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Thermodynamic behavior of monazite-type La1-xLnxPO4 (Ln = Pr, Nd, Eu Gd) solid solutions

Monazite-type orthophosphates (monoclinic LnPO4; Ln = La – Gd) are promising candidates as potential waste forms for the immobilization of specific nuclear waste streams, such as separated Pu from civilian or military sources unsuitable for further use, or separated minor actinides due to their specific physicochemical properties.
The thermochemical behavior of single phase La1-xLnxPO4 (Ln = Pr, Nd, Eu, Gd) solid solutions has been investigated by high temperature oxide melt solution calorimetry. The experimental enthalpy of mixing, as well as complementary atomistic modelling methods, demonstrate the thermodynamic stability of the solid solutions. Margules interaction parameters were calculated applying both approaches. The obtained values are in good agreement, emphasizing the progress of modern numerical methods. These results are an essential basis for thermodynamic models for the assessment of the long term stability of monazite solid solution ceramics as matrices for the safe immobilization of radionuclides in a geological repository.

Summary

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