Synthesis and characterisation of various monazite solid solution series

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Why monazite?

Monazite type ceramics are considered as potential storage materials for minor actinides from high-level nuclear waste. Natural analogues can form solid solutions incorporating up to 30 w% ThO2 and UO2.

Properties of monazite

- Long term stability
- Chemical durability
- Structural flexibility
- High waste loading
- Low critical temperature of amorphisation

Structure

Powders synthesised via solid state reaction with NH4H2PO4 excess [1]

Micro-sized crystallite aggregates are highly porous and homogeneous (BSE)

Lattice parameters from RT-XRD for six solid solutions showing almost ideal behaviour

Single crystals obtained by flux growth routine using MoO3 and Li2CO3 [2]

Single crystals of (La,Pr)PO4 show no sign of zonal growth in EPMA

LaPO4 phase transformation at 27GPa from monazite (P 2n/n) to post-barite structure (P2221) [5]

Thermodynamics of (La,Pr)PO4

HT drop solution calorimetry on powders indicating a high sensitivity to impurities and an almost ideal solid solution [11]

LT microcalorimetry on single crystals revealing a Schottky-contribution resulting from forbstal electrons [12]

Microstructure

Ceramics produced via cold-isostatic pressing and sintering in two steps (1273 K; 1673 K)

SEM images of pre- (a) and final ceramics (b) show increasing theoretical density from 64 % up to 99.3 %

Average grain size of (La,Pr)PO4 ceramics via intercept method [6] showing a higher grain growth of intermediate compositions

Thermochemical Biharmonic Equations

\[ \Delta H_{\text{fus}} = x \Delta H_{\text{fus},0} + (1-x) \Delta H_{\text{fus},1} \]

\[ C_p = x \rho RT + (1-x) \rho_1 RT_1 \]

\[ \rho = x \rho_0 + (1-x) \rho_1 \]

References

[8] Hikichi et al., 1997, MINERALS, 19, 123-130

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