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Comparison of the structural, electrochemical, and spectroscopic properties of two cryptates of trivalent uranium†

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We describe a study of the influence of cryptand density on the structural, electronic, and electrochemical properties of U^{III}-containing cryptates. Two cryptands (2.2.2 and 2.2.1) are reported. The cryptand with the smaller density leads to negative electrochemical potentials and shorter bond lengths that are consistent with a better fit for U^{III} than the larger cryptand. These studies provide insight into the rational design of cryptand-based ligands for trivalent uranium.

Introduction

The accumulation of depleted uranium as a waste product from uranium enrichment encourages the development of research focused on uranium coordination chemistry.^{1–3} Reported studies in this area provide insight into potential uses of depleted uranium and fundamental knowledge of uranium coordination chemistry involved in applications such as actinide separations in nuclear waste.^{4–8} Within this context, there have been widespread reports of redox-active and redox-inactive ligands used to form complexes of U^{III},^{9–16} and one of those ligands, 2.2.2-cryptand, has been widely used to encapsulate metal ions, including U^{III}.^{17–19} Further, the coordination chemistry of U^{III}, Np^{III}, and Pu^{III} with 2.2.2-cryptand has been reported recently,²⁰ expanding cryptand chemistry into the actinides. The thermodynamic and kinetic stability of a cryptate is governed by the cavity size and denticity of the coordinated cryptand as well as the ionic radii and oxidation state of the given metal ion.^{21–24} For example, 2.2.1-cryptand fits better with Eu^{III}, and 2.2.2-cryptand fits better with Eu^{II}; moreover, the Gibbs free energy of Eu^{III}(2.2.1-cryptand) is 1.8 times greater than that of Eu^{III}(2.2.2-cryptand), and the dissociation constant of Eu^{III}(2.2.1-cryptand) is 2.7 × 10⁵ times smaller than that of Eu^{III}(2.2.2-cryptand).²⁴ Because the size of U^{III} (1.025 Å) is closer to the size of Eu^{III} (0.947 Å)

than Eu^{II} (1.17 Å)²⁵ and the charge density of U^{III} is closer to Eu^{III} than Eu^{II}, we suspected that 2.2.1-cryptand would be a good ligand for U^{III}. Additional support for this suspicion is in our recent report that the flexible counterpart of 2.2.2-cryptand, tris[2-(2-methoxyethoxy)ethyl]amine (TDA-1), forms U^{III}-containing complexes with smaller coordination numbers (nine) compared to all reported 2.2.2-cryptates (with coordination numbers of ten).²⁶ This report of an acyclic ligand implies that trivalent uranium can be encapsulated by cryptands with smaller denticities than that of 2.2.2-cryptand. Therefore, based on the studies of Eu^{III} cryptand chemistry and U^{III} chemistry with acyclic TDA-1, we hypothesized that 2.2.1-cryptand is a better match for U^{III} than 2.2.2-cryptand. Here, we report U^{III}-containing cryptates of 2.2.2- and 2.2.1-cryptand (Fig. 1) to investigate how ligand denticity affects the structural, spectroscopic, and electrochemical properties of U^{III}.

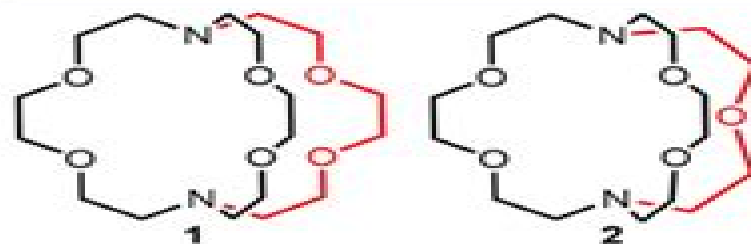


Fig. 1 2.2.2-Cryptand, 1, and 2.2.1-cryptand, 2, that were studied with U^{III}. The red color highlights the difference between the two ligands.

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Chao Zhang



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