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Title:	Design and Development of a Heterogeneous Catalyst for the Michael Addition of Malononitrile to 2-Enoylpyridines: Influence of the Primary Amide Decorated Framework on Catalytic Activity and Selectivity
Authors:	Markad, D. (/jspui/browse?type=author&value=Markad%2C+D.) Mandal, S.K. (/jspui/browse?type=author&value=Mandal%2C+S.K.)
Keywords:	Malononitrile Heterogeneous Isostructural analogue
Issue Date:	2019
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Citation:	Inorganic Chemistry, 58(19), pp.12547-12554.
Abstract:	<p>For the Michael addition of malononitrile to 2-enoylpyridines, we report the first heterogeneous catalyst, {[Zn2(2-bpbpg)(fum)2]·4H2O·EtOH}<sub>n</sub> (1) (where 2-bpbpg = N,N'-bis(2-pyridylmethyl)-1,4-diaminobutane-N,N'-diacetamide and where fum = fumarate), which is decorated with primary amide side arms. It is prepared from the self-assembly of starting materials in methanol at room temperature (27 °C). Using 3 mol % of 1, greater than 99% conversion of substrates to the desired product is achieved within 1 h at 27 °C. Moreover, the catalyst is recyclable up to five consecutive cycles without significant loss of activity and structural integrity. In order to show the uniqueness of 1, the reaction under the same conditions was catalyzed by a fully pyridyl-based (i.e., having no primary amide group) and isostructural analogue, {[Zn2(tpbn)(fum)2]·6H2O}<sub>n</sub> (2) (where tpbn = N',N',N'',N''-tetrakis(pyridin-2-ylmethyl)butane-1,4-diamine), but resulting only in 7% conversion. This demonstrates the selective catalytic activity of 1 over 2 due to the presence of the primary amide side arms, where it acts as a bifunctional catalyst through the excellent hydrogen bond donating (HBD) ability in this reaction. For providing an insight into its mechanism of action involving a cyclic seven-membered hydrogen bonding motif, the reaction was performed with freshly synthesized (E)-chalcone, 3-enoylpyridine, and 4-enoylpyridine instead of 2-enoylpyridine under the same conditions. In the case of (E)-chalcone no product formation was observed, whereas for 3-enoylpyridine and 4-enoylpyridine the conversions were only 29% and 25%, respectively. Both 1 and 2 were fully characterized by infrared spectroscopy, elemental analysis, thermogravimetric analysis, and single-crystal and powder X-ray diffraction.</p>
Description:	Only IISERM authors are available in the order.
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