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Structural - compositional relations in synthetic murataite-structure compounds

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Among the host phases for immobilization of actinide elements occurred in high level waste (HLW) murataite phases are considered as very perspective due to their high chemical durability, radiation resistance, and high capacity with respect to actinide, rare earth and corrosion elements. Rare mineral murataite (only two samples from USA and Russia are known) contains major sodium, zinc, yttrium, titanium and niobium. Synthetic murataites found in ceramics designed for immobilization of HLW from Savannah River Plant, USA, and "Mayak", Russia, contained Ce-group lanthanides and uranium. We have found murataites with various multiplicity (3x, 5x, 7x, 8x) of the fluorite cell and suggested that these phases belong to a series of compounds where pyrochlore, Py (2x) and murataite, M (3x) are the end-members [Urusov et al., Dokl. Russ. Ac. Sci, 2005, 401, 226]. The phases structure is built of modules of pyrochlore and murataite: Py+M = 5x; M+Py+M = 8x, and Py+M+Py = 7x. Hence, their capability to incorporate actinides and lanthanides should increase in a row: 3x $<$ 8x $<$ 5x $<$ 7x. To confirm this suggestion ceramics were fabricated by melting of oxides at 1400-1500 ^{o}C for 1-3 hours. The samples had bulk composition (wt.%): 5 Al_2O_3, 10 CaO, 55 TiO_2, 10 MnO, 5 Fe_2O_3, 5 ZrO_2, 10 WO (WO = La_2O_3, CeO_2, Nd_2O_3, Gd_2O_3, Y_2O_3, ZrO_2, ThO_2, UO_2). They are examined with XRD and electron microscopy (SEM/EDS, TEM). Most of them are composed of murataite and minor perovskite and crichtonite. Murataites with five-, eight-, and three-fold fluorite cell are observed in core, intermediate zone, and rim of the grains, respectively. Rare earths and actinides concentrations are reduced in a row: M5x $>$ M8x $>$ M3x. The research was supported by the US DOE (project RUC2-20009-MO-04) and the Russian Foundation for Basic Research (project 05-05-08000).