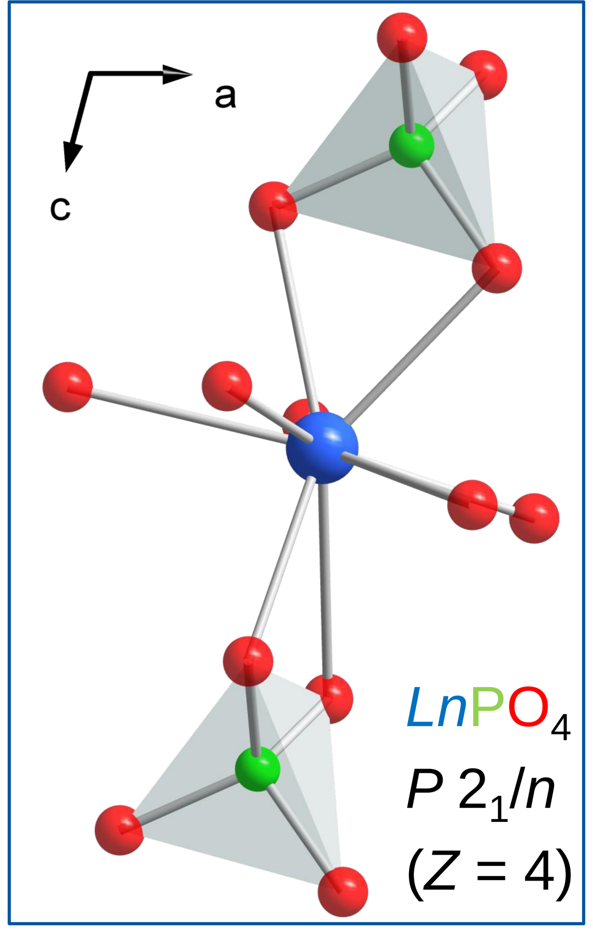
Synthesis and characterisation of various monazite solid solution series

Antje Hirsch[1*], Philip Kegler[2], Igor Alencar[3], Javier Ruiz-Fuertes[3], Anja Thust[3], Johannes Bauer[3], Charlotte Schausten[4], Lars Peters[1], Georg Roth[1]



Why monazite?

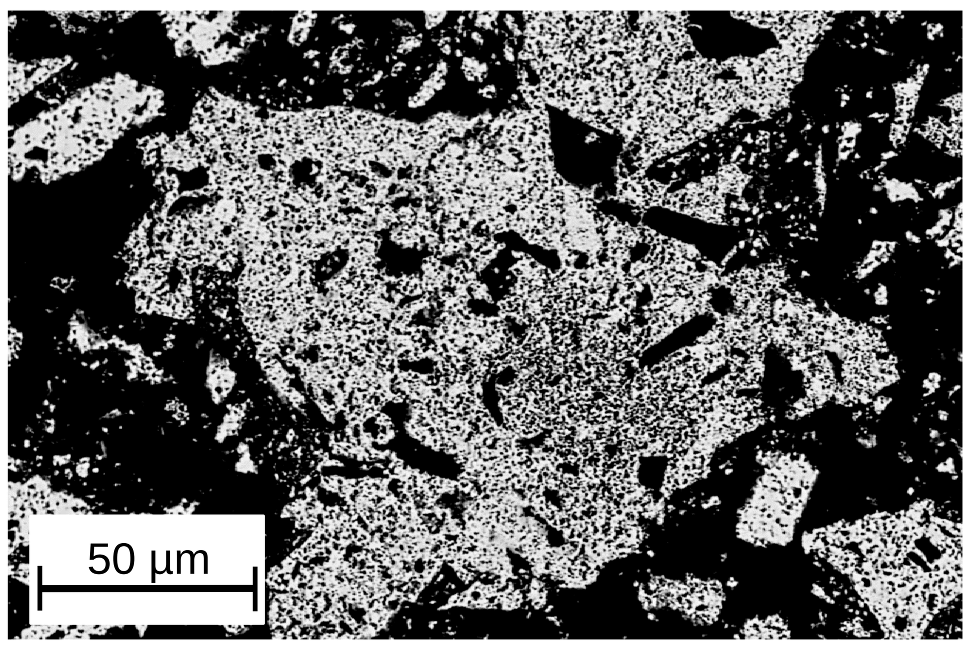
Monazite type ceramics are considered as potential storage materials for minor actinides from high-level nuclear waste. Natural analogues can form solid solutions incorporating up to 30 w% ThO_2 and UO_2.

Properties of monazite

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

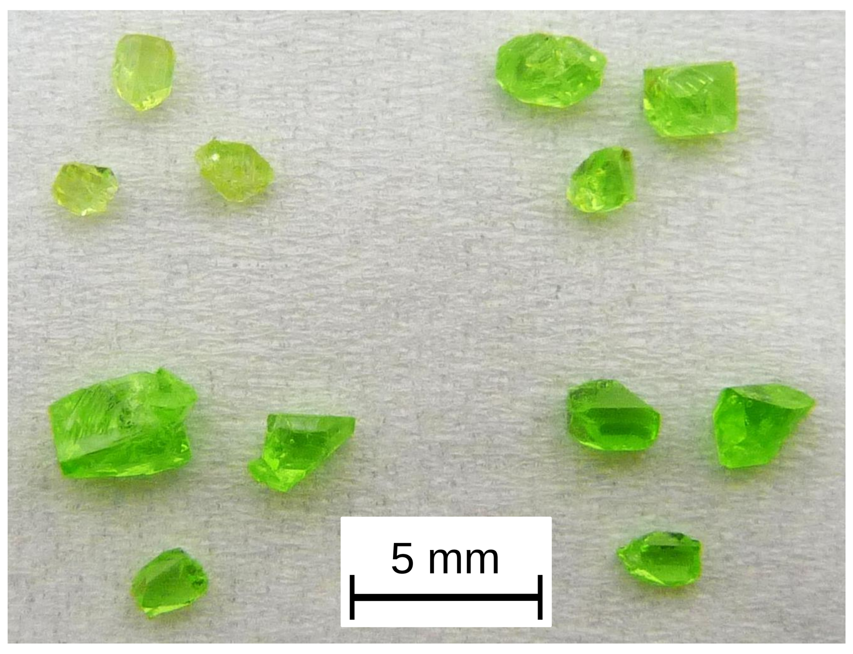
Structure

Powders synthesised via solid state reaction with $NH_4H_2PO_4$ excess [1]

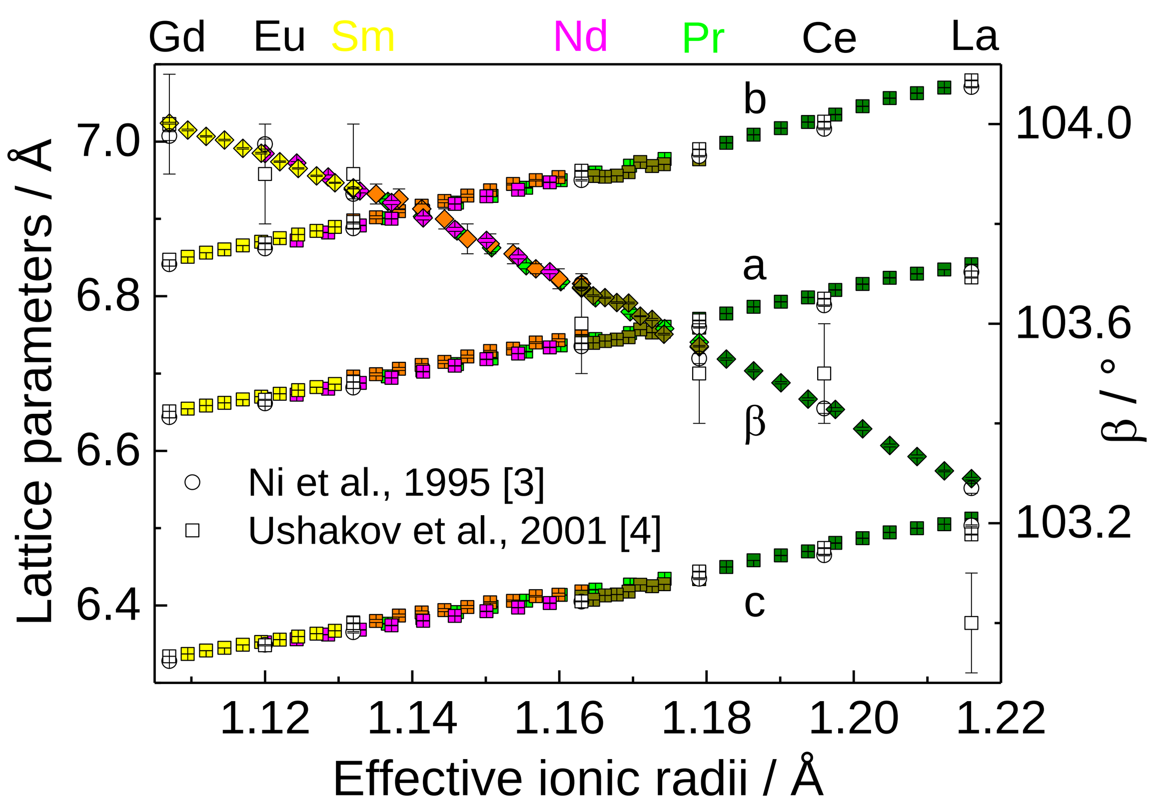


Micro-sized crystallite aggregates are highly porous and **homogeneous** (BSE)

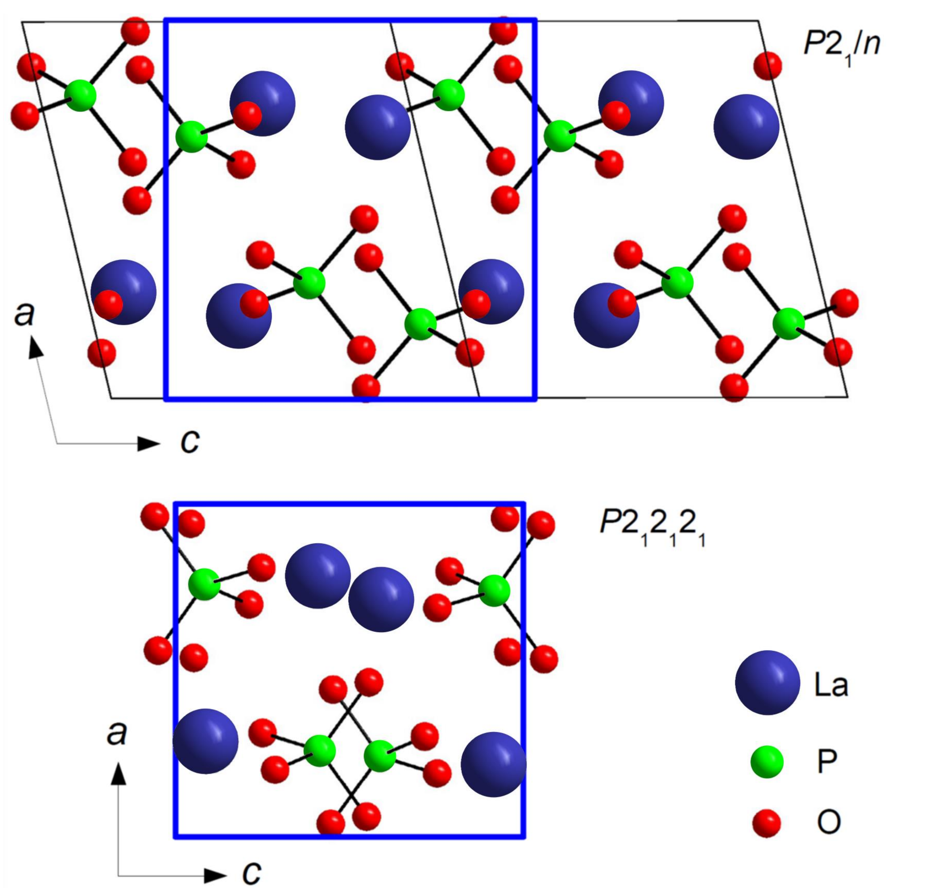
Single crystals obtained by flux growth routine using MoO_3 and Li_2CO_3 [2]



Single crystals of $(La,Pr)PO_4$ show **no** sign of **zonal growth** in EPMA



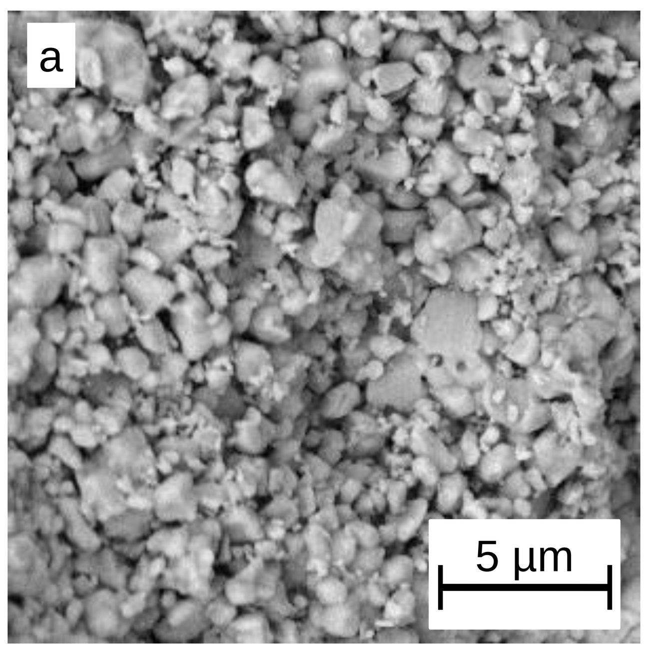
Lattice parameters from RT-XRD for six solid solutions showing **almost ideal behaviour**

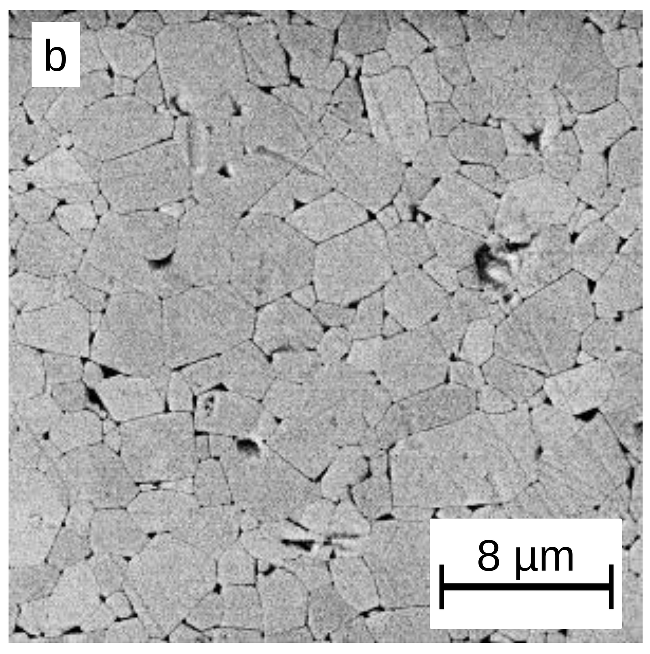


$LaPO_4$ **phase transformation** at 27GPa from monazite ($P\ 2_1/n$) to **post-barite** structure ($P\ 2_12_12_1$) [5]

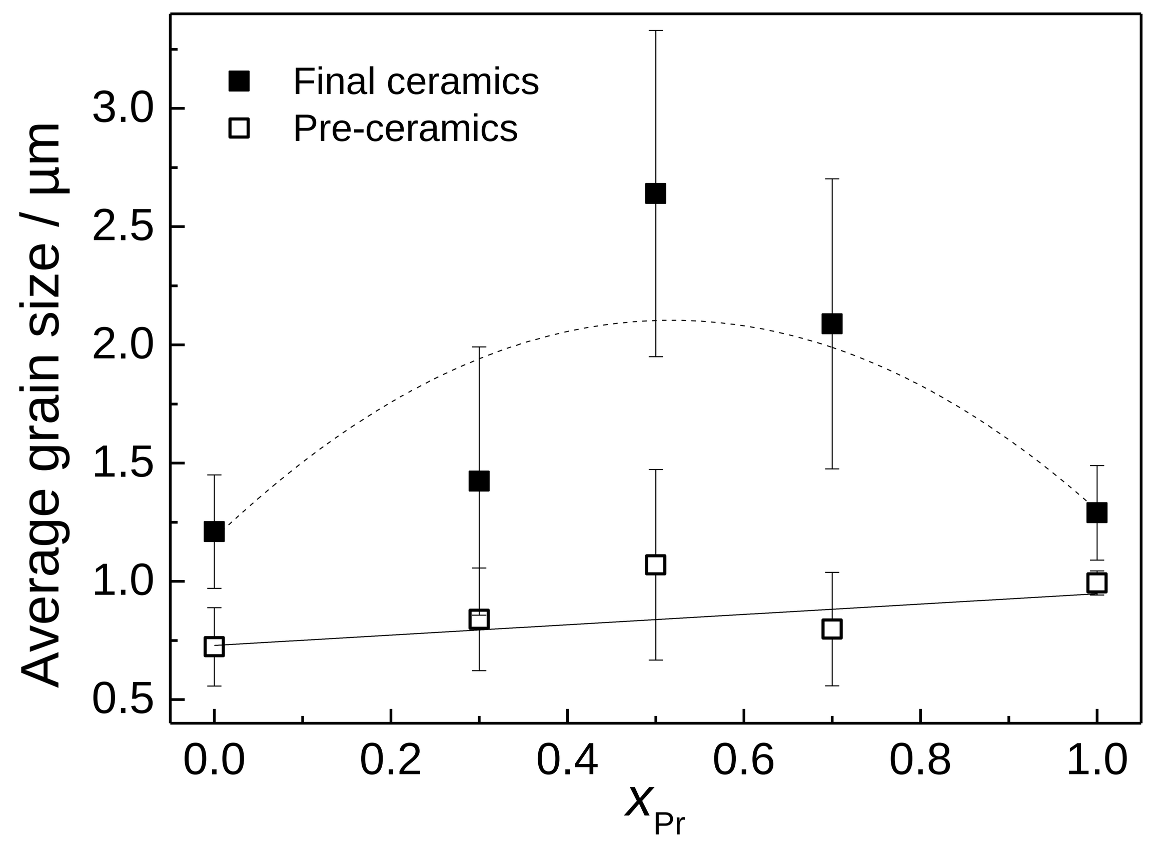
Microstructure

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



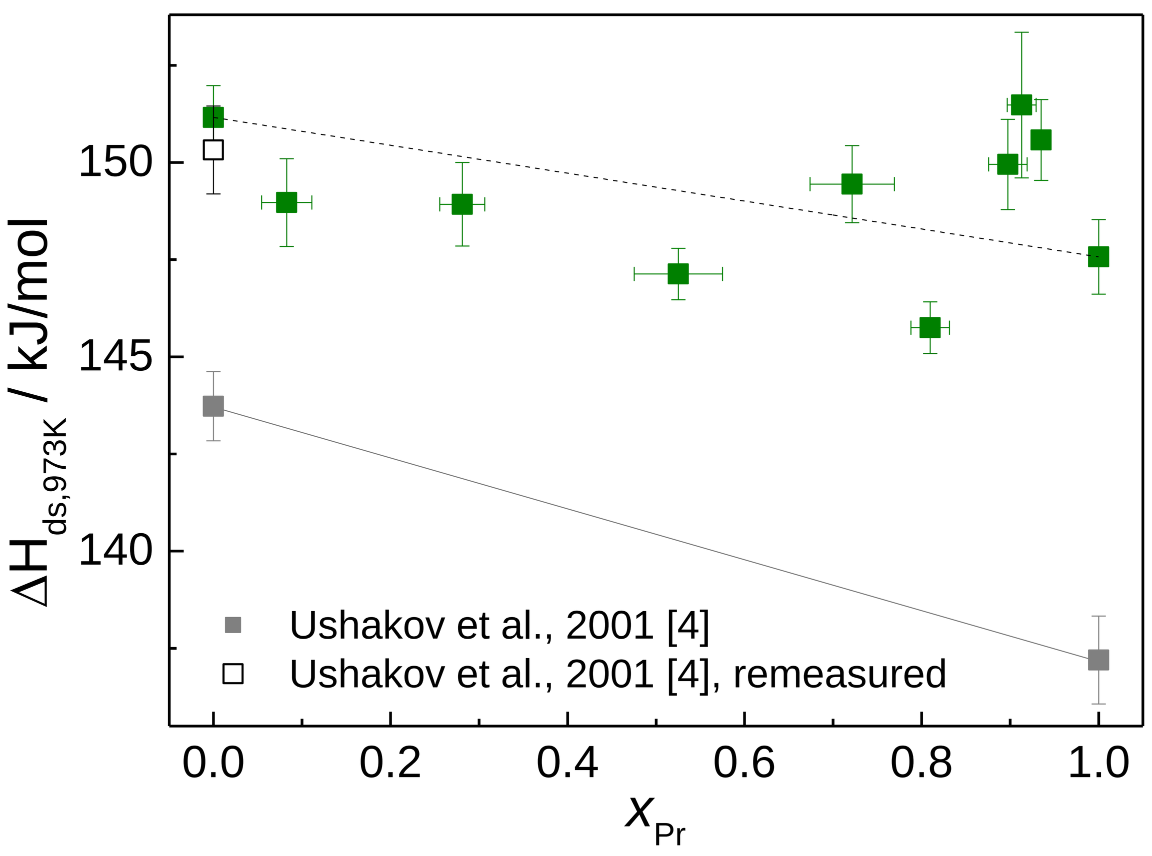


SEM images of *pre-* (a) and *final ceramics* (b) show **increasing** theoretical **density** from 64 % up to 99.3 %

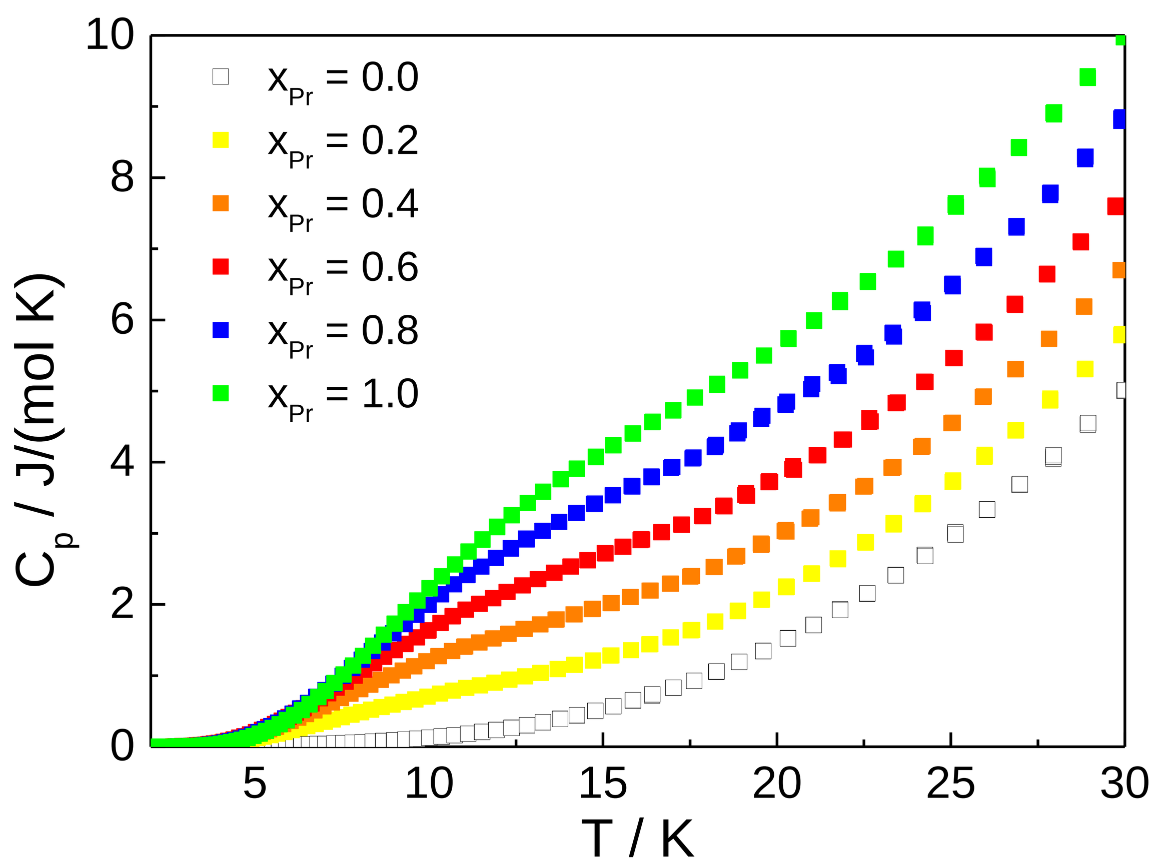


Average grain size of $(La,Pr)PO_4$ ceramics via intercept method [6] showing a **higher grain growth of intermediate compositions**

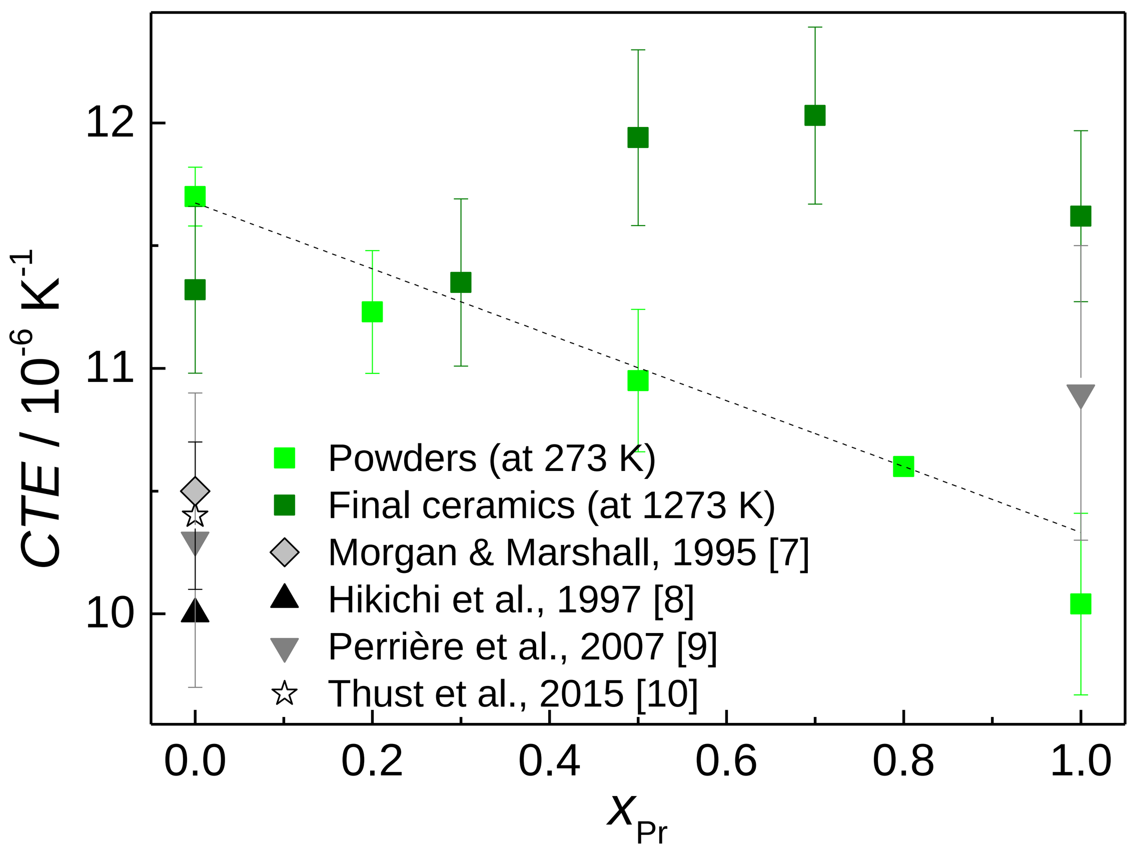
Thermodynamics of $(La,Pr)PO_4$



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 *f*-orbital electrons [12]



Coefficients of thermal expansion for **powders** depend on **composition**, while CTE of **ceramics** depend on **density**

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Institutional Affiliations

[1]RWTH Aachen University, Institute of Crystallography; [2]Institute of Energy and Climate Research (IEK6), Forschungszentrum Jülich; [3]Goethe University Frankfurt am Main, Department of Geoscience, [4]RWTH Aachen University, Institute of Mineral Engineering.

* hirsch@ifk.rwth-aachen.de

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