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Optimal Sample Formulations for DNP SENS: The Importance of Radical-Surface Interactions

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Abstract:

The efficacy of dynamic nuclear polarization (DNP) surface-enhanced NMR spectroscopy (SENS) is reviewed for alumina, silica, and ordered mesoporous carbon (OMC) materials, with vastly different surface areas, as a function of the biradical concentration. Importantly, our studies show that the use of a "one-size-fits-all" biradical concentration should be avoided when performing DNP SENS experiments and instead an optimal concentration should be selected as appropriate for the type of material studied as well as its surface area. In general, materials with greater surface areas require higher radical concentrations for best possible DNP performance. This result is explained with the use of a thermodynamic model wherein radical-surface interactions are expected to lead to an increase in the local concentration of the polarizing agent at the surface. We also show, using plane-wave density functional theory calculations, that weak radical-surface interactions are the cause of the poor performance of DNP SENS for carbonaceous materials.

Keywords: dynamic nuclear polarization, solid-state NMR, surface-enhanced NMR, mesoporous materials, carbonaceous materials