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Progresses on the structural chemistry of the actinide phosphates

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Data on structure and properties of actinide phosphates are needed for the development of phosphate ceramics envisaged as possible hosts for long-term sequestration of actinides generated by the nuclear fuel cycle. In that aim an extensive study on the crystal structures, thermal stability, and thermodynamic properties on a number of crystalline trivalent and tetravalent actinide phosphates was undertaken.

PuPO_4 with monazite structure was obtained and characterized in terms of thermal stability and thermal expansion. The decomposition of PuPO_4 to Pu_2O_3 was observed at 1573 K. Its high-temperature heat capacity is to be measured up to 1500 K by drop calorimetry.

$\text{M}^{\text{II}}\text{Np}(\text{PO}_4)_2$ ($\text{M}^{\text{II}} = \text{Ca, Sr, Ba}$) were obtained by solid state reaction. The influence of ionic radius of M^{II} ($\text{M}^{\text{II}} = \text{Ca, Sr, Ba}$) on the crystal structure is further discussed. Pure phase of $\text{CaNp}(\text{PO}_4)_2$ with monazite structure (P2/n) was obtained by solid state reaction after several thermal treatments and readjustments in stoichiometry. The lattice parameters are $a = 6.65085(3) \text{ \AA}$, $b = 6.83893(5) \text{ \AA}$, $c = 6.35378(3) \text{ \AA}$, and $\beta = 104.117(9)^\circ$. $\text{SrNp}(\text{PO}_4)_2$ seems to adopt an inedited double-monazite structure with $a = 6.895 \text{ \AA}$, $b = 13.272 \text{ \AA}$, $c = 6.892 \text{ \AA}$, and $\beta = 99.22^\circ$, probably due to the Sr/Np ordering responsible for the non-negligible reflections with $k = 2n + 1$; in this case, the two cations seem too different in size to allow disorder, contrarily with $\text{CaNp}(\text{PO}_4)_2$. Pure $\text{BaNp}(\text{PO}_4)_2$ with monoclinic C2/c was also obtained. Its structure was inferred with the one reported for $\text{RbEu}(\text{SO}_4)_2$, the lattice parameters being $a = 12.69095(13) \text{ \AA}$, $b = 5.36179(6) \text{ \AA}$, $c = 9.40949(11) \text{ \AA}$, and $\beta = 102.516(68)^\circ$. The pattern looks similar with the one previously found for the Th-counterpart.

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