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Title: Pd(II)-Catalyzed Arylation and Intramolecular Amidation of γ -C(sp³)-H Bonds: En Route to Arylheteroarylmethane and Pyrrolidone Ring Annulated Furan/Thiophene Scaffolds

Authors: Parella, R. (/jspui/browse?type=author&value=Parella%2C+R.)
Babu, S.A. (/jspui/browse?type=author&value=Babu%2C+S.A.)

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Abstract: We report the Pd(II)-catalyzed, bidentate directing group (BDG)-assisted arylation and successive arylation/intramolecular amidation of γ -C(sp³)-H bonds. The Pd(II)-catalyzed BDG-assisted C-H activation and functionalization of the β -C(sp³)-H bonds of carboxylic acids are well documented, but only a few reports are available that deal with the BDG-directed functionalization of the γ -C(sp³)-H bonds. Various 3-methylthiophene/furan-2-carboxamides (1a-e) were derived from their corresponding carboxylic acids and bidentate directing groups. These compounds were then used as substrates to investigate the arylation and successive arylation/intramolecular amidation of the γ -C(sp³)-H bonds. The γ -C(sp³)-H arylation arose from the Pd(II)-catalyzed reactions of these compounds with aryl iodides with reaction periods of 4–24 h (except a few reactions which required 36 or 48 h). Notably, these reactions led to the construction of various unsymmetrical diarylmethane scaffolds, such as thiophene/furan-based arylheteroarylmethanes (3–6). Prolonging the reaction period to 48–70 h led to successive γ -C(sp³)-H arylation/intramolecular amidation and the construction of both C–C and C–N bonds. Accordingly, these reactions led to the construction of new classes of pyrrolidone-ring annulated thiophene/furan-based heterocyclic scaffolds (e.g., 4,5-dihydro-6H-thieno[2,3-c]pyrrol-6-ones (8), 4,5-dihydro-6H-furo[2,3-c]pyrrol-6-ones (10), and 1-phenyl-1,2-dihydro-3H-benzo[4,5]thieno[2,3-c]pyrrol-3-ones (12)), and notably, compounds 8, 10, and 12 resemble the skeletons of 3-phenylisoindolin-1-ones.

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