Abstract

For the transmutation of long-lived radionuclides and reduction in nuclear waste management timelines, trivalent lanthanide/actinide separations are necessary. Previous research has established that one of the most effective separation approaches uses soft N-donor ligands to selectively interact with the trivalent actinides over the trivalent lanthanides. Several of the currently and previously studied ligands fall in the polyamino polycarboxylate class of complexants. DOTA, 1,4,7,10-tetraazacyclododecane-\(N, N', N'', N''''\)-tetraacetic acid, is a structurally rigid polyaminopolycarboxylate ligand with a heterocyclic core containing four nitrogen donors. Trivalent lanthanide-DOTA complex stability is well characterized under biological conditions, courtesy the application of gadolinium-DOTA contrast reagents in medical imaging. There have been no reports of trivalent actinide bonding patterns with DOTA. This reagent has not been considered for separations applications previously due to the slow kinetics of complexation as it is known that the prearrangement of the donor atoms promotes slow reaction kinetics.

This work highlights that the kinetic barriers associated with DOTA complexation can be significantly lowered through work at elevated temperatures. The results indicate that an operating temperature of 60°C provides enough energy to the system to overcome the kinetic barrier. By operating at this elevated temperature, preliminary Eu/Am separation studies have been done with DOTA as the trivalent actinide holdback reagent. The results indicate the predicted selectivity of DOTA for the trivalent actinides is present. Furthermore, the data offers an alternative interpretation of the rate limiting step in the lanthanide-DOTA complex formation mechanism.