

Synthesis of heterometallic compounds with uncommon combinations of elements for oxide nanomaterials using organometallics.

Autorzy

Łukasz John

Piotr Sobota

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Oxide nanomaterials with interesting electronic and magnetic properties have applications including superconductors, magnetic core materials, high-frequency devices, and gas sensors. They can also serve as efficient oxide lattices for luminescent ions. Highly phase-pure BaHfO₃ nanopowders are extremely desirable as matrices for luminescent doping, and barium hafnate is an attractive host lattice for new X-ray phosphors, which are much more effective than the phosphors currently used in radiology and computed tomography.

This wide range of applications creates a strong impetus for novel and inexpensive methods for their synthesis. Classically, mixed-cation oxide ceramics are synthesized according to conventional solid-state reactions involving oxides, carbonates, or nitrates at relatively high temperatures (~1500 °C). These procedures are inefficient and often lead to inhomogeneous by-products and poor control over the stoichiometry and phase purity. Among the new preparation techniques are those involving metal alkoxides and aryloxides with strictly defined metal stoichiometries at the molecular level.

In this Account, we describe several structurally interesting heterometallic alkoxoorganometallic compounds prepared via reactions of organometallic compounds (MMe₃ where M = Al, In, Ga) with group 2 alkoxides having additional protonated hydroxyl group(s) in the alcohol molecule present in the metal coordination sphere. Using lower temperatures than in the conventional solid-state thermal routes involving carbonate/oxide mixtures, we can easily transform these new complexes, with rarely found combinations of metallic precursors (Ba/In, Sr/Al, and Ba/Ga), into highly pure binary oxide materials that can be used, in a similar manner to perovskites and spinels, as host matrices for various lanthanide ions.

Furthermore, our studies on titanium, zirconium, and hafnium metallocenes showed them to be attractive and cheap precursors for an extensive range of novel molecular and supramolecular materials. This unique synthetic method comprises elimination of the cyclopentadienyl ring (as CpH) from Cp_2MCl_2 ($\text{M} = \text{Ti, Zr, Hf}$) in the presence of $\text{M}'(\text{OR})_2$ ($\text{M}' = \text{Ca, Sr, Ba}$; $\text{ROH} = \text{CH}_3\text{OCH}_2\text{CH}_2\text{OH}$), in an alcohol as a proton source. The resulting compounds are suitable for obtaining highly phase-pure perovskite-like oxides including BaTiO_3 , BaHfO_3 , SrHfO_3 , etc.

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