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Greener solvent systems in synthesis and polymer processing

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Solvents can contribute significantly to waste and hazard associated with both chemical synthesis and materials preparation. Solvent alternatives, such as water or supercritical CO2, have been proposed (and some expound the greenness of solvent-free processes), but there are applications that remain challenging. These include organic synthetic procedures requiring dipolar aprotic solvents and polymer dissolution and processing. It is often difficult to identify alternatives to traditional dipolar aprotic solvents such as DMF, NMP or DMAc, yet these are associated with reproductive hazards and may add significantly to process mass intensity, due to the downstream processing required. Many of the replacements suggested are likely to exhibit similar characteristics when fully tested and legislation in Europe will make it increasingly difficult to use such solvents in production-alternatives are actively sought. Here we describe both a methodology for "intelligent selection of greener solvents", focused on identification of replacements for traditional dipolar aprotic solvents (e.g. in SNAr reactions) and organic solvent/ionic liquid combinations for processing of renewable polymers (e.g. cellulose) in composite materials production. In the former, we demonstrate a systematic approach to identifying solvent alternatives for specific applications and, in the latter, how understanding intermolecular interactions and the mechanism of dissolution can lead to identification of the "best" system, with respect to safety and production of materials with specific properties. The approaches described here are applicable in a wide range of solvent applications and may provide opportunities in many areas, e.g. the production of greener inks and coatings, or identification of solvents for use in cleaning operations.

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Chemo selective reactions of Diaminomaleonitrile (DAMN) in water

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Diaminomaleonitrile (DAMN) is one of the most versatile reagents in Organic Chemistry. It has been used as a precursor for producing nucleotides and for synthesizing a wide variety of heterocyclic compounds including purines, pyrimidines, pyrazines (some which are widely employed in the fluorescent dye industry), imidazoles, biphenylenes, porphyrazines (which have great potential in optical sensor technology) and diimines that are used as catalysts. The reaction of DAMN with aromatic aldehydes is widely known to produce monoimines which are important as synthetic intermediates in the synthesis of drugs, conjugated linear polymers and in the thermostable optical material industry. Most reaction methods described involve the use methanol as a solvent without catalysts, but ethanol and acid catalysis are required if the aldehyde bears a strong electron-withdrawing group. Earlier in our laboratory we have sought to apply green chemistry principles to the reaction between DAMN and various aromatic aldehydes employing either water as solvent without catalysts or under solvent-free conditions, thus obtaining the respective monoimines. The reactions carried out in water were also shown to be chemo-selective. The fact that water was used as solvent in this type of reaction makes this a cleaner, more efficient and attractive method for preparing this type of substances.

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