Transformation of molecular compounds with Ba(Sr)/Al/Si and Ca(Sr, Ba)/Ti(Zr, Hf)/Si heteroelements as new efficient route to metal silicate materials.

Autorzy

Rafał M. Petrus

Anna Drąg-Jarząbek

Józef Utko

Tadeusz Lis

Piotr Sobota

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Streszczenie

Silicon-based moieties were anchored in heterometallic alkoxide platforms to obtain molecular clusters based on the M–O–M′–O–Si motif for the generation of mixed-metal silicate materials. Single-source molecular precursors with structures $[M\{(\mu\text{-ddbfo})_2Al(OSiPh_3)_2\}_2]$, where dbbfoH = 2,3-dihydro-2,2-dimethylbenzofuran-7-ol and M = Ba (**1**), Sr (**2**), $[Sr_4Ti_2(\mu_6\text{-O})(\mu_3,\eta^2\text{-}OCH_2CH_2OMe)_8(\eta\text{-}OCH_2CH_2OMe)_2(OSiPh_3)_4]$ (**3**), $[Sr_4M'_2(\mu_4\text{-OH})(\mu_3,\eta^2\text{-}OCH_2CH_2OMe)_4(\mu,\eta^2\text{-}OCH_2CH_2OMe)_6(\mu\text{-X})(OSiPh_3)_4]$ (**4**, with M′ = Zr and X = Cl;**5** with M′ = Hf and X = OH), $[Sr_3Hf_2(\mu_5\text{-O})(\mu_3,\eta^2\text{-}OCH_2CH_2OMe)_4(OCH_2CH_2OMe)_4(OSiPh_3)_4]$ (**6**), and $[Ca_2M'_2(\mu_3\text{-OH})_2(\mu,\eta^2\text{-}OCH_2CH_2OMe)_4(\eta^2\text{-}HOCH_2CH_2OMe)_2(\eta\text{-}OCH_2CH_2OMe)_2(OSiPh_3)_4]$ for M′ = Zr (**7**), Hf (**8**), were prepared by substitution of methyl groups, using Ph_3SiOH or chloride atoms with $KOSiPh_3$, in $[M\{(\mu\text{-ddbfo})_2AlMe_2\}_2]$ (M = Sr, Ba) or $[M_4M'_2(\mu_6\text{-O})(\mu_3,\eta^2\text{-}OCH_2CH_2OMe)_8(OCH_2CH_2OMe)_2(HOCH_2CH_2OMe)_xCl_4]$ (M = Ca, Sr, Ba, M′ = Ti, Zr, Hf and *x* = 0, 4). The precursors were characterized by elemental analysis, NMR spectroscopy, and single-crystal X-ray structural analysis. Thermal decomposition of compounds **1–8** at 1000 °C led to formation of ceramic materials that consisted of mixed-metal oxide nanocrystallites embedded in an amorphous SiO_2 matrix. Compounds **1** and **2** decomposed to $BaAl_2Si_2O_8$ and $SrAl_2Si_2O_8$ aluminosilicates, **4** gave a mixture of $SrSiO_3$, ZrO_2, $SrZrO_3$ and $Sr_7ZrSi_6O_{21}$, **5** and **6** gave mixtures of various compounds, including $SrHfO_3$, $Hf_{0.96}Si_{0.04}O_2$, and SiO_2, **7** lead to $CaZrO_3$, ZrO_2 and $Ca_3ZrSi_2O_9$ and **8** gave a HfO_2, $Ca_2HfSi_4O_{12}$ and SiO_2 phase. The thermolysis of compound **3** gave SiO_2 particles of average size 40–90 nm, which contained spherical $SrTiO_3$, $SrSiO_3$ and $Sr_2TiSi_2O_8$ nanocrystallites of diameter 5–16 nm.