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Non-conventional methodologies for transition-metal catalysed carbon–carbon coupling: a critical overview.

Part 1: The Heck reaction

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Keywords: Heck reaction; Supercritical fluids; Ionic liquids; Fluorous media; Aqueous solvents; Microwave; Ultrasound; High pressure; Nanofiltration; Microreactors; Ball-milling conditions.

Abbreviations: A, ampere; AAEMA, deprotonated form of 2-(acetoacetoxy)ethyl methacrylate; acac, acetylacetonate; Ad, adamantyl; A336, tricaprylmethylammonium chloride; atm, atmosphere; bbim, 1,3-di-*n*-butylimidazolium; *b*, block copolymer; bmim, 1-butyl-3-methylimidazolium; BINAP, 2,2'-bis(diphenylphosphino)-1,1'-binaphthyl; Boc, *tert*-butoxycarbonyl; BTF, benzotrifluoride (α,α,α -trifluorotoluene); Cbz, benzyloxycarbonyl; *co*, copolymer; COD, 1,5-cyclooctadiene; Cy, cyclohexyl; DAB, 1,4-diaminobutane; dba, dibenzylideneacetone; DBU, 1,8-diazabicyclo[5.4.0]undec-7-ene; DEA, *N,N*-diethylacetamide; DEC, dendrimer-encapsulated catalyst; dendr, dendrimer; DIPEA, diisopropylethylamine; DMA, *N,N*-dimethylacetamide; DMF, dimethylformamide; DMG, *N,N*-dimethylglycine; dppp, 1,3-diphenylphosphinopropane; EDG, electron-donating group; EWG, electron-withdrawing group; Fc, ferrocenyl; F-dppp, fluorine-tagged 1,3-bis(diphenylphosphino)propane; HDAPS, *N*-hexadecyl-*N,N*-dimethyl-3-ammonio-2-propanesulfonate; Hex, hexyl; IL, ionic liquid; LDH, layered double hydroxide; M, metal; MCM-41, hexagonally packed mesoporous molecular sieves; MW, microwave; Nf, nonaflate (nonafluoro-*n*-butane-1-sulfonyl); NMP, *N*-methylpyrrolidinone; P_c , critical pressure; PAMAM, poly(amidoamine); PEG, poly(ethylene glycol); pmim, 1-*n*-pentyl-3-methylimidazolium; PNIPAM, poly(*N*-isopropylacrylamide); PNP, *p*-nitrophenyl; PS, polystyrene; Py, pyridyl; RCM, ring-closing metathesis; ROMP, ring-opening metathesis polymerisation; rt, room temperature; SAPO, silico aluminophosphate; sc, supercritical; SCF, supercritical fluid; SCM, shell cross-linked micelles; SDS, sodium dodecyl sulfate; TBAA, tetra-*n*-butyl ammonium acetate; TBAB, tetra-*n*-butyl ammonium bromide; TBAC, tetra-*n*-butyl ammonium chloride; TBAF, tetra-*n*-butyl ammonium fluoride; TEAC, tetraethylammonium chloride; Tf, trifluoromethanesulfonyl; TPPDS, bis(*p*-sulfonatophenyl)phenylphosphane dipotassium salt; Na-TPPMS, mono(*m*-sulfonatophenyl)diphenylphosphane monosodium salt; *m*-TPPTC, tris(*m*-carboxyphenyl)phosphane trilithium salt; TPPTS, tris(*m*-sulfonatophenyl)phosphane trisodium salt; TXPTS, tris(4,6-dimethyl-3-sulfonatophenyl)phosphane trisodium salt; T_c , critical temperature; THF, tetrahydrofuran; THP, tetrahydropyran; TFA, trifluoroacetic acid; Tol, tolyl; TOF, turnover frequency (mol of product per mol of catalyst h^{-1}); TON, turnover number (mol of product per mol of catalyst); Ts, *p*-toluenesulfonyl; ttmp, tris-(2,4,5-trimethoxyphenyl)phosphane; wt, weight; XAFS, X-ray absorption fine structure.

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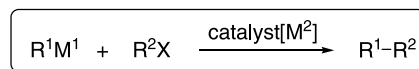
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1. General introduction

The formation of carbon–carbon bonds is a fundamental reaction in organic synthesis the efficiency of which has interested organic chemists for a long time ago. Aryl–aryl bond formation has been known for more than a century and was one of the first reactions involving a transition metal.¹ Modern synthetic chemistry is also sustained by the use of transition-metal catalysts as powerful tools for carbon–carbon bond-forming processes.² Among these, carbon–carbon coupling reactions through the activation of carbon–hydrogen bonds,³ as well as addition reactions,⁴ have experienced an increasing interest in the preparation of molecules, the access to which is not so straightforward using other methodologies. On the other hand, the transition-metal catalysed carbon–carbon bond formation developed in the 1970s represented a milestone in synthetic organic chemistry that allowed the cross coupling of substrates in ways that would have previously been thought impossible.⁵ This protocol has been substantially improved

and expanded over the past 30 years, providing an indispensable and simple methodology for preparative organic chemists (Scheme 1).



M¹ = Li (Murahashi)
Mg (Kumada-Tamayo, Corriu)
B (Suzuki-Miyaura)
Al (Nozaki-Oshima, Negishi)
Si (Tamayo-Kumada, Hiyama-Hatanaka)
Zn (Negishi)
Cu (Normant)
Zr (Negishi)
Sn (Miyata-Kosugi, Stille).....
M² = Fe, Ni, Cu, Pd, Rh,.....
X = I, Br, Cl, OSO₂R,.....

Scheme 1.

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