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## Immobilisation of cesium and divalent metals into single-phase stuffed tridymite-based ceramics

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The mixed phosphates with high caesium concentration of the compositions  $\text{CsMePO}_4$  (Me – divalent metal with a tetrahedral coordination) adopting a stuffed  $\beta\text{-SiO}_2$  tridymite structure are studied as perspective materials for a  $^{137}\text{Cs}$   $\gamma$ -radiation sources to be used in medic applications. For the purpose of  $^{137}\text{Cs}$  source production from commercial radiocaesium liquid wastes (nitrate caesium solutions containing Me impurities) it is necessary to know the possibilities of single-phase composition formation in the systems  $\text{CsMe}^{1-x}\text{MexPO}_4$  with different Me cations.

In this work we have studied phase formation regularities and thermal behavior of mixed complex phosphates of the  $\text{CsMg}_{1-x}\text{MexPO}_4$  (Me = Mn, Co, Ni, Zn, Cu) systems. The samples were synthesized by precipitation method. The aqueous solution of phosphoric acid taken in accordance with the stoichiometry sample was added to stoichiometric mixture of caesium and metal (Me) nitrate or chloride solutions. The reaction mixture was dried at 353 K and thermally treated at 873 and 973 K. All the thermal treatment stages were alternated with careful grinding. The obtained samples were crystalline powders.

X-ray powder diffraction measurements indicated that continuous (Me = Mn, Co, Ni, Zn;  $0 \leq x \leq 1.0$ ) or limited (Me = Cu;  $0 \leq x \leq 0.4$ ) solid solutions of the tridymite structural type were obtained. The results of scanning electron microscopy and electron microprobe analysis monitored the homogeneity of the samples and conformity of their compositions to the theoretical values.

The phase formation regularities and the dependences of the unit cell parameters of solid solutions  $\text{CsMg}_{1-x}\text{MexPO}_4$  on their compositions  $x$  were studied. Three polymorphic forms with similar type of a framework are possible for tridymite structure type phosphates: monoclinic (sp. gr. P21/a) and two orthorhombic (sp. gr. Pn21a and Pnma). For many phosphates with temperature increase the phase transitions  $\text{P21/a} \rightarrow \text{Pn21a} \rightarrow \text{Pnma}$  are observed.

Due to the differential scanning calorimetry results, the phosphates  $\text{CsMePO}_4$  with Me = Co and Zn had two polymorphic transitions in the temperature interval from 423 to 583 K. The phosphate  $\text{CsMgPO}_4$  underwent phase transition at 236 K. The second harmonic generation results showed phosphate crystallization in centrosymmetric (P21/a or Pnma) or non-centrosymmetric (Pn21a) space groups. The transition from Pnma to Pn21a space group was accompanied by absorption band disappearance of valency symmetric vibrations in IR-spectra of phosphates.

A leaching rate of approximately  $10^{-5} \text{ g}\cdot\text{cm}^{-2}\cdot\text{d}^{-1}$  for Cs was determined from Soxhlet leaching of the  $\text{CsMgPO}_4$ .

Thus, the possibility of simultaneous incorporation of caesium and different metals with oxidation degree +2 (which present in commercial radiocaesium liquid wastes) is shown in the tridymite structure with formation of wide or continuous solid solutions. Such tridymite-like phosphates may be a perspective candidate both for radiocaesium immobilisation and caesium isotopic source production instead of soluble CsCl used nowadays. This work was supported by the Russian Foundation for Basic Research (Projects nos. 08-03-00082, 09-03-90731).

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