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Identifying prenucleation species in concentrated, alkaline sodium aluminate solutions to improve aluminum processing efficiency

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The Bayer process is used to refine bauxite ore and produce alumina (Al_2O_3). In this process, bauxite is heated and dissolved in aqueous sodium hydroxide (NaOH), forming predominantly tetrahedrally coordinated aluminate ($Al(OH)_4^-$). The solution is then cooled and seeded to precipitate gibbsite, ($Al(OH)_3$), which exhibits octahedrally coordinated Al. Gibbsite precipitation is slow and inefficient, and the mechanism by which the tetrahedrally coordinated solution species precipitate as octahedrally coordinated gibbsite is unknown. This is because it has not been possible to isolate and identify the intermediate solution species in this process. Here, we prepared synthetic solutions representative of those in the Bayer process that can be poised in metastable states. We are able to control the timelines for precipitation by the relative Al^{3+}:NaOH concentrations. By determining the Al^{3+} speciation and local solution structures in these controlled metastable solutions, their role in the formation of prenucleation species and gibbsite precipitation can be established. We provide evidence that only two Al^{3+} solution species can be identified across the various solution compositions, and that these species are key to the metastability of the system. Rather than changes in speciation, the interplay between hydrogen-bonding, proton transfer reactions, and ion pairing drive the dynamics of gibbsite precipitation in these concentrated alkaline electrolytes. The results have far reaching implications beyond aluminum processing, including water-in-salt electrolytes and the processing of the nation's high level radioactive waste.