopropenes underwent reaction with chromium hexacarbonyl to give Dötz-like products (Eq. 131) [451], while cyclopropenones underwent reaction with alkoxycarbene complexes to give cyclobutenones and naphthoquinones [452]. Other Dötz-type reactions are shown in Eqs. 132 [453], 133 [454], 134 [455] and 135 [456].

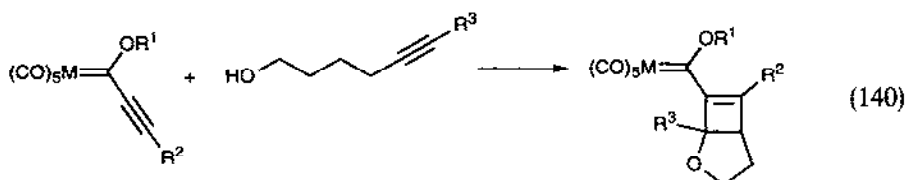
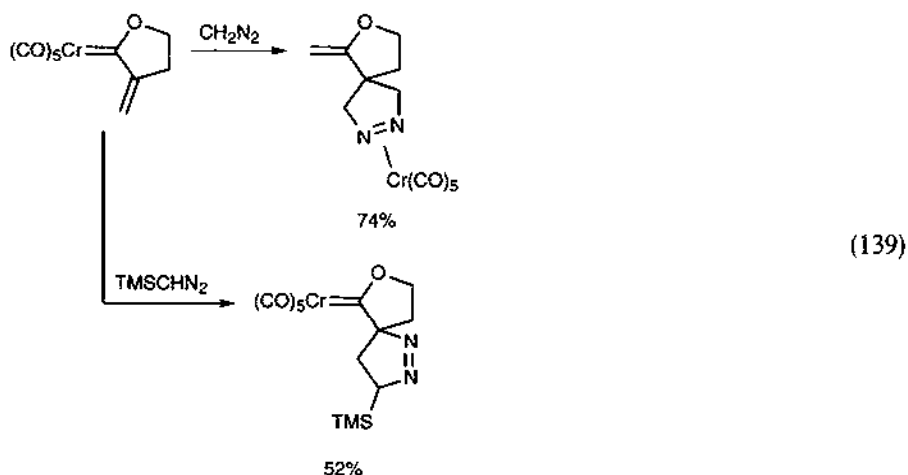
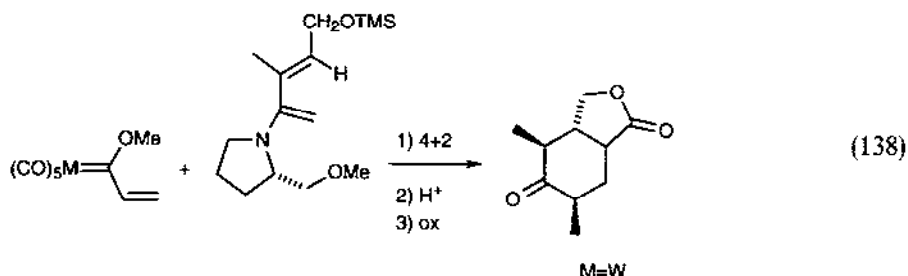


Acetylenic alcohols underwent reactions with $\text{Cr}(\text{CO})_5$ THF to form cyclic carbene complexes which were oxidized to lactones (Eq. 136) [457]. Photolysis of chromium acylate complexes with conjugated enones resulted in the Michael acylation of the enone [458]. Treatment of (aryl)(methoxy)carbene complexes with LiCH_2Cl produced aryl methyl ketones [459], while allenes produced CH insertion products (Eq. 137) [460].



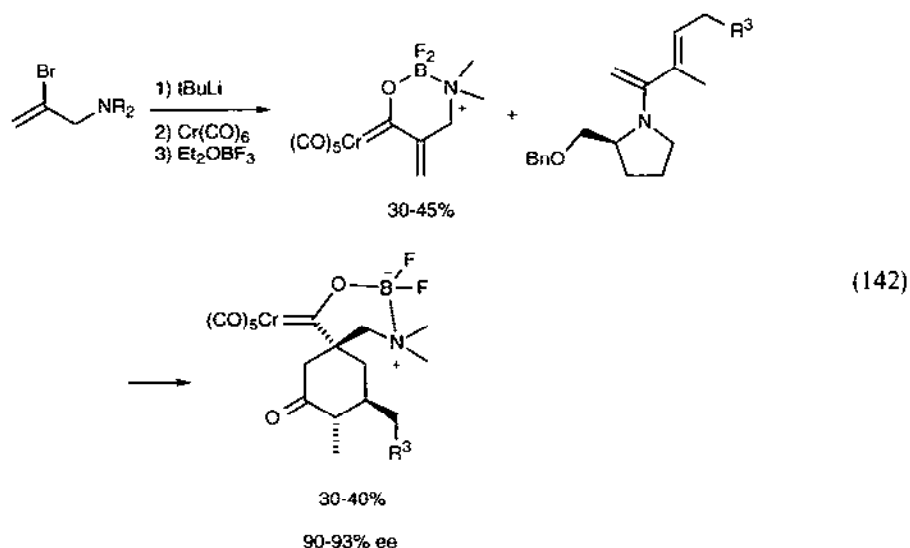
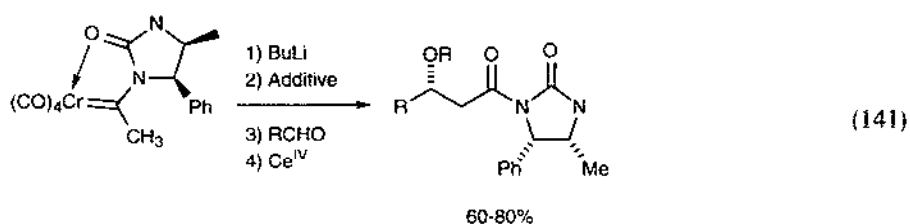


Vinylcarbene complexes of tungsten were comparable to maleic anhydride in Diels–Alder reactions [461], diastereoselectivity (Eq. 138) [462]. Diazo methane (Eq. 139) [463] and acetylenic alcohols (Eq. 140) [464] also added.



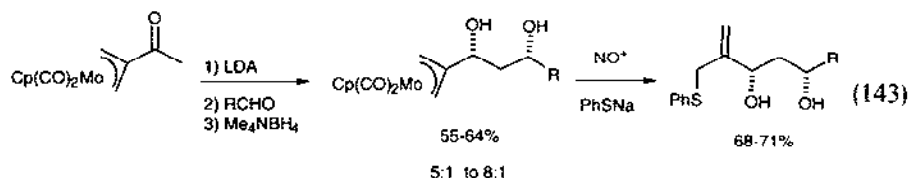
Chromium(amino)(propenyl) carbene complexes underwent deprotonation/alkylation to give mixtures of α - and γ -alkylation products [465]. Chromium carbene

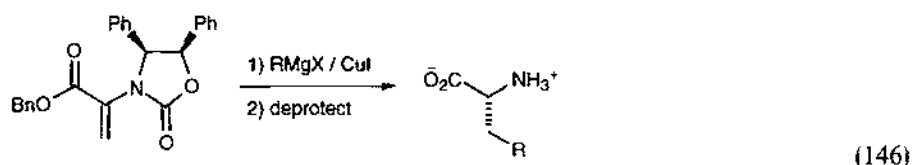
complexes with chiral imidazolidinone ligands underwent aldol chemistry with high stereoselectivity (Eq. (141)) [466]. A complex carbene reaction is shown in Eq. (142) [467].



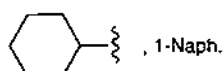
2.1.14. Alkylation of metal acyl enolates

Simple manganese(II) enolates were used for regiospecific monoalkylations [468,469]. Complexation of cyclopentenone to $\text{CpMn}(\text{CO})_2$ allowed exclusive α -alkylation on the position opposite the double bond [470]. With aldehydes, *anti*-addition was favored [471]. Enolates of 2-acetonyl- η^3 -allylmolybdenum complexes underwent *syn*-selective aldol reactions (eqn. 143) [472]. Optically active iron acyl enolates opened carbohydrate 5,6-epoxides (Eq. 144) [473].

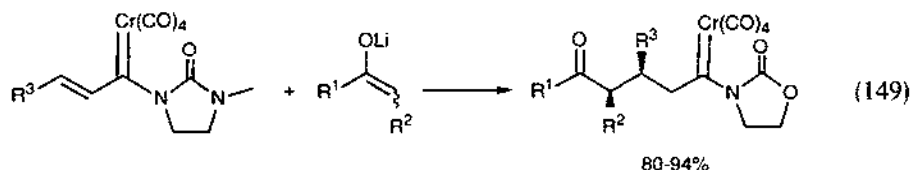
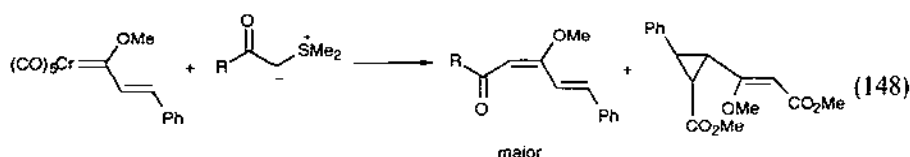
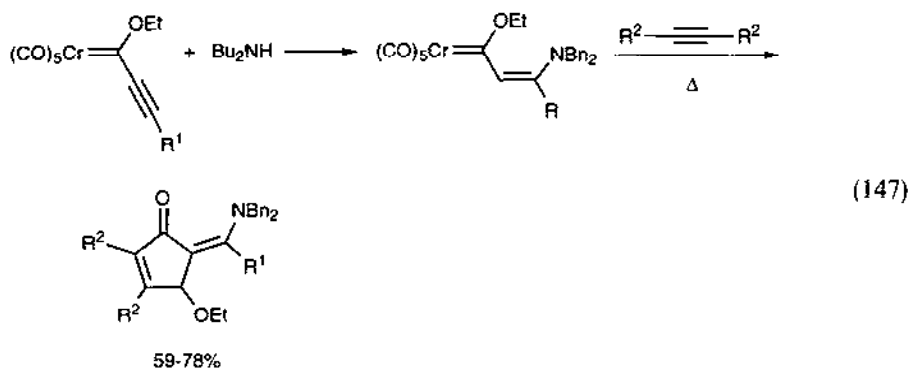


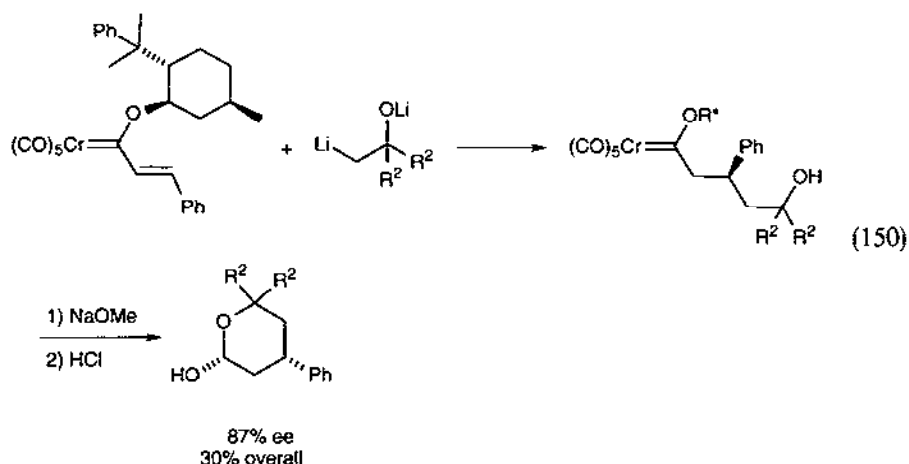


R = *t*Bu, *p*MeOPh, TMSCH₂, 1-adaman, *i*Pr,



Chromium alkynylcarbene complexes underwent conjugate amination (Eq. (147)) [481], alkoxycarbene complexes underwent insertion as well as cyclopropanation by sulfur ylides (Eq. (148)) [482] and optically active carbene complexes underwent Michael alkylation with high stereoselectivity (Eqs. (149) [483], (150) [484]).

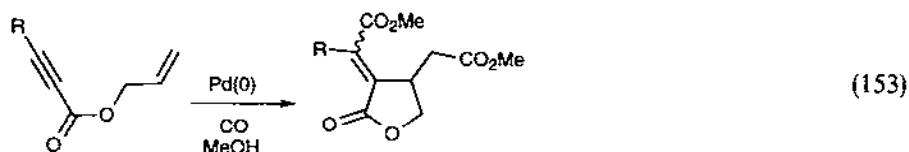
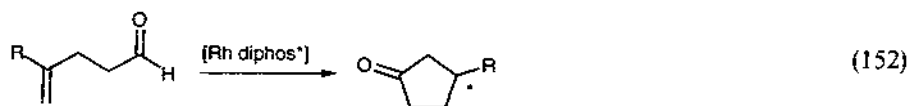
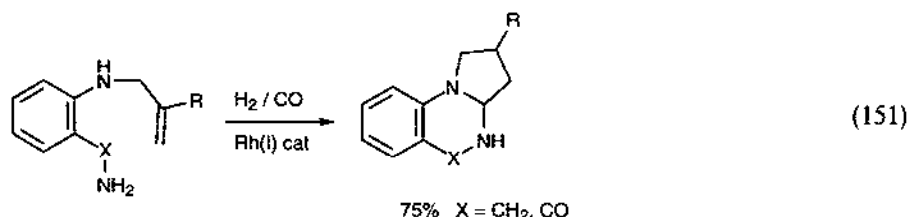




2.3. Acylation reactions excluding hydroformylation

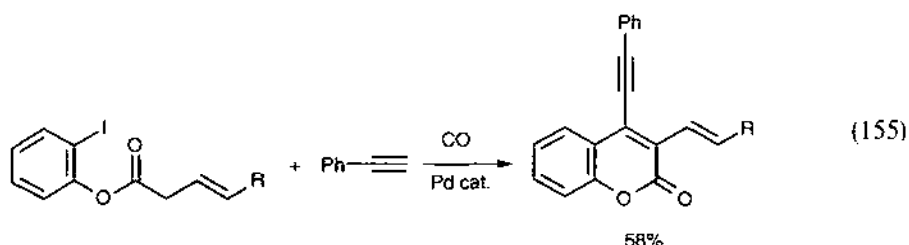
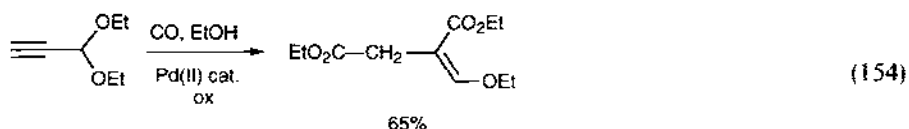
2.3.1. Carbonylation of alkenes and arenes

Ruthenium(II) [485] and palladium(II) complexes [486] catalyzed the cyclocarbonylation of allylic alcohols to unsaturated lactones. Rhodium(I) complexes catalyzed the hydroformylation of allylic amines [488,489] but unusual rearrangements were sometimes observed (Eq. (151)) [489]. Palladium catalyzed the carbonylation of allenic alcohols to produce 1,3-butadiene-2-carboxylates (Sn_2') [490], and the hydrocarboxylation of 1,3-dienes to give unsaturated acids [491]. γ -Olefinic aldehydes were cyclized to cyclopentanones with excellent ee using optically active rhodium catalysts (Eq. (152)) [492], while palladium(0) catalyzed the cyclocarbonylation of enynes (Eq. (153)) [493].

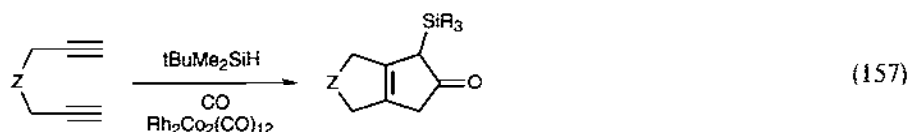


2.3.2. Carbonylation of alkynes (including the Pauson–Khand reaction)

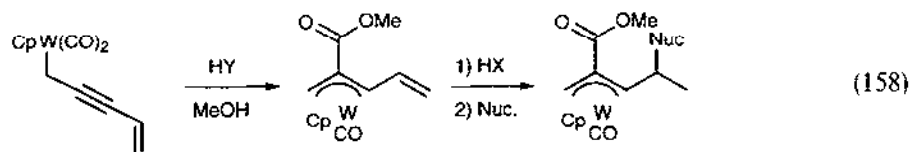
Terminal alkynes were carbonylated to propiolic esters by palladium chloride/copper chloride and carbon monoxide [494], bis carbonylated to maleic esters by palladium iodide/potassium iodide catalysts [495] and acylated to give acetylenic ketones by alkyl halides and carbon monoxide in the presence of palladium(0) catalysts [496]. Propargyl alcohols were cyclocarbonylated to α -methylene- β -lactones by palladium iodide catalysts [497], and to butenolides by phosphine palladium catalysts [498]. More complex palladium-catalyzed alkyne carbonylations are seen in Eq. (154) [499] and Eq. (155) [500].



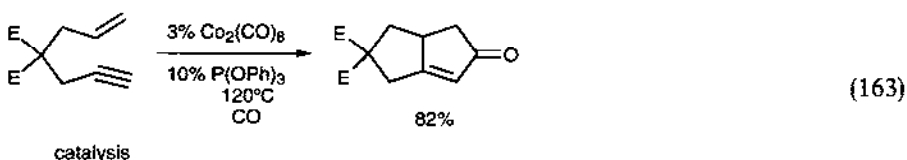
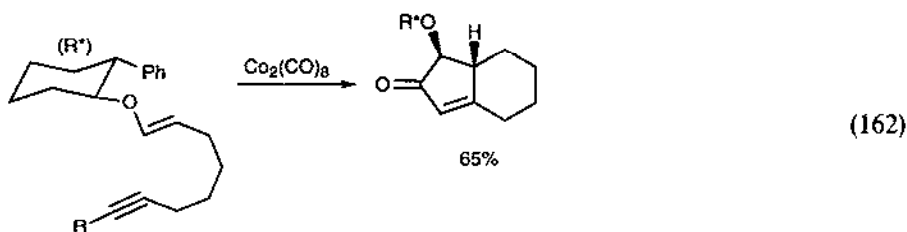
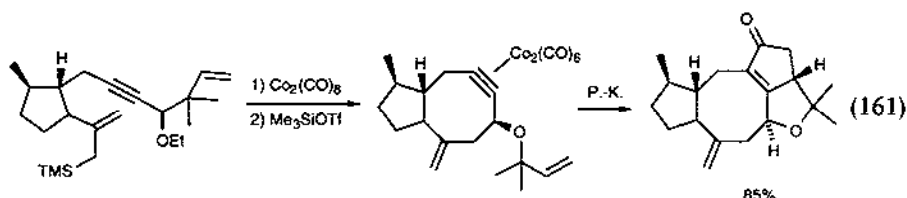
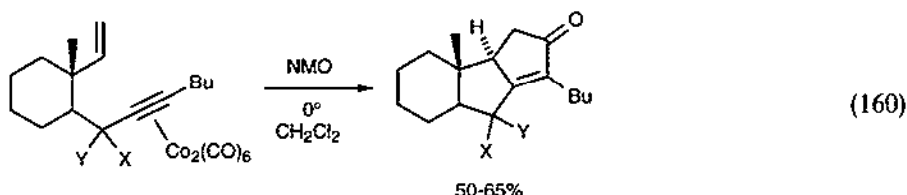
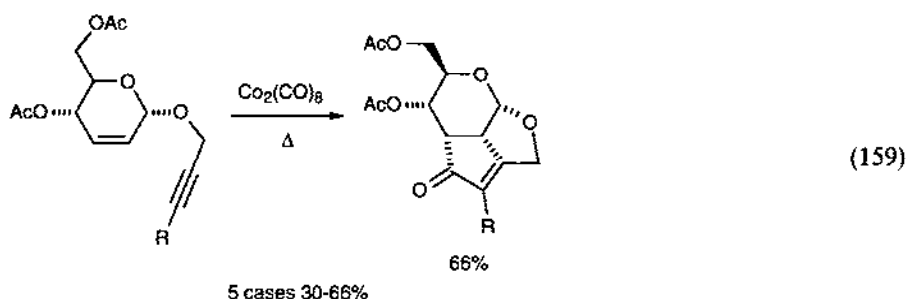
Terminal alkynes were regiospecifically silylformylated using rhodium catalysts (Eq. (156)) the formyl group always ending up in the internal position [501–503]. Diynes were cyclosilylformylated by similar catalysts (Eq. (157)) [504].

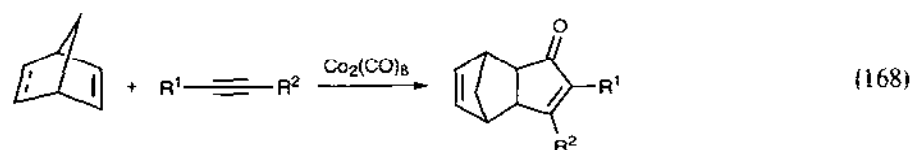
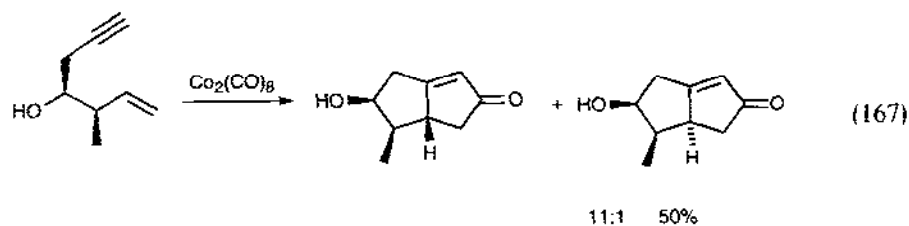
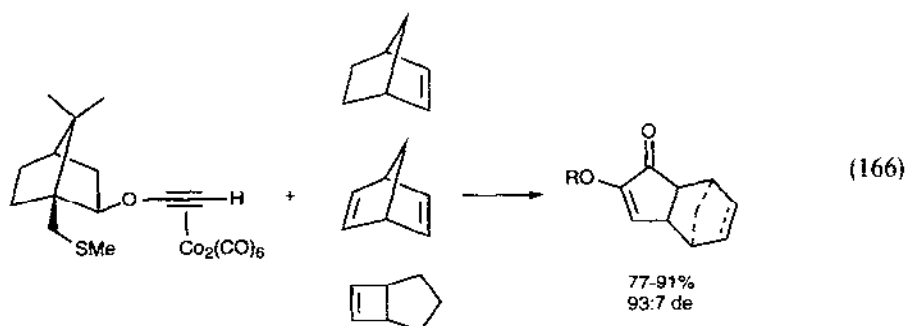
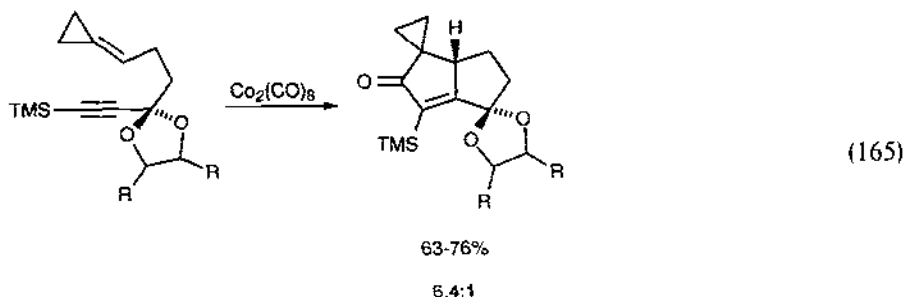
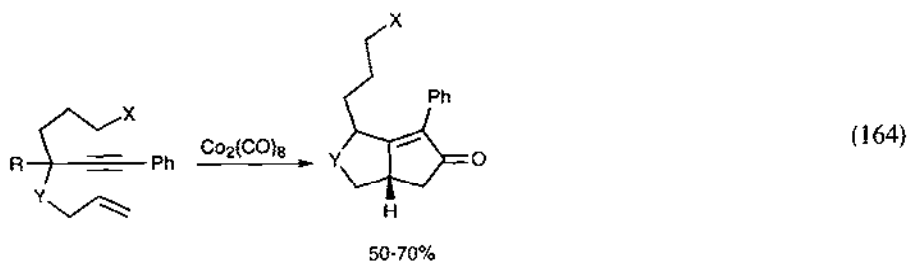


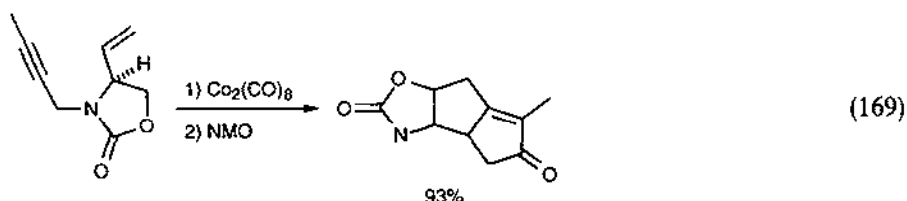
Iron carbonyl cyclocarbonylated diynes to cyclopentadienones [505] and enynes to cyclopentenones [506]. Dicobalt octacarbonyl effected similar cyclocarbonylations of enynes [507]. Treatment of a tungsten enyne complex with acid resulted in carbonylation (Eq. (158)) [508].



Alkyne- $\text{Co}_2(\text{CO})_8$ complexes, preformed by the treatment of alkynes with cobalt(II) bromide, zinc and carbon monoxide, were efficient in Pauson-Khand reactions [509]. Pauson-Khand reactions are impossible to describe in words so this years crop is presented in Eq. form (Eqs. (159) [510], (160) [511], (161) [512], (162) [513], (163) [514], (164) [515], (165) [516], (166) [517], (167) [518], (168) [519,520] and (169) [521]).

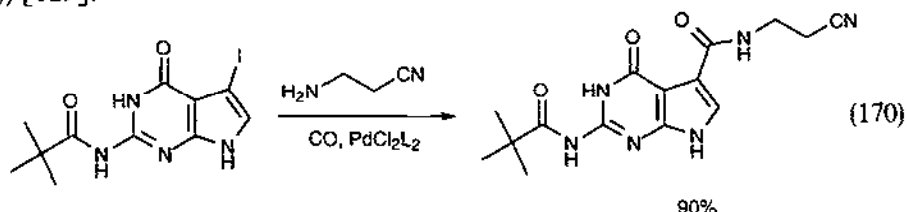




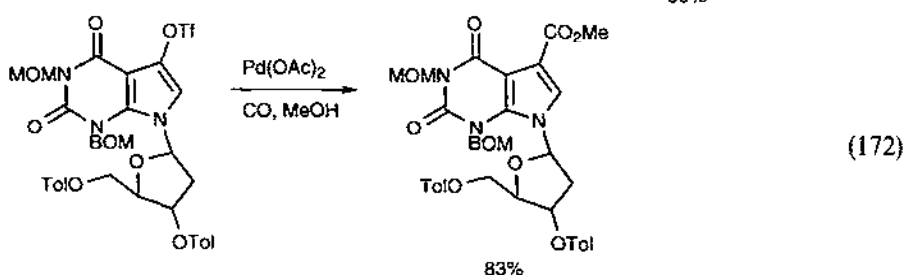
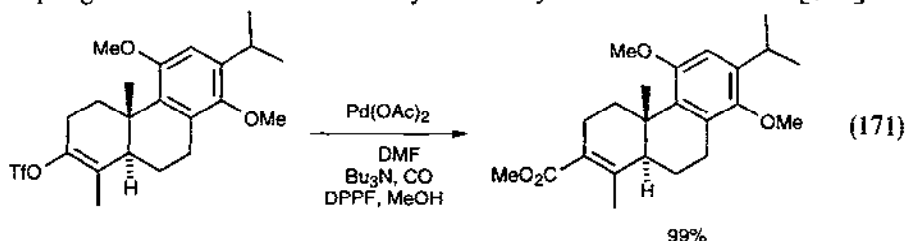


2.3.3. Carbonylation of halides and triflates

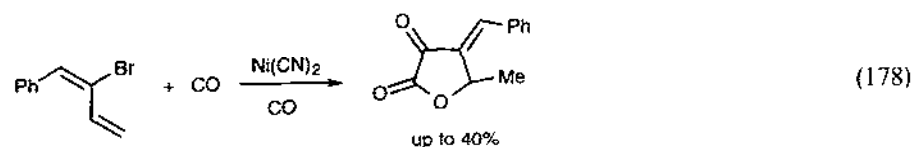
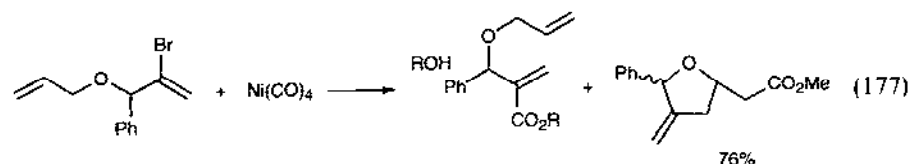
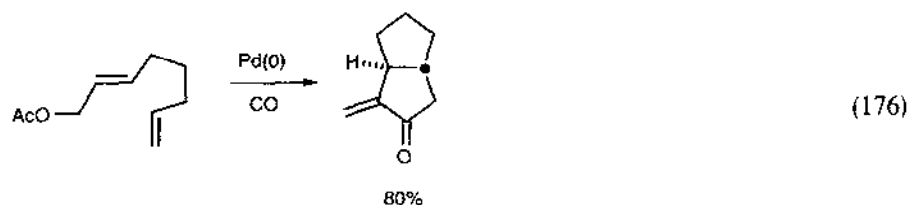
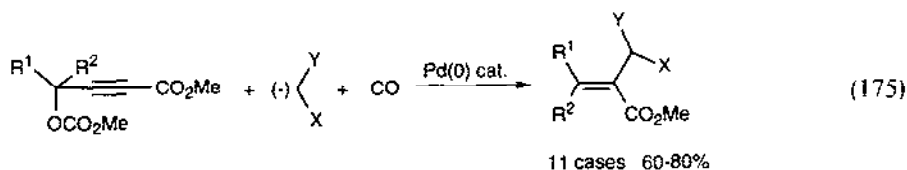
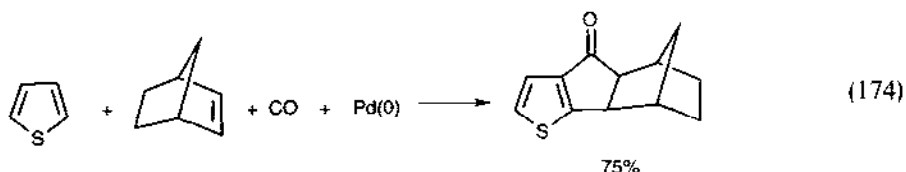
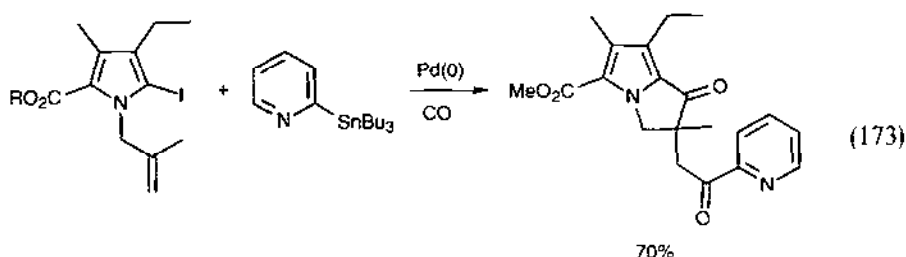
Copper(I) halides and iron carbonyl were cocatalysts for the palladium-catalyzed carbonylation of iodobenzene [522]. Palladium(0) complexes catalyzed the formylation of aryl bromides by formate [523], the carbonylation of calix[4]-aryl iodides [524], the carbonylation of chromium tricarbonyl-bound chlorobenzenes [525], and the selective para formylation of 2,4-diiodoanisole (CO , Bu_3SnH) [526]. Biphenyl aryl esters were synthesized by the palladium(0)-catalyzed carbonylation of 4-bromobiphenyl in the presence of phenols, even sterically hindered 2,6-ditertbutylphenol [527,528]. Heteroaryl iodides could also be carbonylated (Eq. (170)) [529].



Palladium catalyzed the carbonylation of (enol)vinyl triflates of β -dicarbonyl compounds [530], C-17 enol triflates of A-ring aromatic steroids [531], as well as A-ring fluorosulfonates (7 kg scale) [532], aryl triflates of dihydro- γ -pyrones [533] and benzodiazepines [534]. This type of carbonylation was efficient in complex systems (Eq. (171)) [535] and (Eq. (172)) [536,537]. Palladium(0) catalyzed the carbonylative coupling of indole-2-borates with aryl- and vinyl-halides and triflates [538].



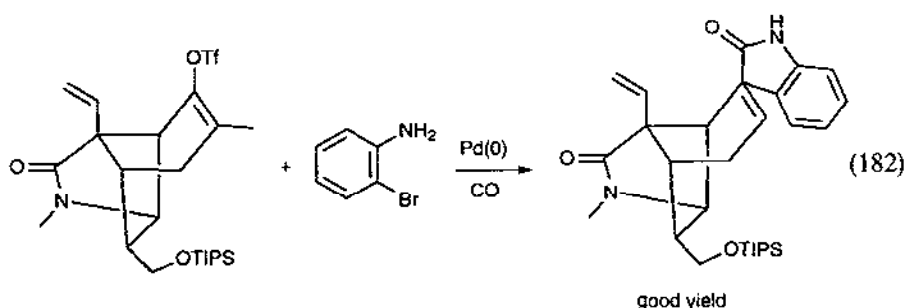
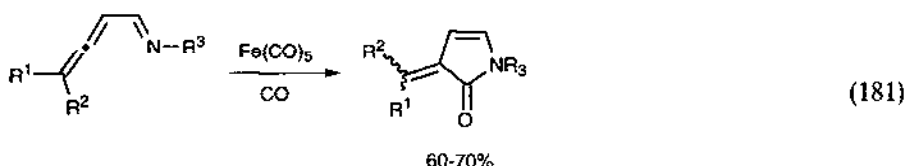
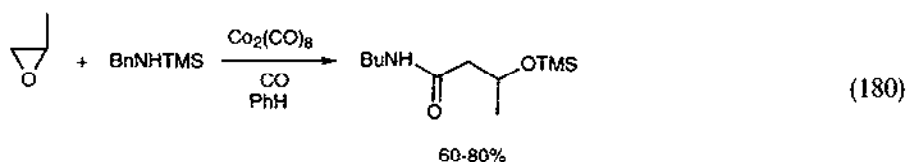
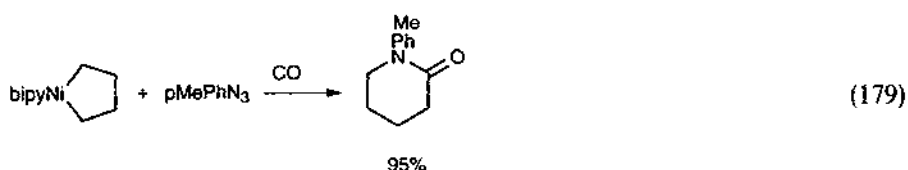
Palladium(0) complexes catalyzed the conversion of β -iodo allylic alcohols into butenolides [539], *o*-bromobenzyl alcohols with butyrolactones [540] and the cyclocarbonylation of allylic phosphates with imines to give α -vinyl- β -lactams [541]. 'Cascade' carbonylations are seen in Eqs. (173) [542], (174) [543], (175) [544] and (176) [545]. Nickel carbonyl also catalyzed cyclocarbonylations (Eq. (177)) [546] and (Eq. (178)) [547].



η^3 -Allyliron carbonyl complexes underwent facile carbonylation when oxidized [548], or treated with organic halides [549]. Treatment of iron pentacarbonyl with KOH followed by tributylphosphine produced $K_2Fe(CO)_4$, a nonpyrophoric Collman's reagent [550].

2.3.4. Carbonylation of nitrogen compounds

Aryl nitro compounds were reductively carbonylated to anilides using palladium catalysts and carbon monoxide [551]. Copper(II) acetate catalyzed the carbonylation of amines to produce carbamates under electrochemical reduction [552]. *o*-Allylcarbamates were converted to allyl amides by palladium(0) catalysts [553]. Nickel(II) alkyls underwent reaction with aryl azides and carbon monoxide to give amides (Eq. (179)) [594]. Dicobalt octacarbonyl catalyzed the carbonylation of epoxides (Eq. (180)) [555]. Iron carbonyl cyclocarbonylated 1-azadienes (Eq. (181)) [556]. A useful cyclocarbonylation is shown in Eq. (182) [557].

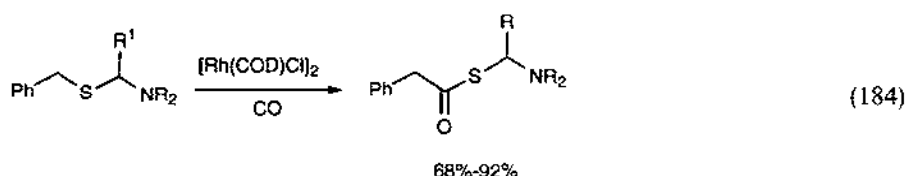
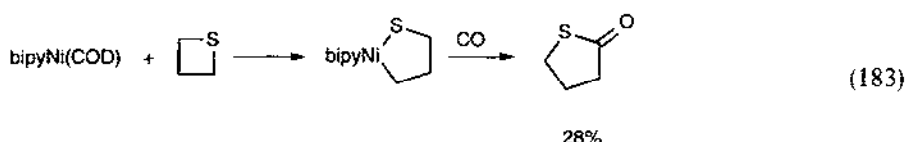


2.3.5. Carbonylation of oxygen compounds

There are no examples of such reactions in the literature.

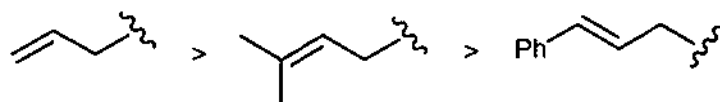
2.3.6. Miscellaneous carbonylations

There were two strange ones: Eqs. (183) [558] and Eq. (184) [559].

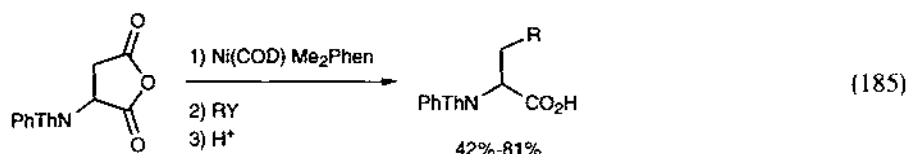


2.3.7. Decarbonylation reactions

Allyl propiolates were decarboxylated to give terminal alkynes using palladium(0) catalysts [560]. The catalyst system $\text{Pd(OAc)}_2/\text{TPPTS}/\text{Et}_2\text{NH}/\text{MeCN}/\text{H}_2\text{O}$ allowed the selective removal of allyl ester protecting groups in the order [561].



Succinic anhydrides were decarbonylated by nickel(0) complexes (Eq. (185)) [562].



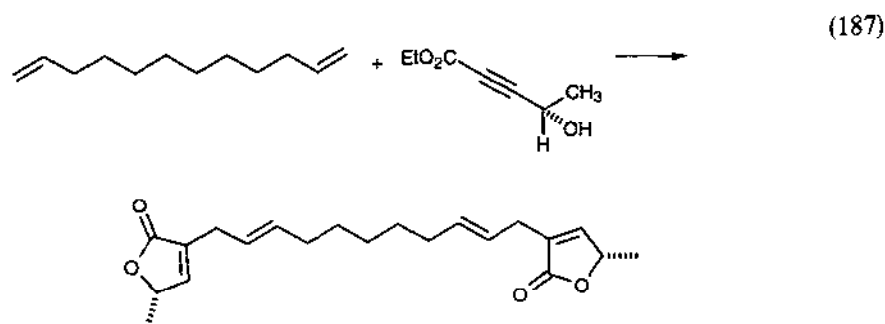
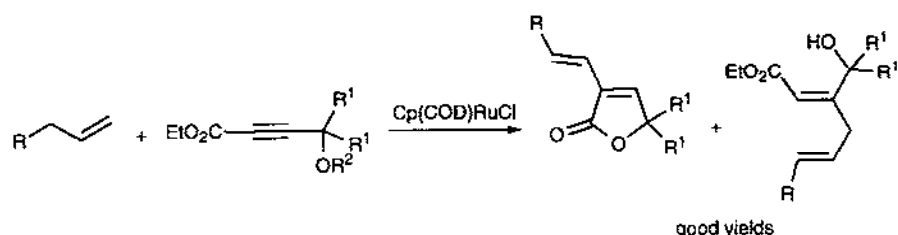
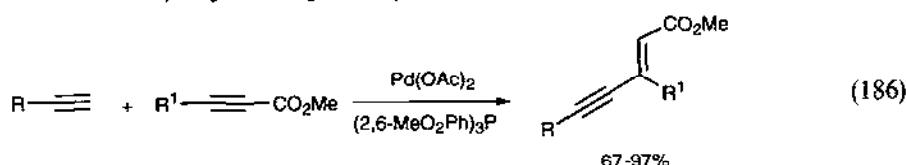
2.3.8. Reactions of carbon dioxide

Aromatic halides were carboxylated by treatment with one equivalent of nickel(0) bis-cyclooctadiene, carbon dioxide and two equivalents of bipyridine [563].

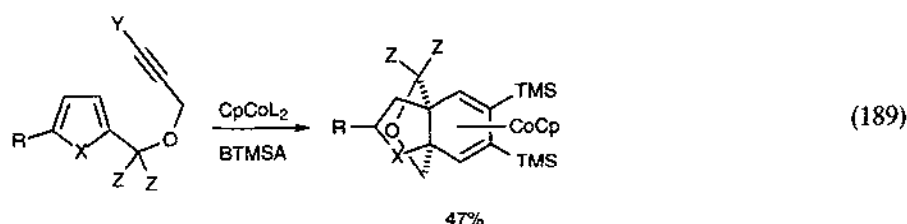
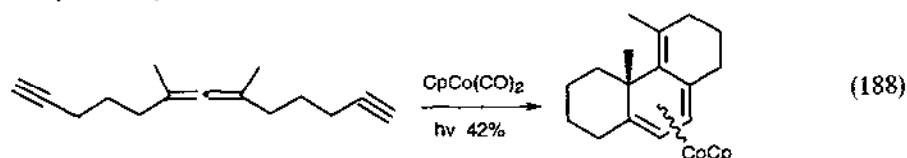
2.4. Oligomerization (including cyclotrimerization and metathesis polymerization)

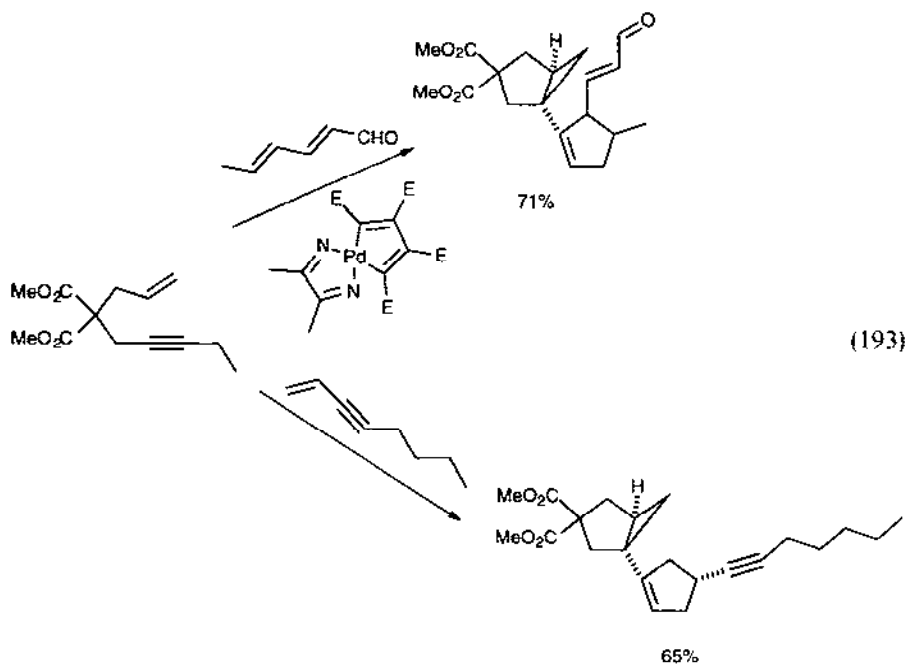
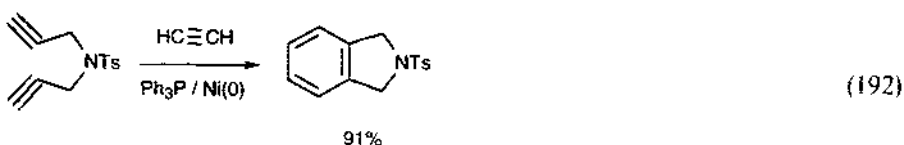
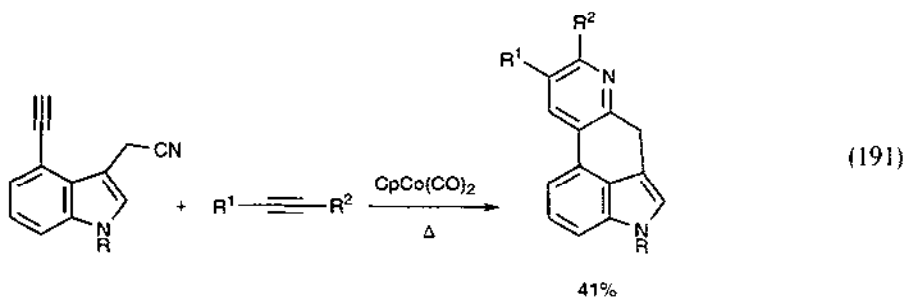
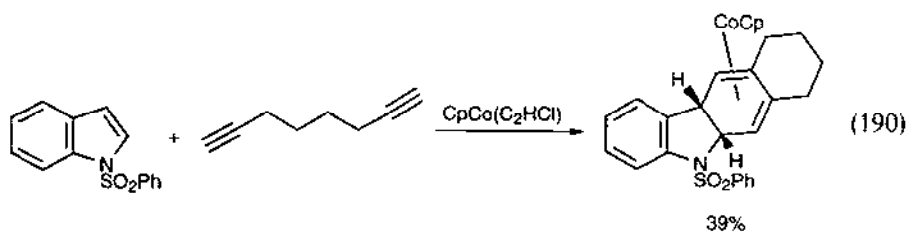
The full mechanistic details of the rhodium(III)-catalyzed tail-to-tail dimerization of methyl acrylate have been published [564]. Arene ruthenium (cyclooctadiene) catalyzed the head-to-tail dimerization of acrolein [565]. Rhodium(III) complexes catalyzed the codimerization of butadiene with homoallylic amines [566]. Palladium (Eq. (186)) [567] and ruthenium (Eq. (187)) [568] catalyzed the codimerization of

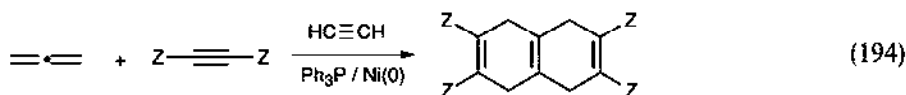
alkynes and alkene/alkynes, respectively.



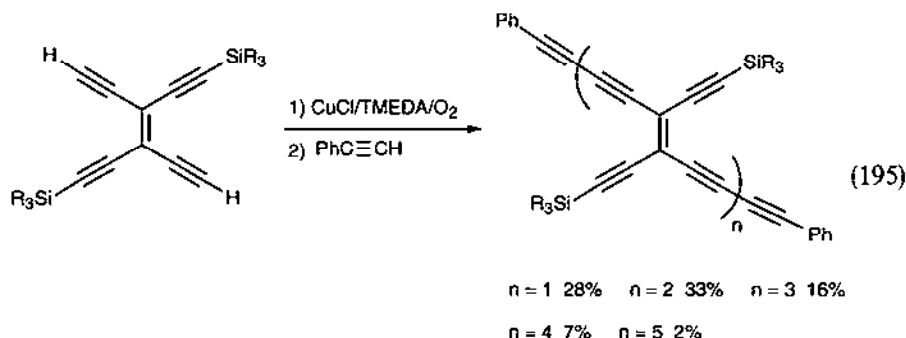
$CpCo(CO)_2$ catalyzed the cocyclotrimerization of two alkynes with an allene (Eq. (188)) [569], heterocycles (Eqs. (189) [570] and (190) [571]), and the cocyclotrimerization of two alkynes with a nitrile (Eq. (191)) [572–574]. Rhodium(II) carboxylates catalyzed the cyclotrimerization of internal alkynes to give hexasubstituted arenes [575]. Nickel (Eq. (192)) [576] and palladium (Eqs. (193) [577] and (194) [578]) also catalyzed cocyclotrimerizations.



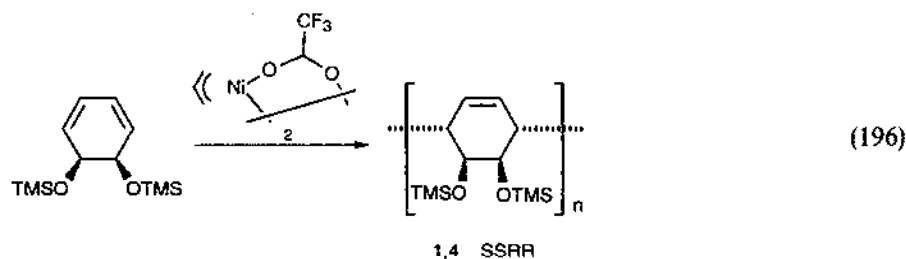




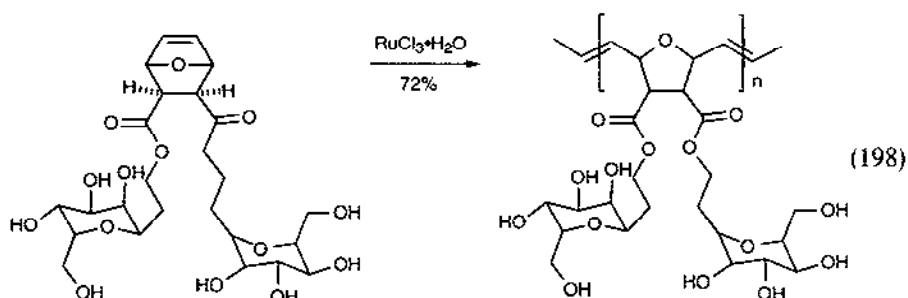
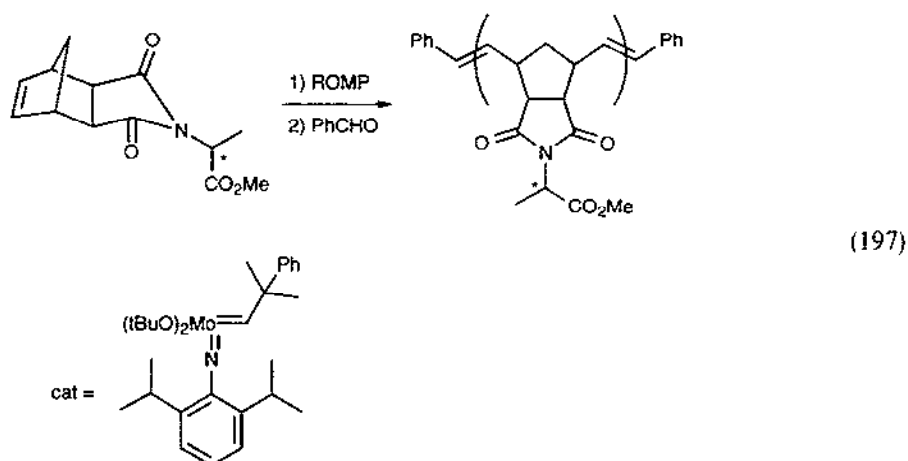
Linear block poly(phenacetylene) copolymers were prepared by the palladium/copper-catalyzed coupling of trimethylsilyl capped polyphenylalkynes with iodoaryl capped polyarylacetylenes [579]. Polyacetylenes were made by copper chloride/ O_2 coupling of diynes (Eq. (195)) [580]. Divergent, convergent and double-stage convergent syntheses of phenylacetylene dendrimers was achieved using palladium-catalyzed coupling of terminal alkynes with gem-propargyl dihalides [581]. Cyclic (poly)arylacetylenes were prepared by palladium(0)-catalyzed coupling of terminal alkynes with arylhalides [582].



'Homogeneous Group 4 Metallocene Ziegler–Natta Catalysts. The Influence of the Cyclopentadienyl-Ring Substituent' was the title of a review (297 references) [583]. Phenylacetylene was linearly oligomerized by rhodium(I) catalysts in the presence of silanes [584]. *Para*-dibromoheteroaromatics (pyridines) were oligomerized by nickel(0)-catalyzed coupling [585]. Molybdenum pentachloride oligomerized propargyl phosphonium salts to give 'polyacetylenes' with pendant phosphonium groups [586]. η^3 -Allylnickel trifluoroacetates were effective catalysts for diene oligomerization (Eq. (196)) [587]. With butadiene, living polymers with high *cis*-1,4 content and both chain ends active were produced [588].



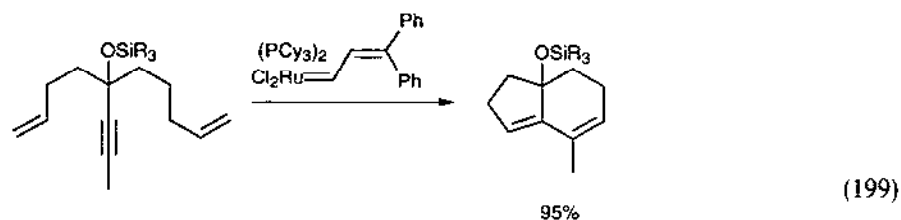
Molybdenum alkylidene complexes produced living ROMP fluorinated homo and block copolymers [589]. Two interesting ROMP processes are shown in Eqs. (197) [590] and (198) [591].



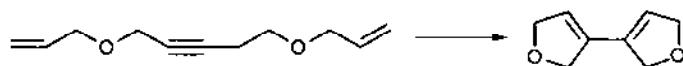
2.5. Rearrangements

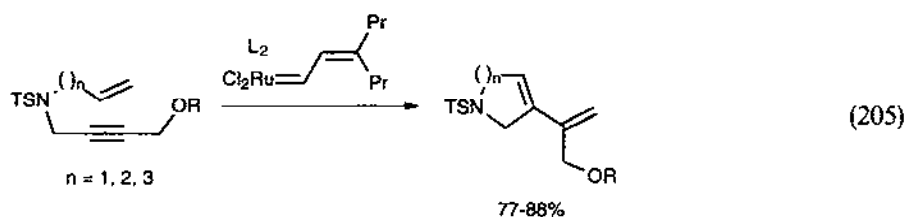
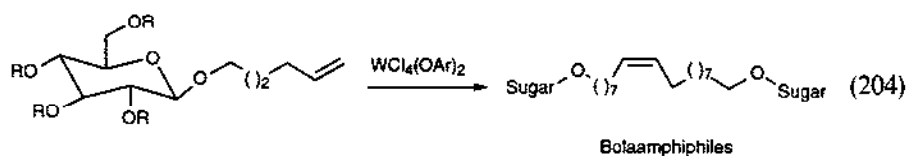
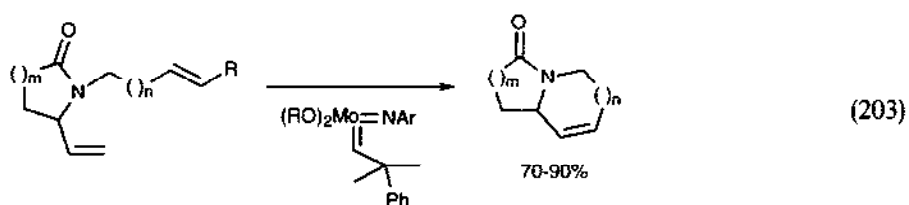
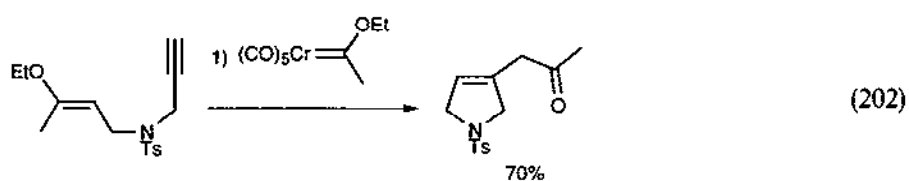
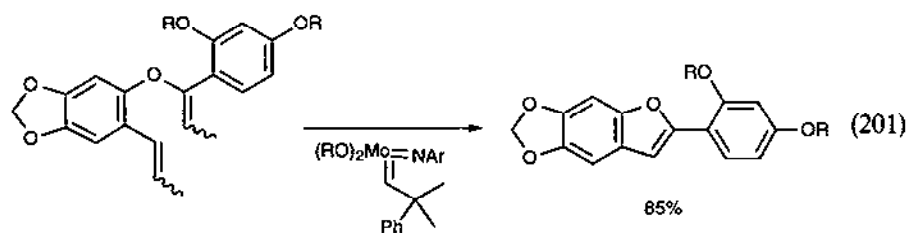
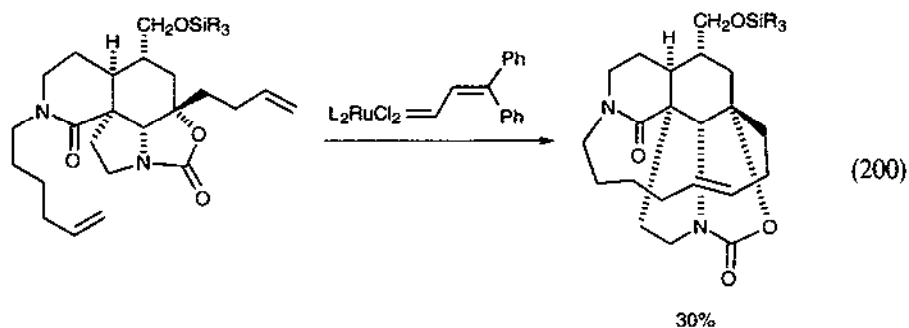
2.5.1. Metathesis

The discovery and development of metathesis catalysis which tolerate functionality has opened new ways to make ring systems. These are shown in Eqs. 199 [592], (200) [593], (201) [594], (202) [595], (203) [596], (204) [597] and (205) [598].



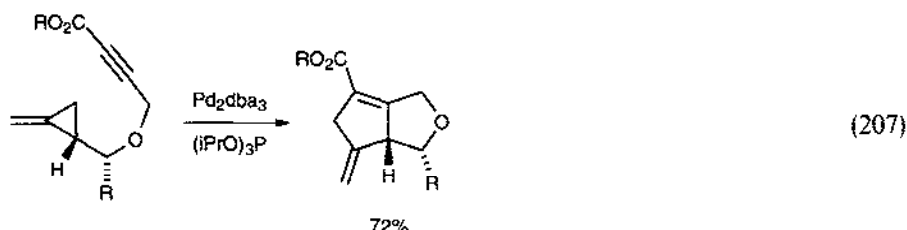
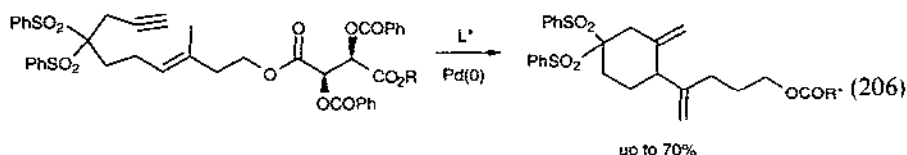
and



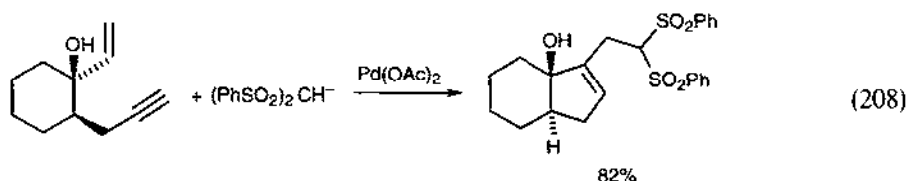


2.5.2. Olefin isomerization including cycloisomerizations

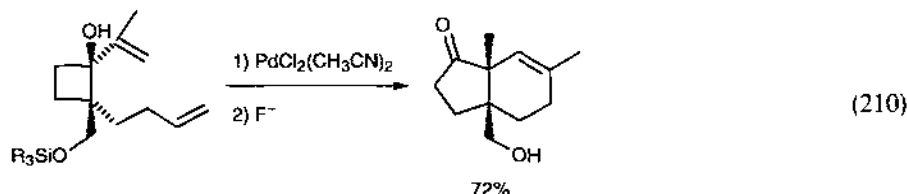
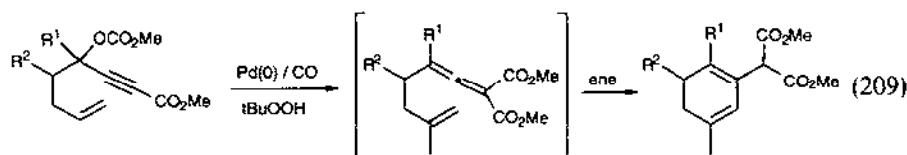
Full papers on palladium-catalyzed cycloisomerization of enynes to exocyclic 1,3-dienes have appeared [599] and [600]. Asymmetric induction has been achieved by having remote chirality present (Eq. (206)) [601]. Methylene cyclopropanes participated, with ring opening (Eq. (207)) [602], and 3,4-bis-methylene pyrroles were synthesized from allyl propargyl amines [603].

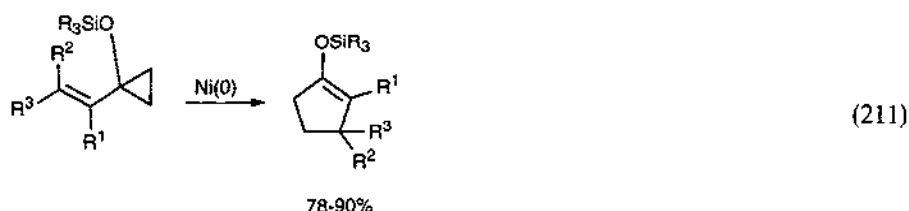


An intermediate in this cycloisomerization could be trapped by nucleophiles (Eq. (208)) [604,605]. Ruthenium(II) complexes catalyzed similar enyne cycloisomerizations [606].



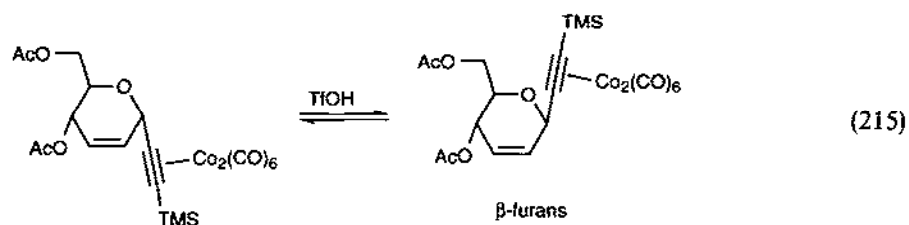
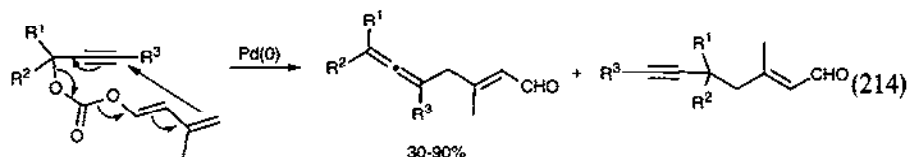
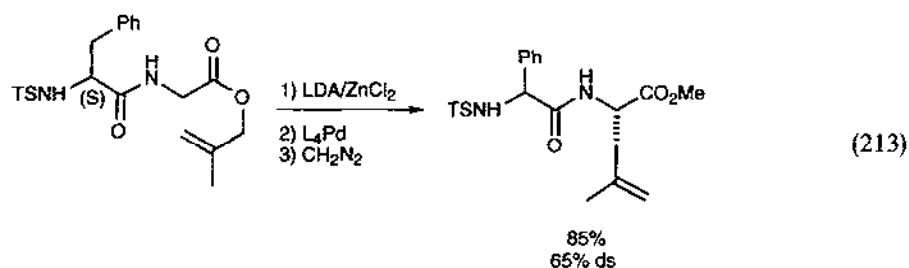
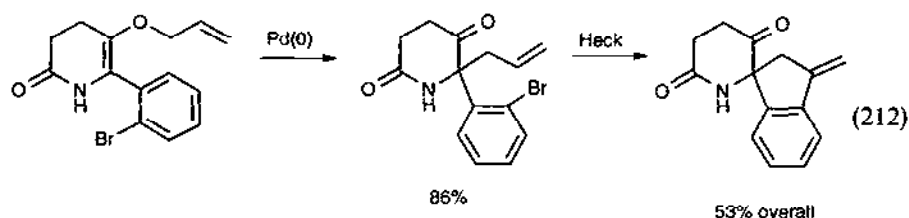
Palladium complexes also catalyzed related isomerizations (Eqs. (209) [607] and (210) [608,609]), while nickel(0) complexes catalyzed ring expansions of vinyl cyclopropanes to cyclopentenes (Eq. (211)) [610], with high ee in the presence of chiral ligands [611].





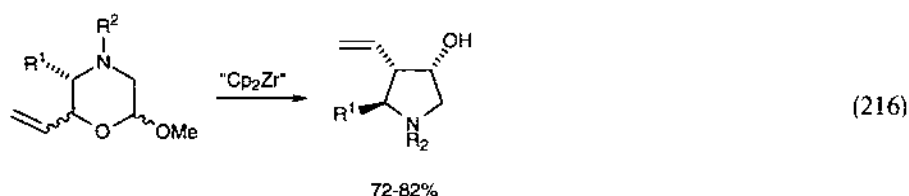
2.5.3. Rearrangements of allylic and propargylic compounds

Palladium(II) complexes catalyzed the allylic transposition of allyl acetates [612] and π -allylpalladium complexes in the presence of chiral bis-amidophosphines and *t*-butoxide deracemized cyclic allylic acetates [613]. Chiral ferrocenyl bis phosphines catalyzed η^3 to η^1 allyl isomerizations [614]. Claisen type rearrangements were catalyzed by palladium(0) complexes (Eqs. (212) [615], (213) [616] and (213) [617,618]). Cobalt-complexed propargyl sugars underwent α to β face isomerizations (Eq. (215)) [619].

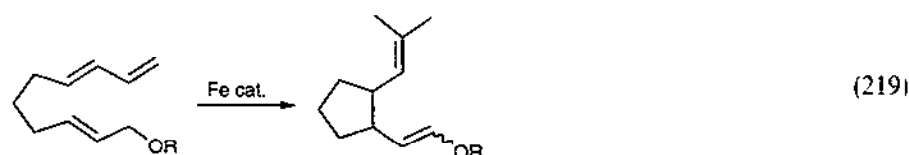
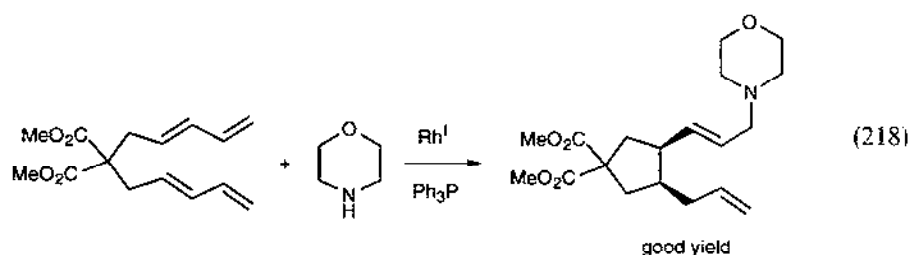
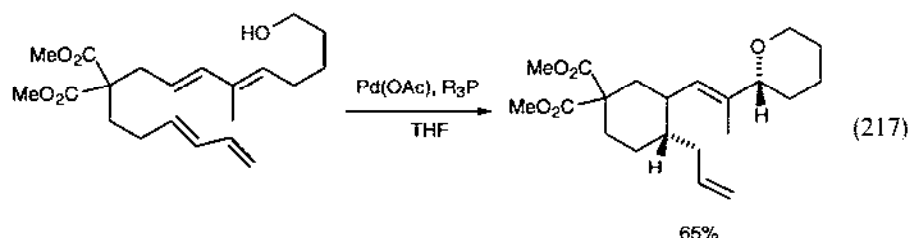


2.5.4. Skeletal rearrangements including diene telomerization

Ring contractions in carbohydrates have been reviewed (8 references) [620]. A related example is seen in Eq. (216) [621].

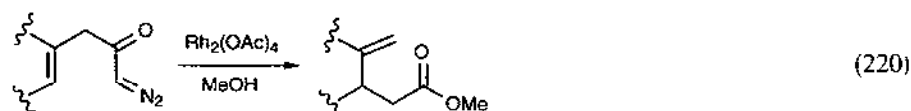


Palladium(II) (Eq. (217)) [622,623] and rhodium(I) (Eq. (218)) [624] complexes catalyzed intramolecular diene telomerizations, while reduced iron species catalyzed the diene/alkene cyclization (Eq. (219)) [624].



2.5.5. Miscellaneous rearrangements

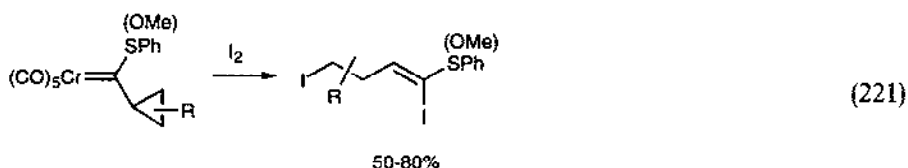
Rhodium(II) acetate-catalyzed Wolff rearrangements (Eq. (220)) [626]. Palladium(II) complexes catalyzed the rearrangements of epoxides to ketones [627].



3. Functional group preparations

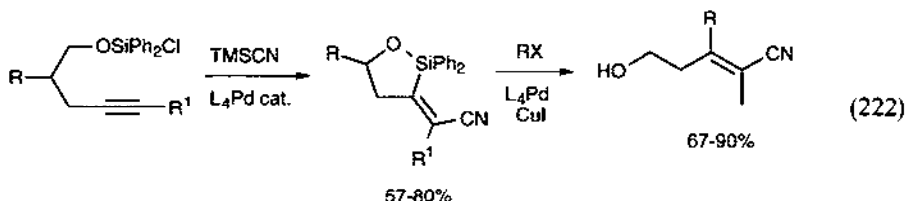
3.1. Halides

Alkenes were 1,2-dichlorinated by hexachloroethane in the presence of RuCl_2L_3 catalysts [628]. Dienes were 1,4-dichlorinated. Cyclopropyl chromium alkoxycarbene complexes were ring opened and cleaved by iodine (Eq. (221)) [629]. Fullerene underwent hydrozirconation by Cp_2ZrHCl , followed by cleavage with *N*-bromosuccinamide to give 7-bromonorbornene appendages [630].



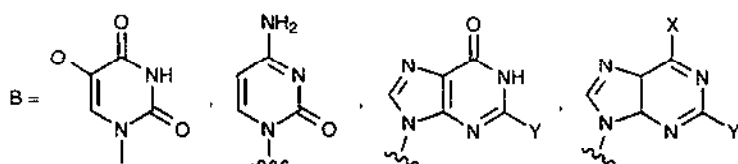
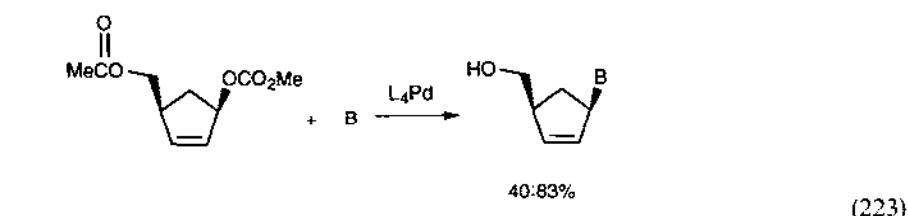
3.2. Amides, nitriles

Zirconocene dichloride and titanocene dichloride catalyzed the conversion (aminolysis) of *N*-aroyl carbamates to amides. The degree of asymmetric induction in the nickel-catalyzed hydrocyanation of styrenes could be enhanced by electronic features in the chiral ligands [632]. ^{11}C isotopically labelled aryl cyanides were prepared by the palladium(0)-catalyzed reaction of aryl halides with ^{11}C labelled cyanides [633]. Palladium(0) catalyzed the silylcyanation of alkynes (Eq. (222)) [634].

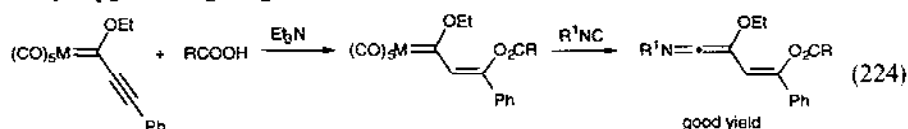


3.3. Amines, alcohols

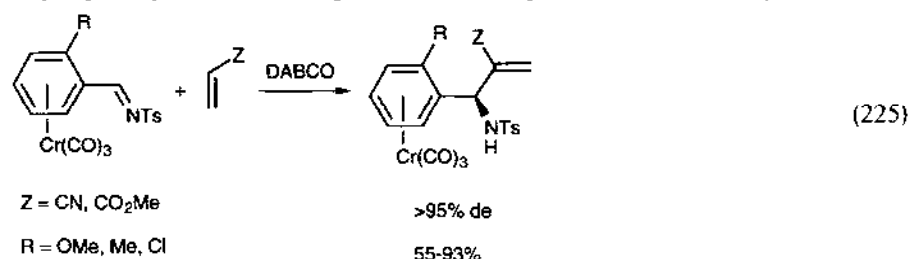
Palladium(0)/tosyl azide was an effective system for the mild removal of alloc protecting groups from amines [635]. Allyl acetates, carbonates and phosphates were asymmetrically aminated using palladium(0)-chiral oxazoline/phosphine ligands [636]. Palladium catalyzed the allylic amination of cinnamyl acetates by silylate uracil to give varying degrees of mono and dialkylation [637]. The regiochemistry of the reaction with the free base was dependent on the solvent [638]. Palladium(0) catalyzed the *N*-allylation of phosphonamides by allylic acetates [639]. Palladium(0)-catalyzed amination of allyl acetates played a major role in the synthesis of carbocyclic nucleosides (Eq. (223)) [640–643]. The bis-tosyl amine (NTs_2) was also a viable leaving group [644]. Propargyl acetates and carbonates were aminated to give propargyl amines in the presence of copper(I) chloride [645].



Palladium complexes catalyzed the amination of aryl halides by triallyl stannyl amines [646,647]. Optically active cyclopentadienyl titanium BINOL complexes catalyzed the asymmetric reduction of imines to amines with excellent enantioselectivity [648–650]. This was used for the kinetic resolution of racemic amines [651]. Chromium aminocarbene complexes were reductively cleaved to the amine by sodium borohydride in trifluoroacetic acid [652]. Isonitriles cleaved carbene complexes to give ketenimines (Eq. (224)) [653]. The cyclic ketal of *cis*-1,4-dihydroxy-2-butene was cleaved by R_2NMgX in the presence of nickel catalysis to give 1-hydrox-4-amino-2-butenes [654]. 2,5-Dihydrofurans, pyrroles and phospholes were ring opened by Cp_2ZrHCl [655].

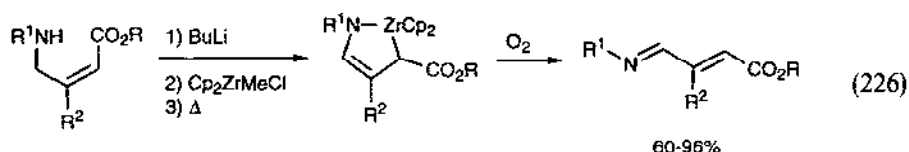


Chlorobenzene complexed to $CpFe^+$ was directly aminated by amines in the presence of *t*-butoxide and DMSO [656]. Chromium tricarbonyl complexed *N*-tosyl benzaldehyde imines underwent the Bayless–Hillman reaction with high diastereoselectivity (Eq. (225)) [657]. α -Methyl styrenes were aminated (NH_2) on the menthyl group by phenyl hydroxyl amine in the presence of iron phthalocyanine catalysis [658].

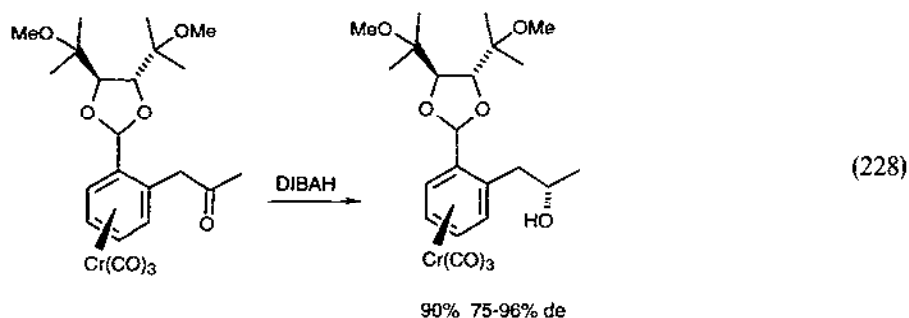
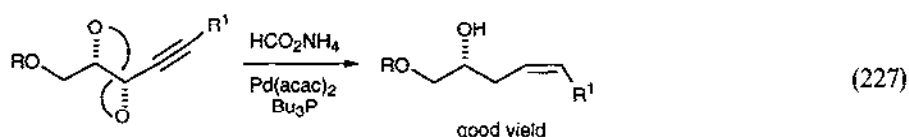


Palladium(0) complexes were used to cleave Fmoc protected peptides from Merrifield resins using allyl anchors [659]. The nitro group of *N*-(*p*-nitrophenyl)histidine was reduced to an amino group using palladium on carbon catalysts [660]. α -Amino acids were oxidized to nitrones by hydrogen peroxide/sodium tungstate [661]. Allyl

amines were converted to azadienes via zirconium chemistry (Eq. (226)) [662]. Iron tricarbonyl complexed acroleins were converted to imines via aza-Wittig chemistry [663]. Cinnamaldehyde RAMP hydrazones complexed stereoselectively to $\text{Fe}_2(\text{CO})_9$ [664].



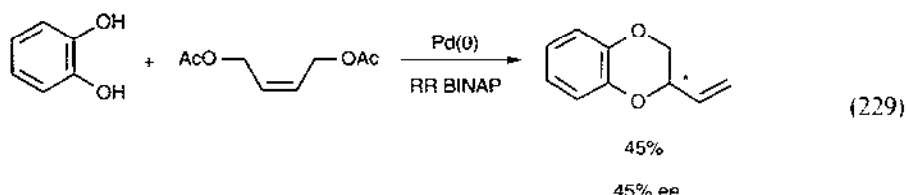
Catalytic asymmetric dihydroxylation has been reviewed (284 references) [665]. *O*-Allylphenols were deallylated to the free phenol by sodium borohydride in the presence of palladium(0) catalysts [666]. Propargyl carbonates were cleaved to homoallyl alcohols by palladium(0) formate (Eq. 227) [667]. Chiral titanocene BINOL complexes were efficient catalysts for the asymmetric reduction of Sp_2 -ketones to alcohols; a wide range of functional groups was tolerated [668]. Chromium tricarbonyl complexed phenethyl ketones were reduced by DIBAH with high diastereoselectivity (Eq. (228)) [669]. Rhodium-catalyzed decomposition of α -diazo esters in the presence of silanes [670] produced α -silyl systems that were converted to alcohols. When carried out in the presence of water α -hydroxy esters were generated directly [671].



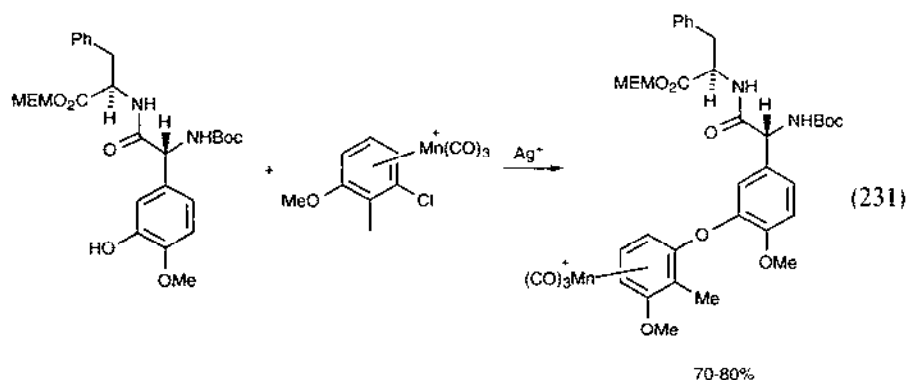
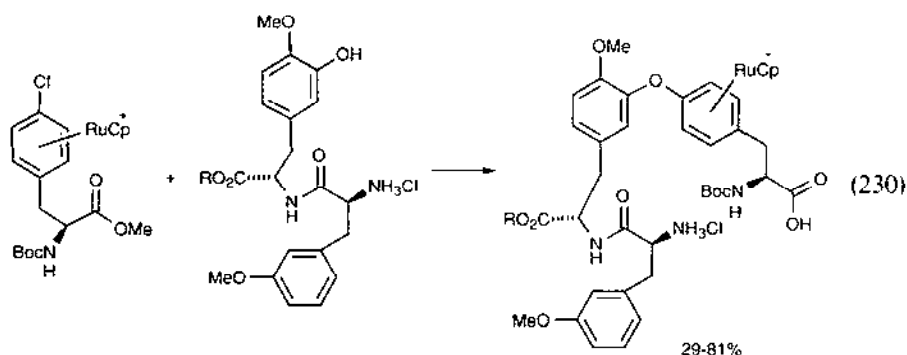
3.4. Ethers, esters, acids

Very efficient reoxidation processes for palladium catalyzed allylic acetoxylation of alkenes have been developed [672]. Evidence for π -allyl intermediates in this process has been acquired [673]. Acrylates also participate in this process [674]. All of the OH groups of a disaccharide were allylated by butadiene using palladium catalysts [675]. Palladium also catalyzed the α -alkoxylation of acetylene dicarboxy-

lates [676] and the formation of cyclic diethers from butenediol diacetates and hydroquinone (Eq. (229)) [677].

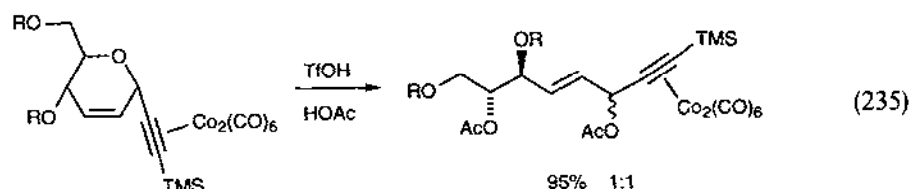
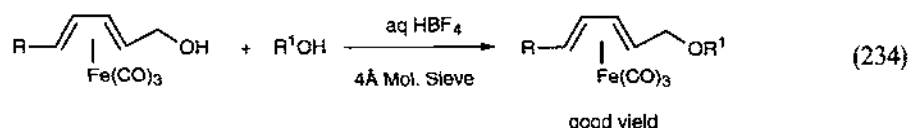
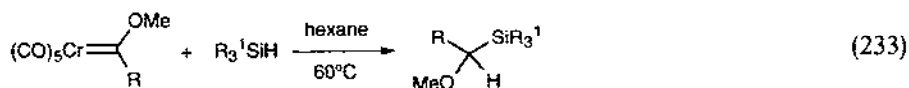
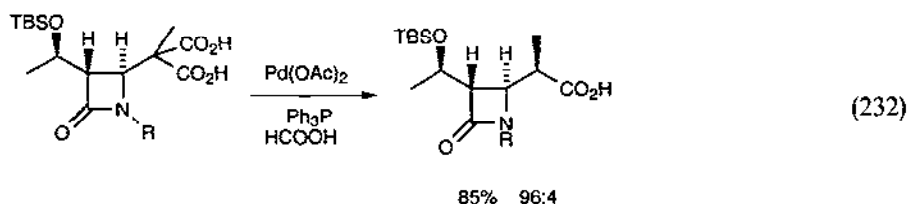


Cationic cyclopentadienyliron complexes of *p*-dichlorobenzene were dialkoxylated by phenols [678]. With *p*-dihydroquinone triarylethers were formed with monochlorobenzenes [679]. Cationic ruthenium (Eq. 230) [680] and manganese arenes (Eq. (231)) [681] were used to synthesize complex diaryl ethers.



Rhodium(II) carboxylate decomposition of α -diazo esters in the presence of alcohols produced α -alkoxyesters [682–684]. The complex RuH_2L_4 catalyzed the hydrolysis of nitriles to produce esters [685]. Palladium acetate catalyzed the decarboxylative deprotection of a β -lactam (Eq. (232)) [686]. The complex $[\text{CpFe}(\text{CO})_2]\text{K}$ disproportionated aldehydes to acids and alcohols, then condensed them to give esters [687]. Chromium alkoxy carbene complexes were cleaved by silane to produce α -alkoxysilanes (Eq. 233) [688]. The *O*-methyl group of anisoles was acylated by

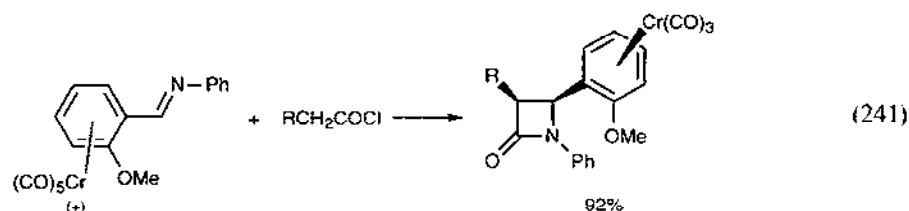
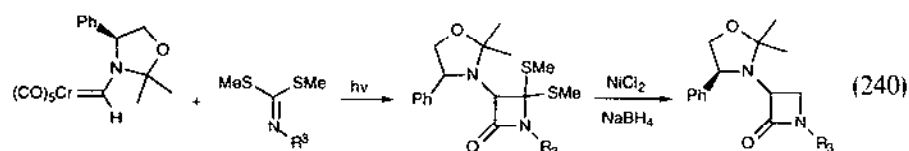
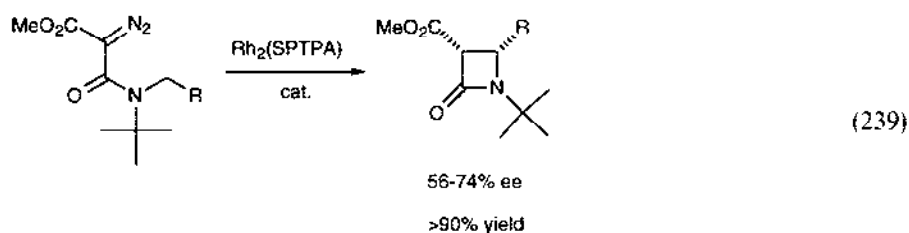
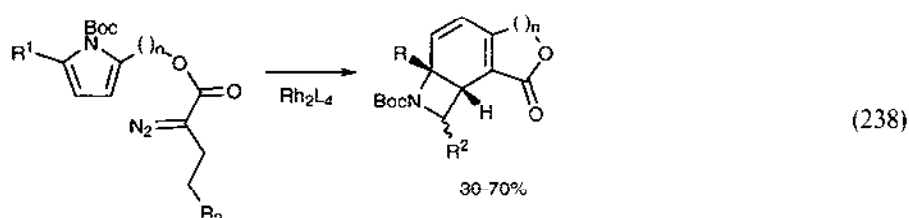
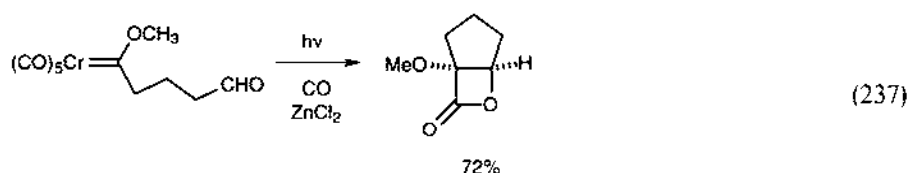
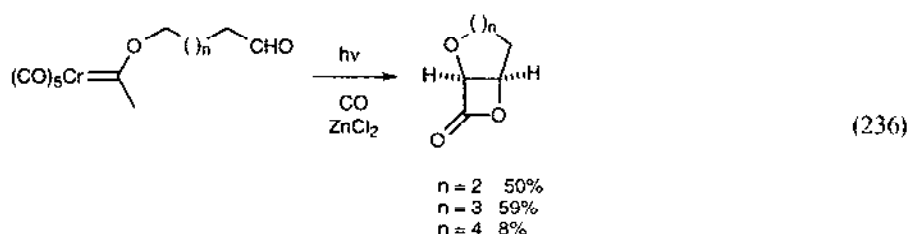
benzoic acid hydride in the presence of $\text{Pd}(\text{OAc})_2/\text{Sn}(\text{OAc})_2/\text{O}_2$ catalyst systems [689]. Iron-complexed diene alcohols were converted to ethers with Lewis acids (Eq. (234)) [690]. Triflic acid ring opened cobalt protected propargyl glycals (Eq. (235)) [691,692].



3.5. Heterocycles

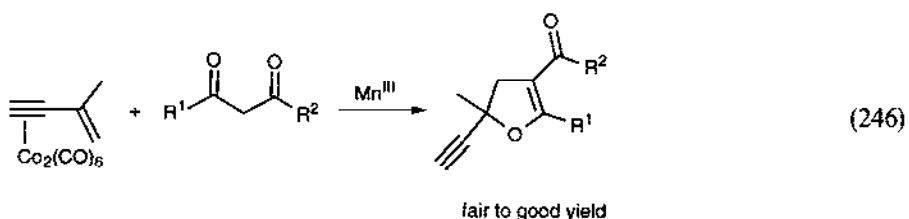
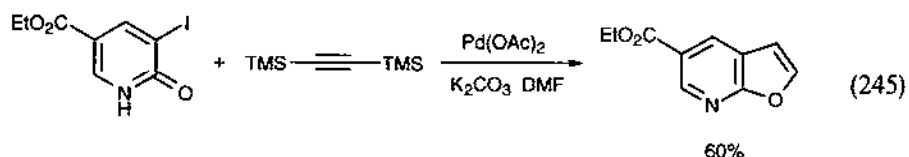
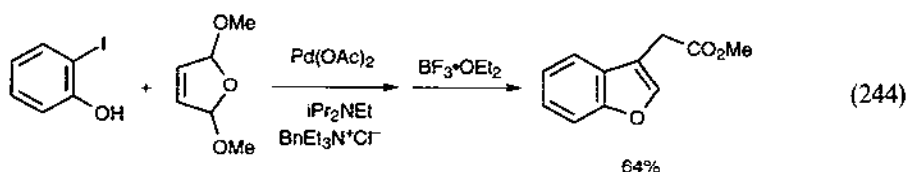
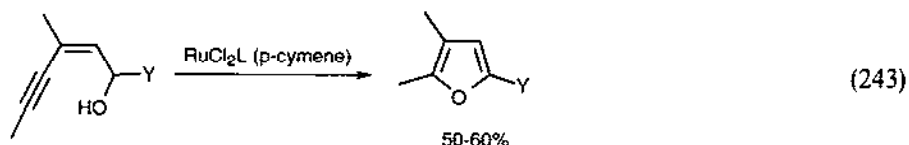
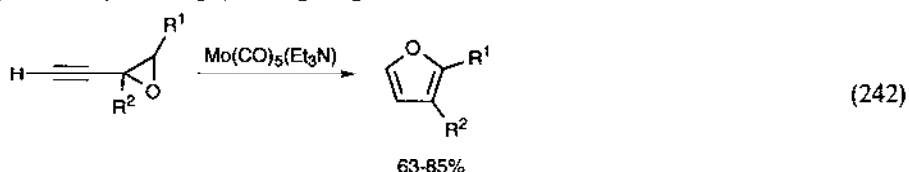
Chiral manganese salen complexes were efficient for the asymmetric epoxidation of trisubstituted olefins [693], and 1,3-dienes to give optical active mono-epoxides [694]. β -Ring steroidal olefins were epoxidized by $\text{MoO}_2(\text{acac})_2$ and aldehydes [695]. Rhodium(II) acetate catalyzed the insertion of phenyl diazomethane into the $\text{C}=\text{O}$ of aldehydes to produce *trans*-epoxides [696]. The full paper on the copper-catalyzed aziridination of alkenes by $\text{PhI}=\text{NTs}$ has appeared [697].

β -Lactones were prepared by the intramolecular reaction of aldehydes with chromium carbene photogenerated ketenes (Eqs. (236) and (237)) [698]. Azetidines (Eq. (238)) [699] and β -lactams (Eq. (239)) [700] were prepared by rhodium(II) catalyzed diazoalkene decomposition. β -Lactams were synthesized by chromium carbene photochemistry (Eq. (240)) [701], and chromium tricarbonyl complexed imine/ketene chemistry (Eq. (241)) [702]. *N*-Propargyl β -lactams were depropargylated via complexation to dicobalt octacarbonyl [703].

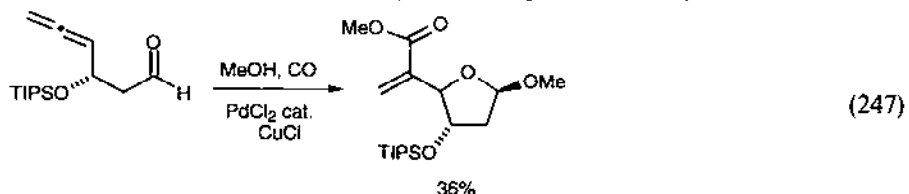


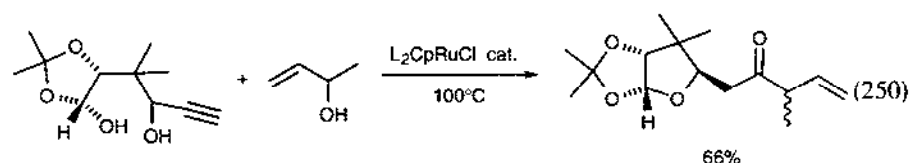
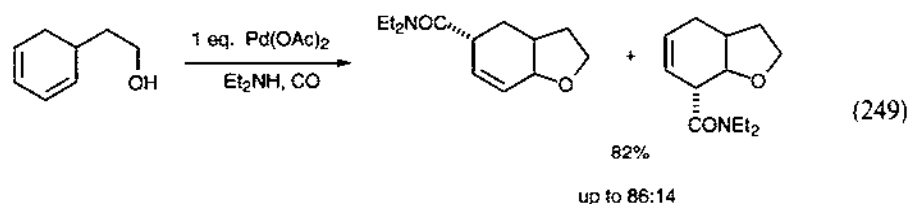
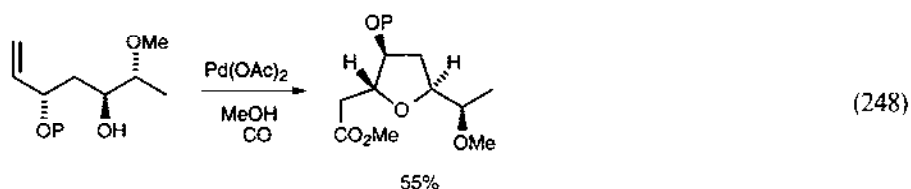
Molybdenum carbonyl rearranged propargyl epoxides to furans (Eq. (242)) [704], while ruthenium(II) complexes cyclized enynols to furans (Eq. (243)) [705]. Rhodium(II) catalyzed decomposition of α -diazo- β -dicarbonyl compounds with

alkynes to produce furans [706,707], as did palladium-catalyzed insertion chemistry (Eqs. (244) [708] and (245) [709]). Dihydrofurans were produced by the manganese(III) promoted reaction of β -dicarbonyl compounds with cobalt-complexed enynes (Eq. (246)) [710].

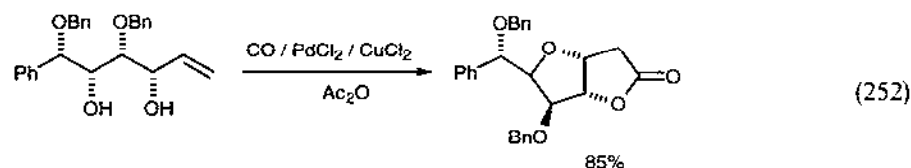
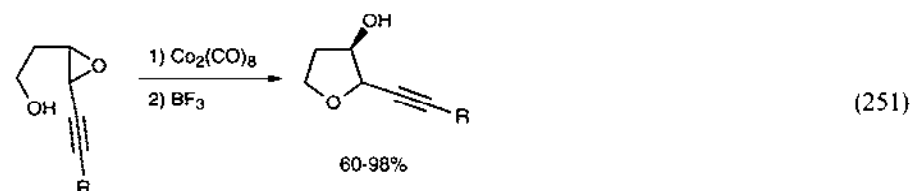


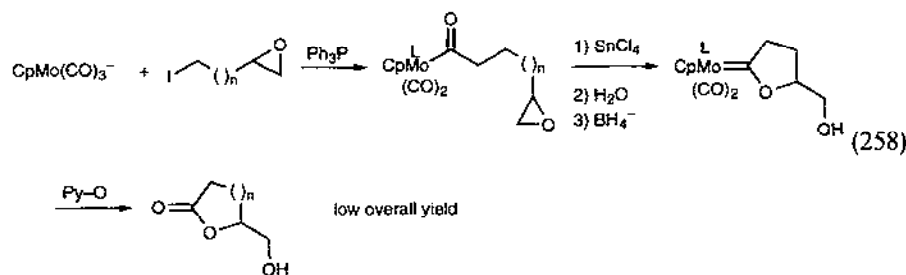
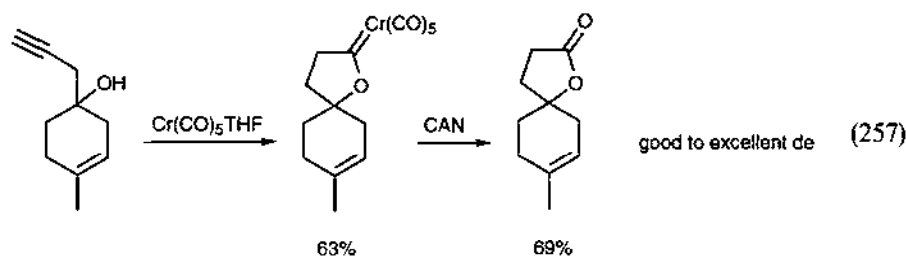
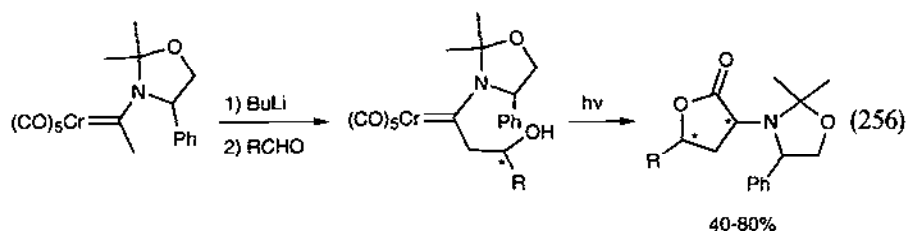
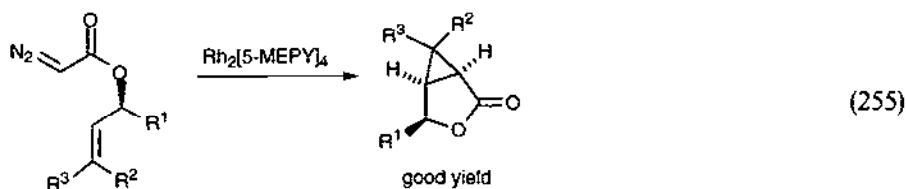
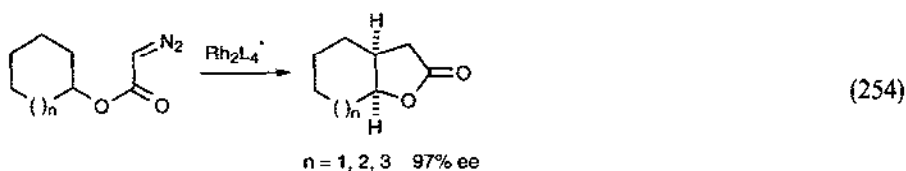
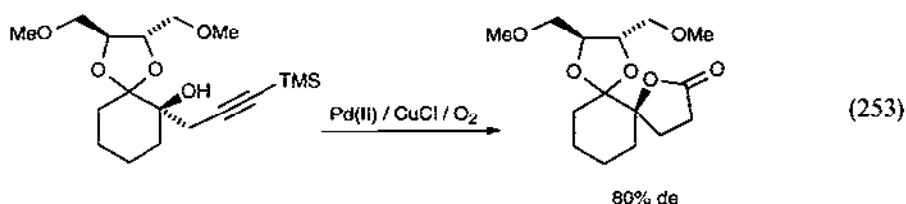
Tetrahydrofurans were produced by palladium-catalyzed alkoxylation of allenes (Eq. (247)) [711,712], alkenes (Eq. (248)) [713] and dienes (Eq. (249)) [714], by copper triflate-catalyzed decomposition of diazoesters in the presence of octanes [715], and aldehydes with electron-deficient olefins [716], by the ruthenium(II)-catalyzed reaction of propargyl alcohols with allylic alcohols (Eq. (250)) [717] and by cobalt carbonyl assisted opening of hydroxy epoxides (Eq. (251)) [718].

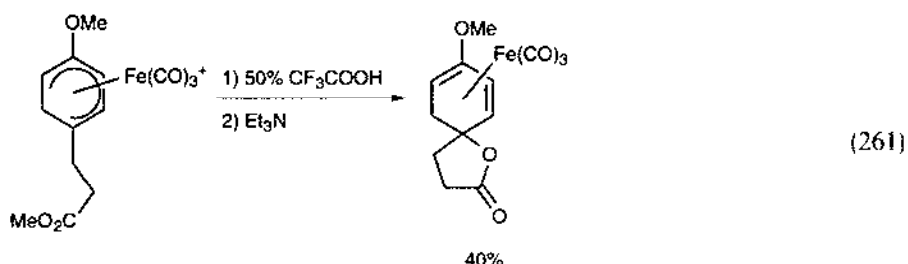
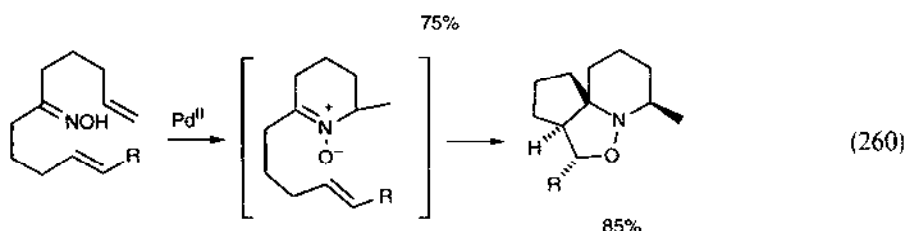
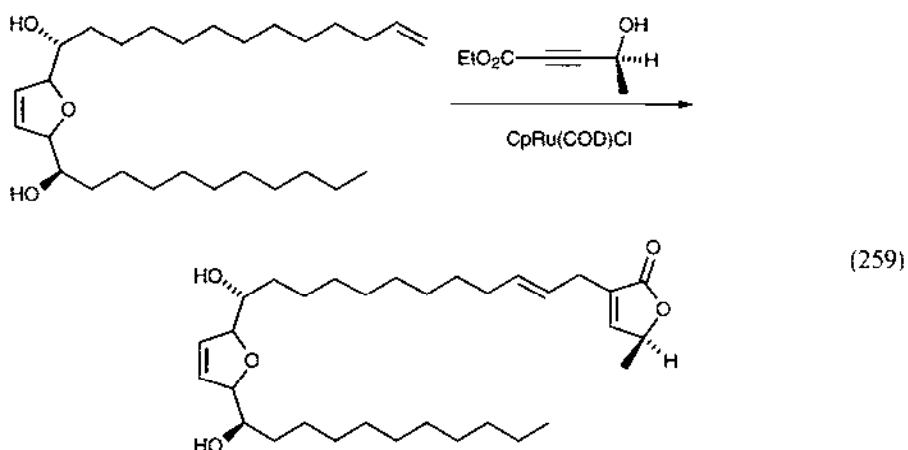




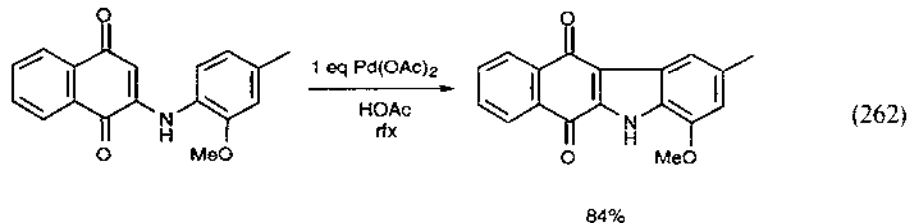
Butyrolactones were synthesized by palladium-catalyzed carbonylation cyclization of allyl (Eq. (252)) [719] and propargyl alcohols (Eq. (253)) [720], and rhodium(II)-catalyzed diazo decomposition C–H insertion (Eq. (254)) [721–724] and cyclopropanation (Eq. 255) [725]. Optically active α -amino- γ -butyrolactones were synthesized by combined aldol/photocycloaddition reactions of optically active chromium carbene complexes (Eq. (256)) [726]. Other carbene complex approaches are shown in Eqs. (257) [727] and (258) [728]. Ruthenium(II) complexes produced butenolides from alkenes and propargyl alcohols (Eq. (259)) [729]. Palladium(II) complexes catalyzed the amination of alkenes by oximes to give nitrones (Eq. (260)) [730]. Cationic iron tricarbonyl dienyl complexes underwent acid-catalyzed cyclization to generate butyrolactones (Eq. (261)) [731].

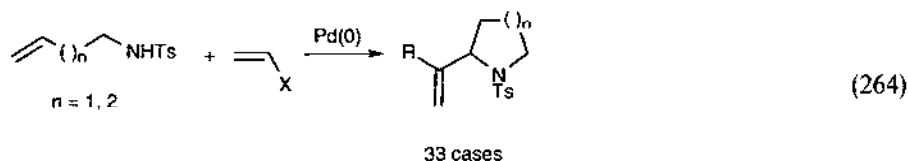
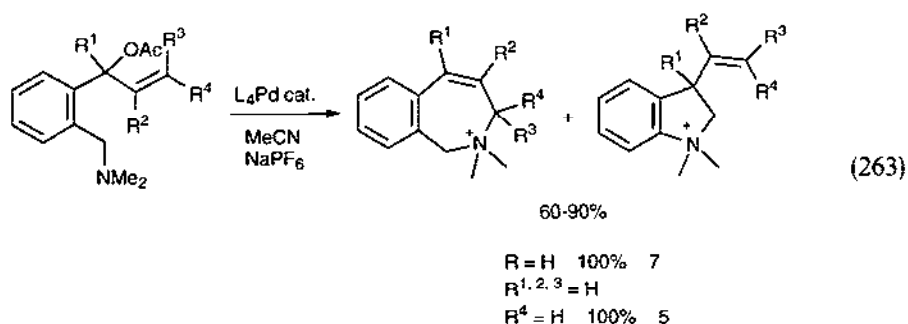




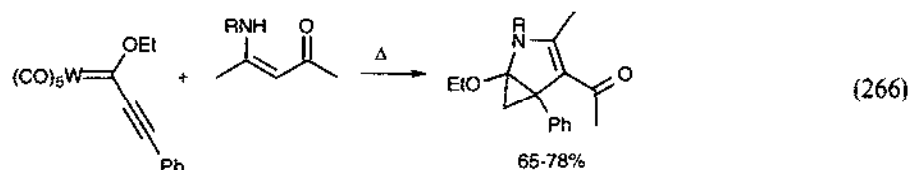
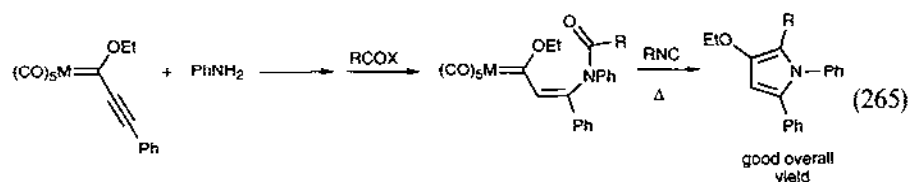


Ortho-nitrostyrenes were reductively cyclized to indoles by $\text{PdCl}_2\text{L}_2/\text{CO}/\text{SnCl}_2$ in fair to good yield [732]. *ortho*-iodo anilines [733] and *ortho*-palladated anilines [734] underwent palladium-assisted reactions with alkynes to produce indoles. One equivalent of palladium(II) acetate was required to couple 2-anilinoanthraquinones to the indoloquinone (Eq. (262)) [735]. Palladium(0) complexes catalyzed allylic (Eq. (263)) [736] and olefinic aminations (Eq. (264)) [737], while palladium(II) complexes catalyzed the cyclization of olefinic amines to make pyrroles and dihydropyrroles [738].

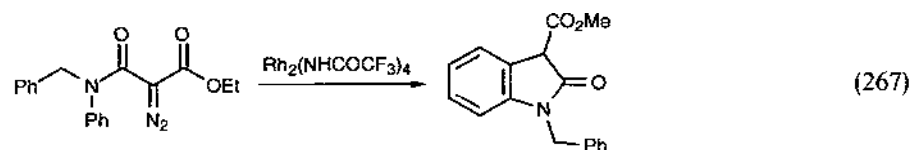


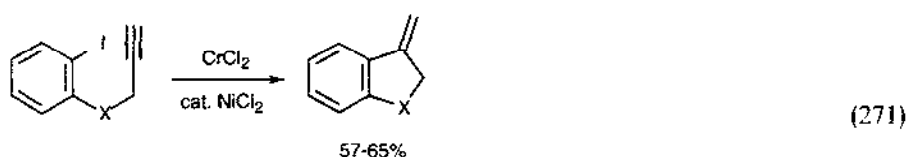
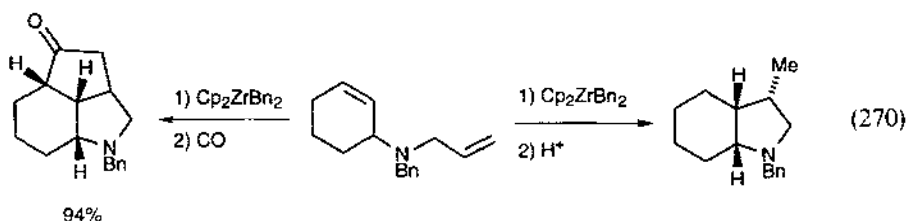
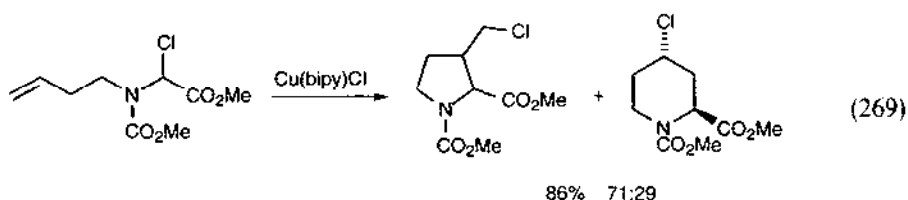
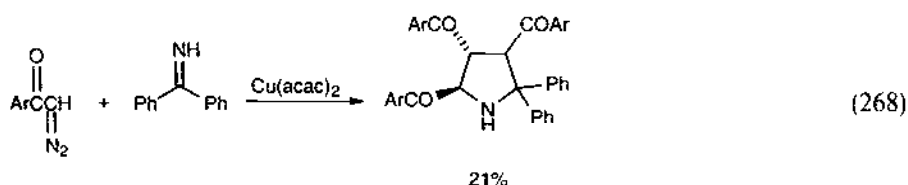


Alkynyl carbene complexes of Group 6 metals underwent reaction with amines followed by isonitriles to produce pyrroles (Eq. (265)) [739,740]. Similar products resulted from the reaction of alkoxy carbene complexes with 1-azadienes [741] and acetylenic carbene complexes with imines [742]. Vinyllogous amides produced bicyclic compounds (Eq. (266)) [743].

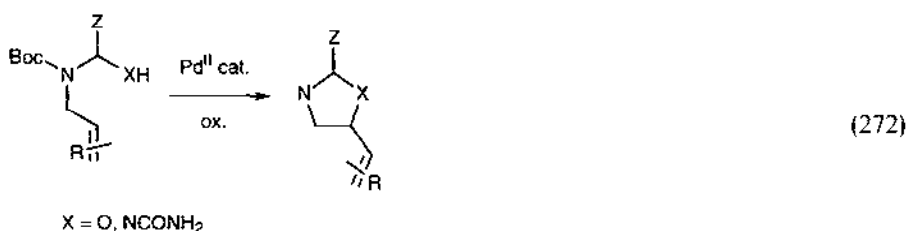


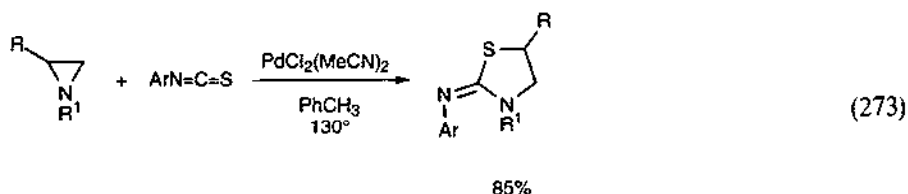
Five-membered nitrogen heterocycles were made by metal-catalyzed decomposition of diazoalkanes (Eqs. (267) [744] and (268)) [745], by copper-catalyzed chloroalkylation of olefinic amines (Eq. (269)) [746], and by zirconium- (Eq. (270)) [747] and chromium-catalyzed (Eq. (271)) [748] reductive cyclization (Eq. (270)) [747].



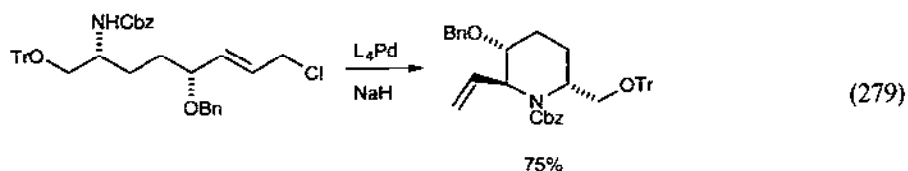
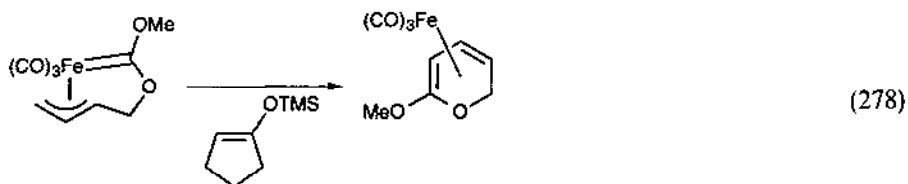
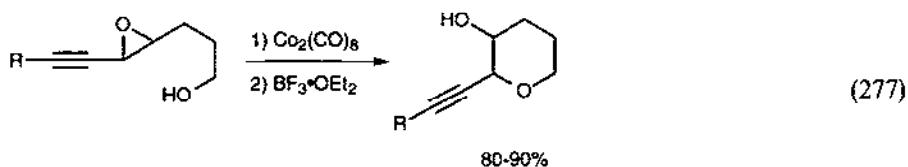
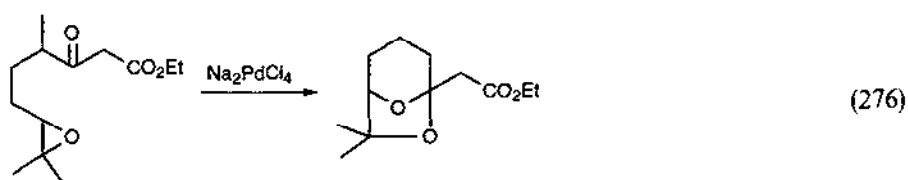
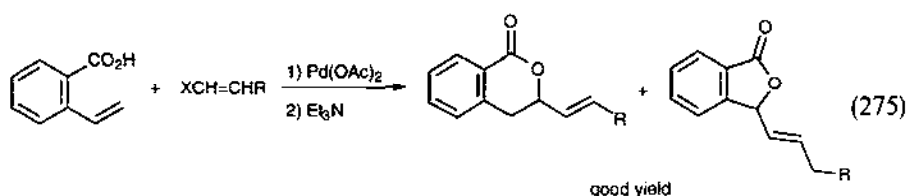
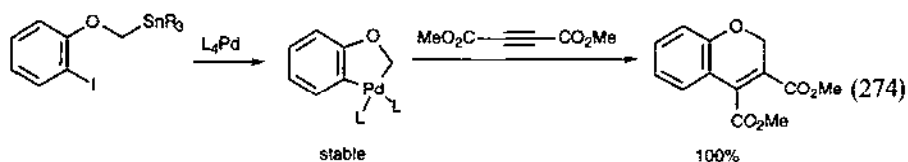


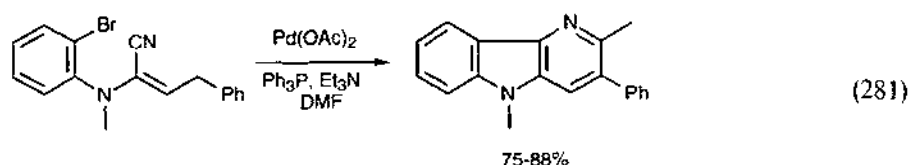
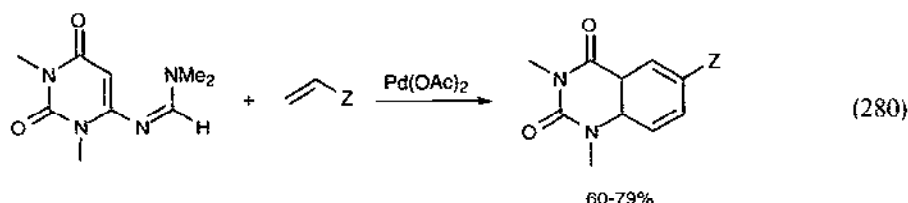
Five-membered *N,O*- [749,750] and *N,N*-heterocycles [751] were made by palladium(II)-catalyzed alkoxylation or amination of olefins (Eq. 272). Vinyl carbonates were converted to vinyl ureas by treatment with isocyanates and a palladium catalyst [752]. Isothiocyanates combined with aziridines in the presence of a palladium(II) catalyst to give *S,N*-heterocycles (Eq. (273)) [753]. Benzothiazoles were produced by the palladium(0)-catalyzed coupling of aryl iodides with *ortho* amino thiophenol under a CO pressure [754]. Rhodium(II)-catalyzed decompositions of α -diazo carbonyls in the presence of nitriles gave oxazoles [755,756]. Rhodium(I) catalyzed the carbonylation of cyclic thioaminals to thioureas [757].





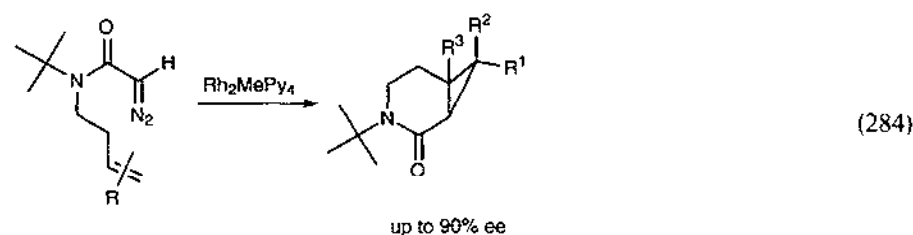
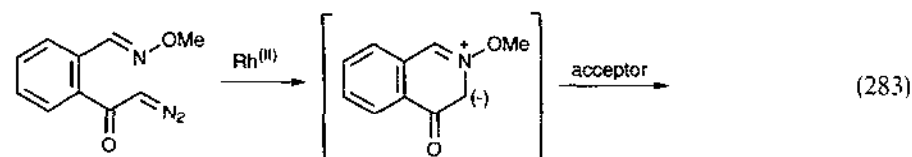
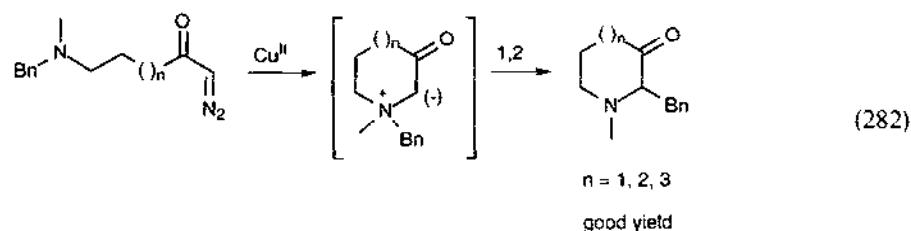
Six-membered oxygen heterocycles were prepared in a variety of ways (Eqs. (274) [758], (275) [759], (276) [760], (277) [761] and (278) [762]).

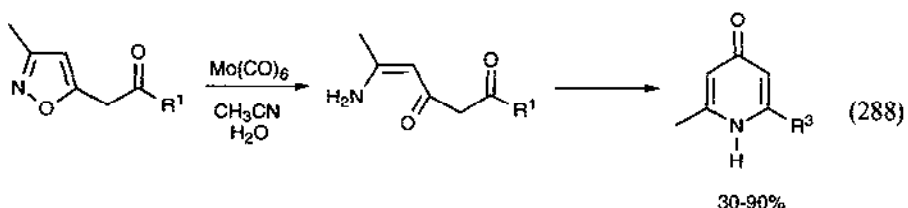
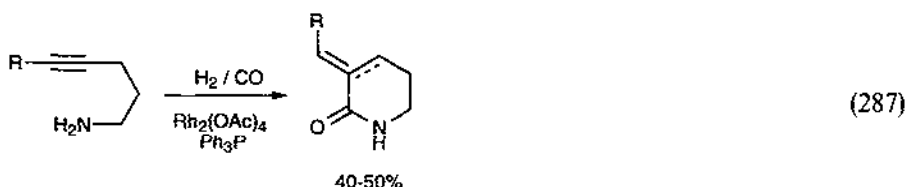
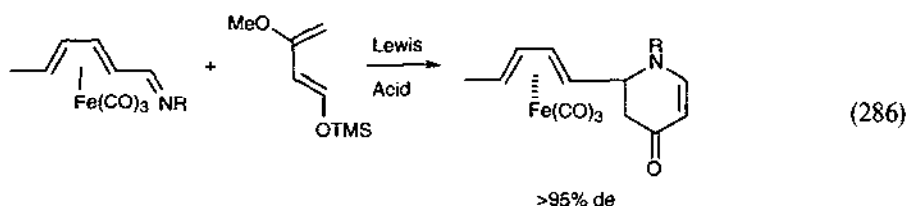
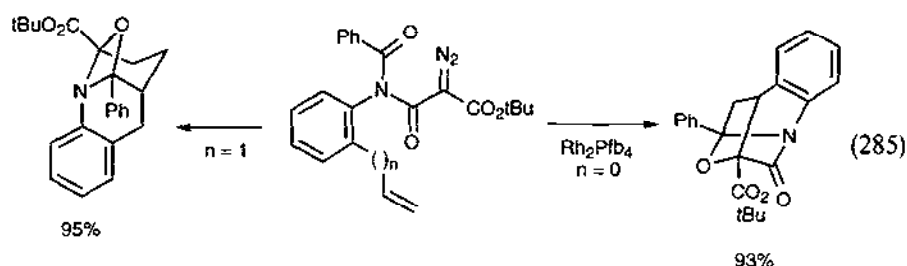




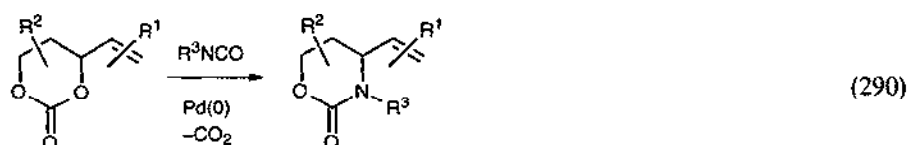
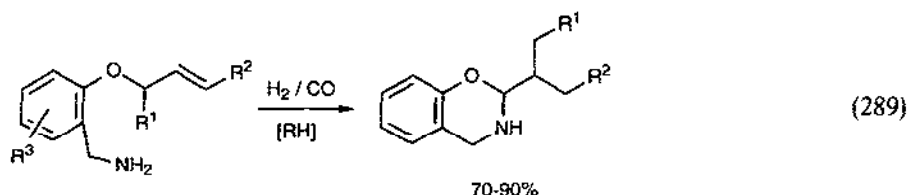
Palladium(0) complexes catalyzed the coupling of aryl halides with *ortho* boronated pyridines [763]. *ortho*-allyl-*N,N*-dimethyl anilines cyclized to the dimethyl dihydroquinoline salts under palladium catalysis [764]. Palladium(0) complexes catalyzed the formation of six-membered heterocycles (Eqs. (279) [765], (280) [766] and (281) [767]).

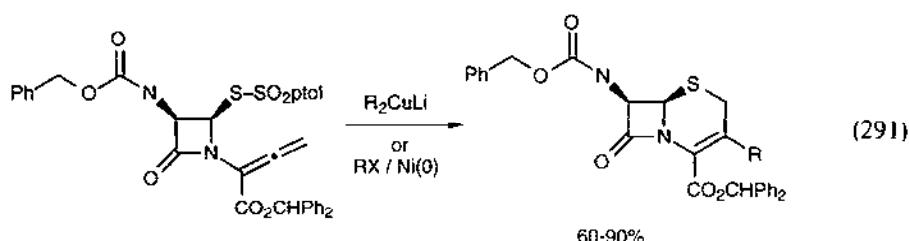
Metal-catalyzed diazo compound decomposition (Eqs. (282) [768], (283) [769], (284) [770] and (285) [771]) gave six-membered nitrogen heterocycles, as did iron diene chemistry (Eq. (286)) [772], rhodium catalyzed hydrocyclization of alkynes (Eq. (287)) [773] and molybdenum chemistry (Eq. (288)) [774].



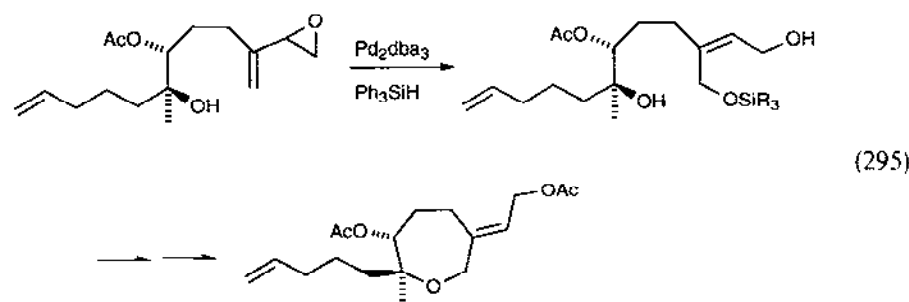
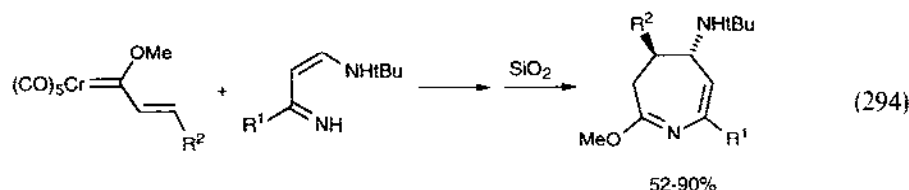
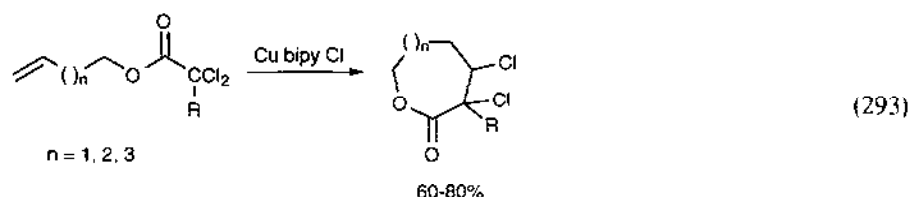
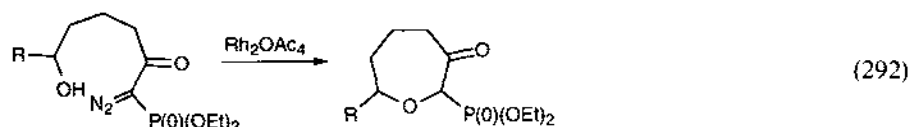


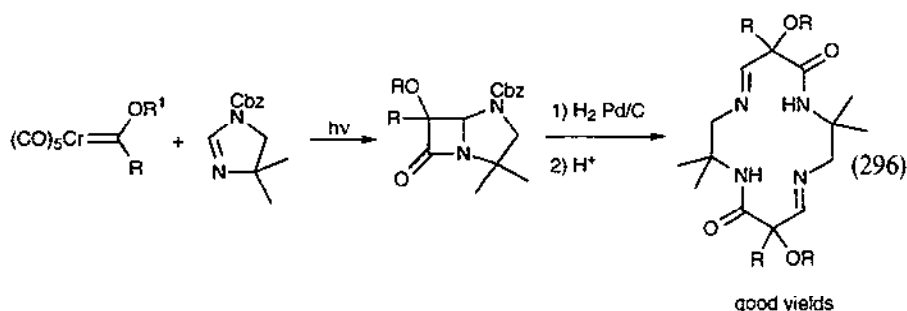
Morpholines were prepared by copper-catalyzed diazo decomposition/C–H insertion [775]. Rhodium-catalyzed olefin amination formed oxazoles (Eq. (289)) [776]. Palladium-catalyzed the O,N-exchange of cyclic carbonates with retention of stereochemistry (Eq. (290)) [777]. Both copper [778] (Eq. 291) and nickel [779] cyclized *N*-allenyl β -lactams.





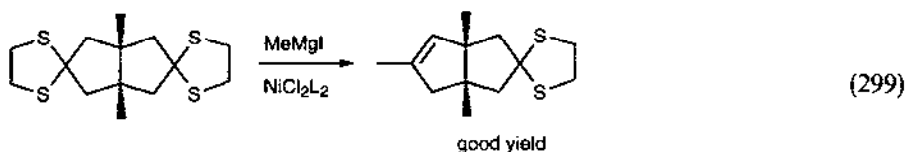
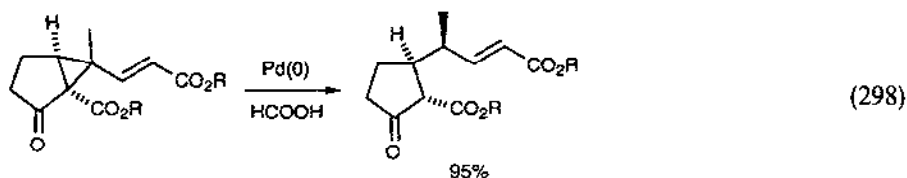
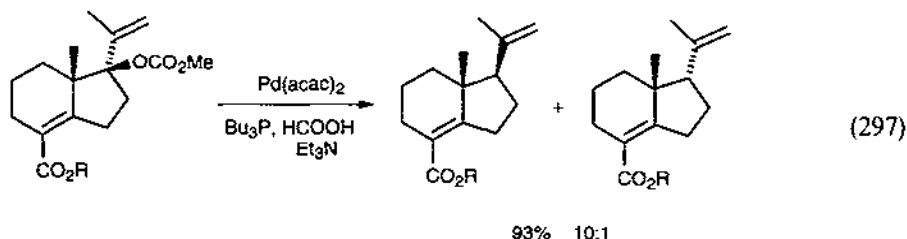
Seven-membered heterocycles were made by diazo decomposition OH insertion (Eq. (292)) [780], copper-catalyzed chloroalkylation of alkenes (Eq. (293)) [781], and cycloadditions to chromium carbene complexes (Eq. (294)) [782]. Palladium catalysis was central in the synthesis of elaborated oxepines (Eq. (295)) [783]. Photolysis of chromium carbene complexes with imidazolines produced azapenams which dimerized to cyclams (Eq. (296)) [784]. 'Recent Advances in the Cycloaddition Chemistry of Isomunchnones and Thioisomunchinones, an Under Utilized Class of Mesionic Compounds' is the title of a review (107 references) [785] with many cases of rhodium-catalyzed diazo decomposition reactions.

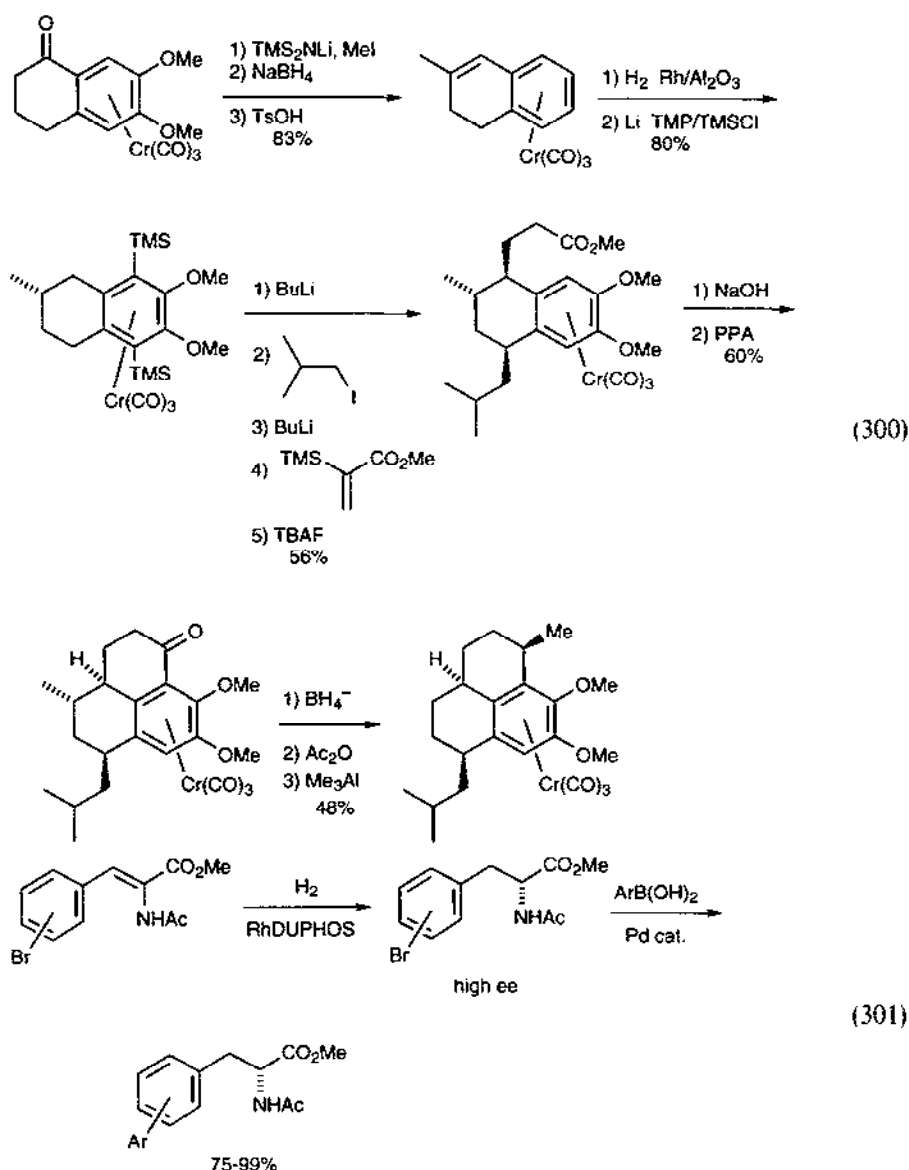




3.6. Alkenes, Alkanes

Allylic carbonates were reduced to alkenes with Sn_2' regiochemistry and with very high ee using palladium(0) R-MOP catalysts and formate as the hydride source [786–788]. Achiral complexes also catalyzed this reduction (Eq. (297)) [789,790]. A similar catalyst system reductively ring opened vinylcyclopropanes (Eq. (298)) [791]. Dithianes were converted to alkenes by nickel-catalyzed Grignard reactions (Eq. (299)) [792]. Reduction and elimination of chromium tricarbonyl complexed decalenones was useful in complex syntheses (Eq. (300)) [793]. Rhodium(II) complexes catalyzed the elimination of α -diazo- β -alkoxy esters to β -alkoxyenones [794,795]. Bromo acetamido acrylates were asymmetrically reduced, then functionalized using Suzuki coupling (Eq. (301)) [796]. Tritiated $\text{Cp}_2\text{Zr} (^3\text{H})\text{Cl}$ was prepared for use in tritium labelling compounds [797]. A new chiral ferrocenyl diphosphine ligand Josiphos, has been developed for the asymmetric reduction of allylic acetates [798].

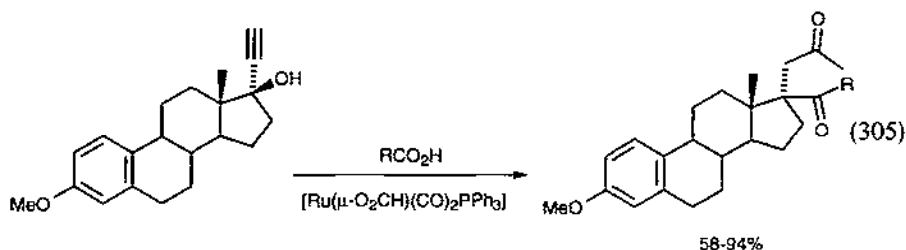
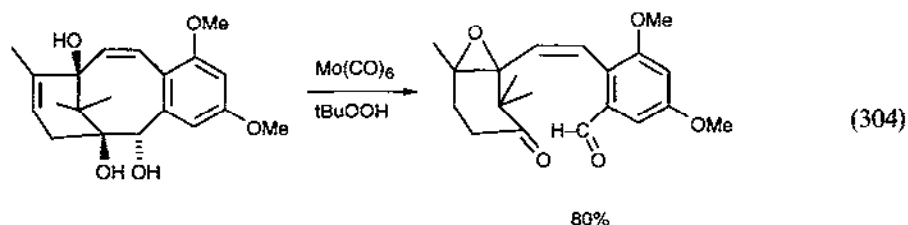
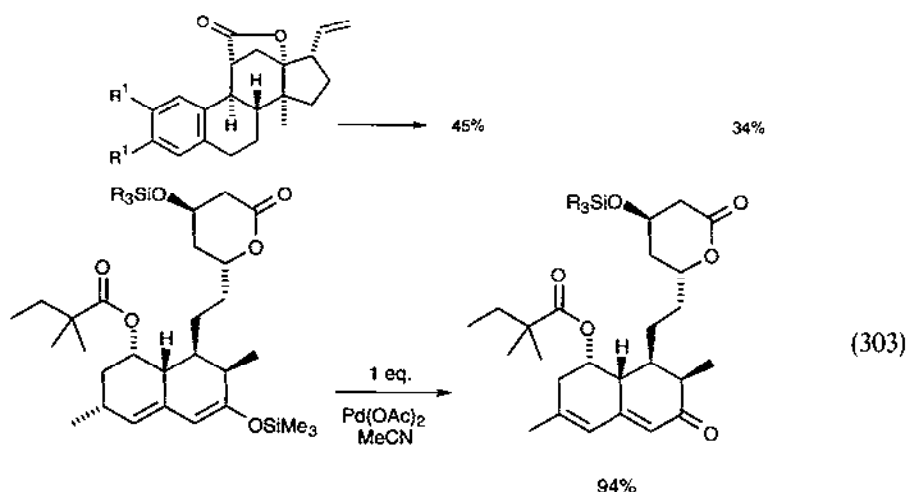
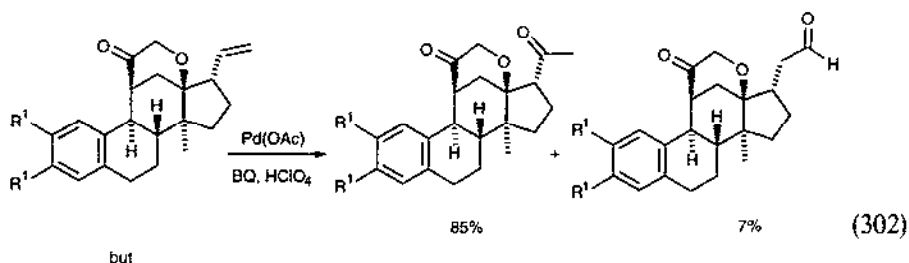




3.7. Ketones, aldehydes

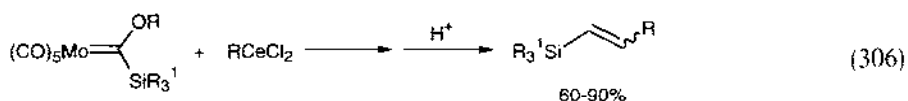
Palladium dibenzilidene acetone complexes catalyzed the oxidation of allylic alcohols and esters to α,β -unsaturated aldehydes [799]. The Wacker oxidation of C-17 steroidal alkenes was directed by adjacent functionality (Eq. (302)) [800]. Palladium(0) catalyzed the conversion of butene 1,2- or 1,4-diols to unsaturated aldehydes [801]. Silylenol ethers were oxidized to enones by stoichiometric quantities of palladium(II) acetate (Eq. (303)) [802]. Pyrrolidines with pendent alkene groups

underwent clean Wacker oxidation to the methyl ketones [803]. Diols were oxidatively cleaved by $\text{Mo}(\text{CO})_6$ and *t*-butylhydroperoxide (Eq. (304)) [804]. Efficient oxidation of secondary alcohols to ketones has been achieved using MnO_2 and ruthenium catalyst [805]. C-17 ethynyl sterols were converted to gem diketones using ruthenium catalysts (eq. (305)) [806].

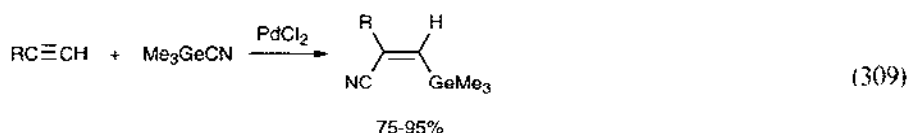
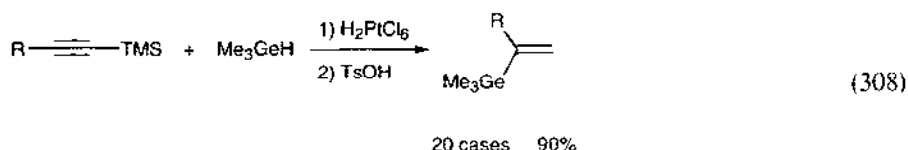
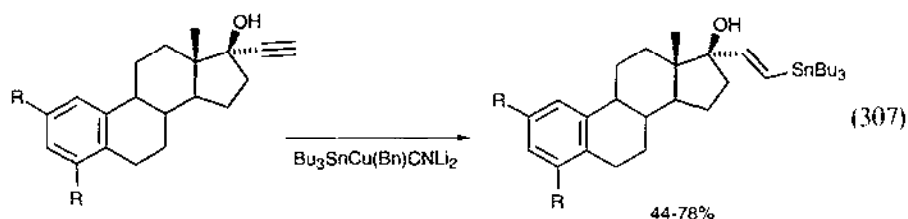


3.8. Organosilanes, germanes and stannanes

α -Diazoesters were converted to α -silyl esters by decomposition with rhodium(II) acetate in the presence of silanes [807]. Rhodium complexes catalyzed the hydrosilylation of enamides to put the silicon to nitrogen [808] and propargyl alcohols to put the silicon γ to oxygen [809]. A ruthenocene bisphosphine/amine ligand was an efficient catalyst for the conversion of allylic chlorides to allyl silanes with high ee [810]. Silyl cuprates β -silylated conjugated esters [811], while palladium(0) complexes-catalyzed the 1,2-*cis*-bis silylation of alkynes by disilanes [812]. Organocerium reagents converted molybdenum silyl carbene complexes into vinyl silanes (Eq. (306)) [813].



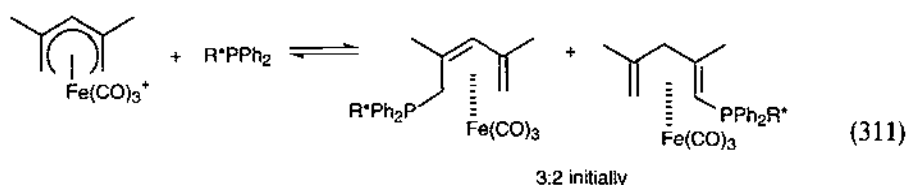
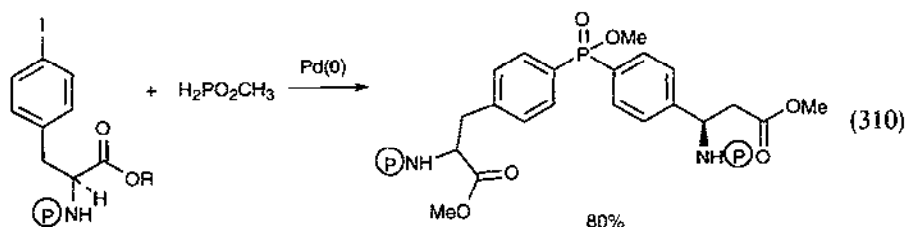
1-Tosyl-3-bromoindole was stannylated in the 3-position by treatment with hexamethylditin and a palladium(0) catalyst [814]. Palladium(0)-catalyzed the hydrostannylation of ynol ethers (Sn α to O) [815], propargyl glycines (Sn internal) [816] and propargyl alcohols (Sn remote from O) [817]. Stannyl cuprates added to ethynylsteroids (Eq. (307)) [818]. Ynol ethers were *cis*-bis-stannylated and silylstannylated by hexamethylditin and $\text{Me}_3\text{SnSiMe}_3$ respectively using palladium(0) catalysts [819]. Alkynes were germylated with platinum (Eq. (308)) [820] and palladium (Eq. (309)) [821] catalysts.



3.9. Organophosphorous and sulfur compounds

Binap phosphine ligands were prepared from binaphthol triflates by nickel [822] and palladium-catalyzed [823] reactions with phosphorous (Ph_2PH , Ph_2POH)

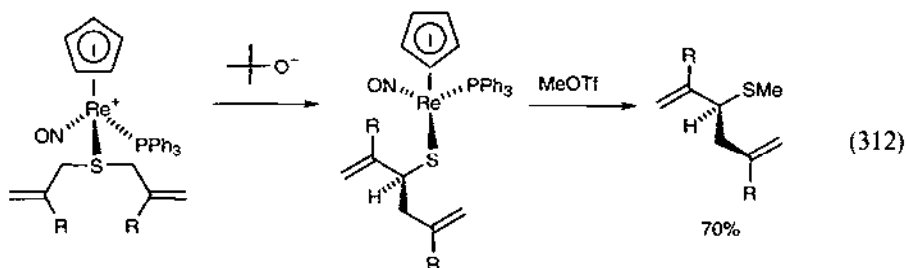
nucleophiles. Phenyl alanine-containing phosphorous compounds were made by palladium(0)-catalyzed coupling of the iodoarene to $\text{H}_2\text{PO}_2\text{CH}_3$ (Eq. (309)) [824]. Optically active tertiary phosphines added selectively to one face of the cationic iron tricarbonyl dienyl complex of the 2,4-dimethyl pentadienyl system (Eq. (310)) [825].

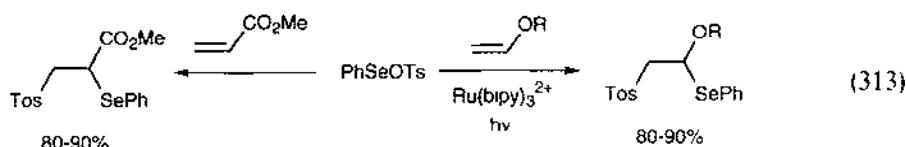


slow reaction $\rightleftharpoons$ >80% of one

Palladium complexes catalyzed the conversion of allyl carbonates to allyl thioethers (RSH nucleophile) [826], vinyl triflates to vinyl sulfides [827] (RSLi nucleophile), vinyl and aryl halides to vinyl and aryl TIPS sulfides [KSTIPS nucleophile] [828] and conjugated enynes to 2-thiophenyl-1,3-dienes (PhSH nucleophile) [829]. The allocam group $-\text{CH}_2\text{NHCO}_2\text{CH}_2\text{CH}=\text{CH}-$ was a good protecting group for the serine SH, stable to acid but not base, and easily removed by tributyltin hydride in the presence of palladium catalysts [830].

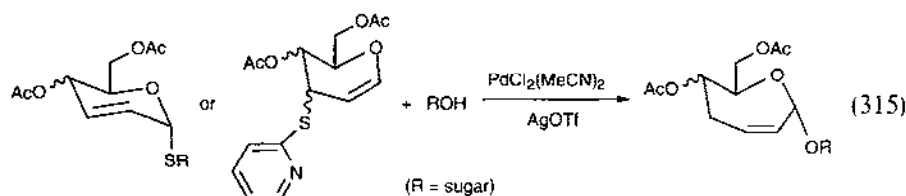
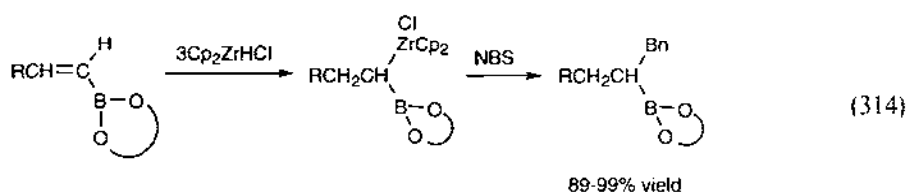
Chromium tricarbonyl complexed *p*-methylthiophenol methyl ether underwent asymmetric oxidation to the sulfoxides using (+)diethyl tartrate and cumene hydroperoxide [831,832]. This could be further oxidized to the sulfone [833]. α -Thiophenyl ketones were produced by rhodium(II) acetate catalyst decomposition of α -diazoketones in the presence of thiophenols [834]. Nickel(II) catalyzed the conversion of thiocarbamoyl chlorides to thioamides by Grignard reagents [835]. Porphyrin iron(III) complexes catalyzed the Markovnikov addition of thiophenol to styrenes [836]. Other unusual processes are shown in Eqs. (310) [837] and (311) [838].





3.10. Miscellaneous

Lipase was used to selectively acetoxyrate one enantiomer of chromium tricarbonyl complexes of *o*-methyl benzyl alcohol and iron tricarbonyl complexes of 5-substituted penta-2,4-dien-1-ols, resulting in resolution [839]. Alkyl diphenylsulfonium salts were effective *o*-alkylating agents for anionic Group 6 pentacarbonyl acylate complexes to produce alkoxy carbene complexes [840]. The iron tricarbonyl complex of substituted *N*-aryl imines of cinnamaldehyde was an excellent source of the iron tricarbonyl fragment for the preparation of dieneiron tricarbonyl complexes [841]. α -Bromoborates were made by hydrozirconation/bromination of vinyl boronates (Eq. (312)) [842]. Palladium(II)/silver(I) catalysis of the coupling of thioglucosides with sugars was α -selective (Eq. (313)) [843].



4. Reviews

The following reviews have appeared:

Enhanced Synthetic Efficiency Towards Natural Products via Transition Metal Catalyzed Reactions (27 references) [844].

Stepwise Electrophilic Addition. Some Novel Synthetic Ramifications of an Old Concept (>100 references) [845].

MOP: A Monodentate Chiral Phosphine Ligand for Catalytic Asymmetric Reactions (35 references) [846].

Catalytic Addition of CH Bonds to Multiple Bonds. The First Stage (34 references) [847].

Resolution Racemization and Epimerization in Acyclic(diene) $\text{Fe}(\text{CO})_2\text{L}$ Complexes (full paper) [848].

Transition Metals in Organic Synthesis: Hydroformylation Reductions and Oxidation 1992 (784 references) [849].

Transition Metals in Organic Synthesis, Annual Survey 1992 (1033 references) [850].

Asymmetric Synthesis of 2,3-Methanoamino Acids (60 references) [851].

The Development of Organometallic Methodologies for the Stereospecific Introduction of Cephalosporin Side Chains (45 references) [852].

Chromium(II) Based Methods for Carbon–Carbon Bond Formation (16 references) [853].

Tetrapropylammonium Perruthenate, $\text{Pr}_4\text{N}^+\text{RuO}_4^-$; A Catalytic Oxidant for Organic Synthesis (142 references) [854].

Tricarbonyliron Lactones Complexes in Organic Synthesis (30 references) [855].

New Synthetic Methods Based on Organozirconium and Organocopper Chemistry (19 references) [856].

Organonickel Chemistry in Organic Synthesis. Some Applications of Alkyl and Metalacyclic Derivatives (>28 references) [857].

Transformations of Chloroarenes Catalyzed by Transition Metals (179 references) [858].

Organic Synthesis with α -Diazocarbonyl Compounds (786 references) [859].

Transition Metal Complexes in the Chemistry of Vinylcyclopropanes (115 references) [860].

Organometallic Ways for the Multiplications of Chirality (333 references) [861].

Activation of Carbon–Fluorine Bonds by Metal Complexes (400 references) [862].

Transition Metal Promoted Free-Radical Reactions in Organic Synthesis: The Formation of Carbon–Carbon Bonds (363 references) [863].

Nitrogen Donors in Organometallic Chemistry and Homogeneous Catalysis (170 references) [864].

Fluorinated Organometallics: Vinyl, Alkynyl, Allyl, Benzyl, Propargyl, Aryl, Fluorinated Organometallic Reagents in Organic Synthesis (297 references) [865].

Review of Taxol (221 references) [866].

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