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Design of a Primary-Amide-Functionalized Highly Efficient and Recyclable Hydrogen-Bond-Title: Donating Heterogeneous Catalyst for the Friedel–Crafts Alkylation of Indoles with  $\beta$ -Nitrostyrenes

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Abstract:

A primary-amide-functionalized metal organic framework, {[Zn2(2-BQBG)(BDC)2]·10H2O}n (1) (in which 2-BQBG = 2,2'-(butane-1,4-diylbis((quinolin-2-ylmethyl)azanediyl))diacetamide and BDC = 1,4-benzenedicarboxylate), has been found to be a highly efficient hydrogen-bond-donating (HBD) heterogeneous catalyst for the Friedel–Crafts alkylation of indole with  $\beta$ -nitrostyrenes under mild reaction conditions (catalyst loading: 3 mol %; reaction conditions: 12 h and 35 °C). The catalyst can be easily separated from the reaction mixture by simple filtration for its reuse in four consecutive cycles with very little loss of activity. More importantly, the one-pot room temperature synthesis of 1 from the self-assembly of Zn(OAc)2·2H2O and 2-BQBG (in CH3OH) and Na2BDC (in H2O) can be easily scaled-up for obtaining multigram quantities in few hours. In order to showcase its versatility, the substrate scope included a variety of substituted indoles and different  $\beta$ -nitrostyrene derivatives forming the desired products in good to high yields. For its catalytic action, a direct proof for the key step in the proposed mechanism, based on the interaction of a primary-amide group in the 2-BQBG ligand with the nitro group of  $\beta$ -nitrostyrene through hydrogen bonding, is provided from the enhancement in fluorescence intensity of 1 upon successive addition of  $\beta$ -nitrostyrene.

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