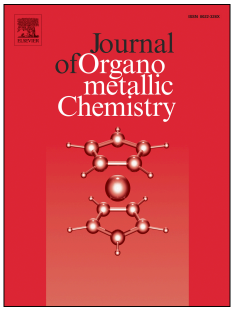

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Generation of Tp*$_2$U(N$_3$) from a Family of New Uranium(III) Alkyl Complexes

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Abstract

A new family of uranium(III) benzyl species supported by bulky hydrotris(3,5-dimethylpyrazolyl)borate (Tp*) ligands has been synthesized and characterized. These derivatives were synthesized by treating Tp*$_2$UI (**1-I**) with various benzylpotassium salts to afford Tp*$_2$U(CH$_2$-*para*-*iso*propylphenyl) (**1-*p*-^{i}Pr**), Tp*$_2$U(CH$_2$-*para*-*tert*-butylphenyl) (**1-*p*-^{t}Bu**), Tp*$_2$U(CH$_2$-*meta*-methoxyphenyl) (**1-*m*-OMe**), and Tp*$_2$U(CH$_2$-*ortho*-picolyl) (**1-*o*-Picolyl**). Along with previously reported Tp*$_2$U(CH$_2$Ph) (**1-CH$_2$Ph**), these uranium alkyl complexes can be treated with an equivalent of SiMe$_3$N$_3$ to yield Tp*$_2$UN$_3$ (**2-N$_3$**), releasing an equivalent of the corresponding trimethylsilylbenzyl compound. All compounds were characterized by multinuclear NMR, IR, and electronic absorption spectroscopies as well as X-ray crystallography.

Introduction

The field of organouranium chemistry has been of interest since the mid-twentieth century, when such compounds were predicted to be useful for isotope separation due to their presumed increased volatility.[1,2] While uranium alkyls did not prove their versatility in this realm, these species have been of fundamental interest for comparison to their transition metal counterparts, with most of the strides being made for uranium(IV) derivatives.[3–7] More recently, new synthetic methodologies have allowed access to tri-, penta-, and hexavalent analogues. [8]

Efforts in our group have focused on the synthesis, characterization and reactivity of organouranium species in the +3[9–11] and +4[12,13] oxidation states. In regard to the former, we have demonstrated that utilizing sterically demanding hydrotris(3,5-dimethylpyrazolyl)borate