

Past and Present Research Systems of Green Chemistry

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Click chemistry: Mediated synthesis of bio-inspired functionalized ionic liquids

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Task-Specific Ionic Liquids (TSILs) or functionalized ionic liquids are a unique subclass of conventional ionic liquids that incorporate an ion-tethered functionality tailored for specific applications ranging from organic synthesis and metal extraction to material and pharmaceutical sciences. The introductions of functional groups on cations and/or anions allow for marked changes of the physicochemical properties of ionic liquids and convey a particular reactivity pattern to them. Since functional groups on TSILs are generally heteroatom-based, the challenge is to develop highly efficient, atom-economical and orthogonal methodologies, such as "click" chemistry. Underlying these synthetic goals is the additional desire to develop scalable reactions that fall within the philosophical realm of green chemistry for the synthesis of ionic liquids. In addition, it has been a challenge to design imidazolium-type ionic liquids (the most common IL class) that incorporate progressively more lipophilic structural elements while maintaining melting points below room temperature. The T_m values of these ILs increases significantly once an appended N-alkyl group exceeds seven carbons in length. To further develop task-specific ionic liquids, the exploitation of robust, efficient and orthogonal "click" reactions is desirable. The author will discuss about the "click" chemistry-mediated syntheses of bio-inspired functionalized ionic liquids. The synthetic methodology will be discussed, as well as the impact of these structural modifications upon ionic liquid melting points and computational insights into their basis.

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Acid-base catalysis for valorization of glycerol and lactic acid

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Glycerol (GL) and lactic acid (LA) are two important bio-derivative platform molecules, whose potential as renewable feedstock for energy and chemicals production have been the subject of many green chemistry research in recent years. This presentation intends to show the richness of chemicals production from both GL and LA, featuring the key roles of heterogeneous catalysts for achieving high-selectivity towards specific products. Results from acid-base catalysis research in our laboratory are then presented to understand the requirement for surface acidity and basicity for the gas-phase GL dehydration to produce acrolein (AC) and the gas-phase LA dehydration to acrylic acid (AA). Catalytic evaluation under identical conditions of many solid catalysts with widely changed acid-base property demonstrates that the strongly acidic surface sites on the dry catalyst, with Hammett acidity function (H_0) in $-8.2 < H_0 \leq -3.0$, are most effective for offering high AC selectivity (60%) in the GL dehydration reaction but coexisting basic sites on the catalyst surface are detrimental to AC formation. On the other hand, the catalytic LA dehydration reaction data point to a mechanism of cooperative acid-base catalysis for the selective formation of AA. A ZSM-5 based catalyst is identified to show the up to date highest AA selectivity (ca. 80%) and yield (70%), and longest catalytic durability (80 h) in the LA dehydration reaction. Results from reaction kinetic studies will be presented to discuss the effects of reaction variables on reactant activation and product selectivity.

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