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

D. Nuwangi Kulasekara," Matthew D. Bailey, 😏 "Cassandra L. Ward and Matthew J. Allen 3 **

We describe a study of the influence of cryptand denticity on the structural, electronic, and electro-Received 22nd February 2024. chemical properties of U^{III}-containing cryptates. Two cryptands (2.2.2 and 2.2.1) are reported. The cryptand with the smaller denticity leads to negative electrochemical potentials and shorter bond lengths that are consistent with a better fit for U^{tt} than the larger cryptand. These studies provide insight into the rational design of expetand-based ligands for trivalent uranium.

Introduction

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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." Within this context, there have been widespread reports of redox-active and redox-inactive ligands used to form complexes of UIII 9-16 and one of those ligands, 2.2.2-cryptand, has been widely used to encapsulate metal ions, including Unit 12-18 Further, the coordination chemistry of U", Np", and Pu" with 2.2.2-cryptand has been reported recently,200 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, 2.2.1cryptand fits better with Eu^{III}, and 2.2.2-cryptand fits better with Eu"; moreover, the Gibbs free energy of Eu" (2.2.1-cryptand) is 1.8 times greater than that of Eu¹¹¹(2.2.2-cryptand), and the dissociation constant of Eu¹¹¹(2.2.1-cryptand) is 2.7 × 10" times smaller than that of Eu¹⁰¹(2.2.2-cryptand).24 Because the size of Ust (1.025 Å) is closer to the size of Eust (0.947 Å) than Eu¹⁰ (1.17 Å)21 and the charge density of U²⁰¹ is closer to Eust than Eust, we suspected that 2.2.1-cryptand would be a good ligand for U". 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 UIII. containing complexes with smaller coordination numbers (nine) compared to all reported 2.2.2-cryptates (with coordination numbers of ten).25 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^{mt} cryptand chemistry and U" chemistry with acyclic TDA-1, we hypothesized that 2.2.1-cryptand is a better match for U" than 2.2.2-cryptand. Here, we report U" 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⁽¹⁾.

Fig. 1 222-Cryptand, 1, and 221-cryptand, 2, that were studied with Ust. The red color highlights the difference between the two ligands.

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Christian Drosten

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