Temperature-Controlled Selective Mono- vs. Di-*ortho*-Arylation for the Synthesis of Arylhydrazine Derivatives

Autorzy

Nurbey Gulia

Jarosław Fornalski

Adrianna Gumienna

Małgorzata Ambroziak

Sławomir Szafert

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Streszczenie

A method of palladium-catalyzed C−H arylation assisted with a 3,4,4-trimethylpyrazol-5-on directing group, selectively providing mono- and di-*ortho*-arylated products is reported. Steric hindrance appearing between the directing group and the already introduced aryl substituent enables control of mono- vs. diarylation selectivity by the temperature of the reaction. Taking advantage of this, a series of monosubstituted and disubstituted derivatives were obtained in good yields. Moreover, unsymmetrical double-arylation in a one-pot procedure was developed to give corresponding products in reasonable yields. Additionally, synthesis and X-ray study of intermediate palladium metallacycles of both, the first and second arylation reactions, were conducted. X-ray structure comparison emphasizes the geometrical differences that were consistent with the observed reactivity. Finally, decarboxylative cleavage of the pyrazolone directing group under mild conditions gave synthetically useful hydrazones. The presented solution opens the alternative synthetic way to such *ortho* arylated derivatives of arylhydrazines.

Słowa kluczowe

arynes, C-H activation, homogeneous catalysis, hydrazones, palladium

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