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First depth-resolved beta-NMR measurements of 1-ethyl-3-methylimidazolium acetate

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Over the past decade, we have been using beta-detected NMR to examine the properties of amorphous materials. While this has typically focused on polymers,[1] we have recently been interested in ionic liquids (ILs). ILs are binary mixtures: they are composed of two oppositely charged molecular species. They are also liquid at room temperature. Their properties, determined by strong electrostatic forces, make them attractive candidates for the development of next-generation battery technology.

The long-range forces between ions also affect their dynamics, one of our primary interests in amorphous materials. This makes ILs a fascinating comparison to the relatively well-understood case of polymers. As with polymers, many ILs are extremely resistant to crystallization and will instead vitrify upon cooling. In our prior work, we showed that β-NMR was a good probe of bulk IL dynamics and dynamic heterogeneity.[2] In our present experiments, we turn to the question of how the surface modifies these properties, presenting the first depth-resolved β-NMR measurements in 1-ethyl-3-methylimidazolium acetate. This interfacial region is important for understanding how constrained dimensionality affects dynamics, which in turn may affect this IL's effectiveness as a potential electrolyte in batteries or capacitors.

We will show that both the surface and the glass transition have large effects on molecular dynamics, which in many aspects differs greatly from our expectations. In the glassy phase, the surface dynamics appear to be simultaneously faster (i.e., liquid-like) and yet still heterogeneous (i.e., glass-like), an apparent departure from our understanding of "normal" behaviour. Additionally, relaxation becomes faster below the glass transition temperature.

[1] McKenzie, I. et al. J. Chem. Phys. 156, 084903 (2022).
[2] Fujimoto, D. et al. Chem. Mater. 31, 9346–9353 (2019).

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