NOVEL MULTICOMPONENT EQUIATOMIC PYROCHLORES FOR FUTURE THERMAL BARRIER COATINGS

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Multicomponent equiatomic oxides (MEOs), also named high entropy oxides, have attracted great interest in recent years. Analogous to high entropy alloys, they consist of five or more different cations on one or more cation sublattice in approximately same amount. This leads to a big variety of compositions with adjustable material properties and promising versatile characteristics like reversible lithium storage, ionic conductivity, catalytic activity and dielectric and magnetic properties. The high configurational entropy obtained by equimolar mixing of the cations on the cation sublattice of the structure results in improved phase stability at high temperatures and low thermal conductivity due to increased phonon scattering caused by different cation size, mass and valency state. Therefore, the new material class of MEOs are proposed as promising materials for future thermal barrier coatings (TBCs).

However, the development of new TBC materials requires a systematic examination of promising candidate materials, ranging from synthesis to the characterization of thermophysical and thermochemical behavior and investigation of mechanical properties and corrosion protection behavior. Furthermore, up to now the prediction of single-phase materials is very complex.

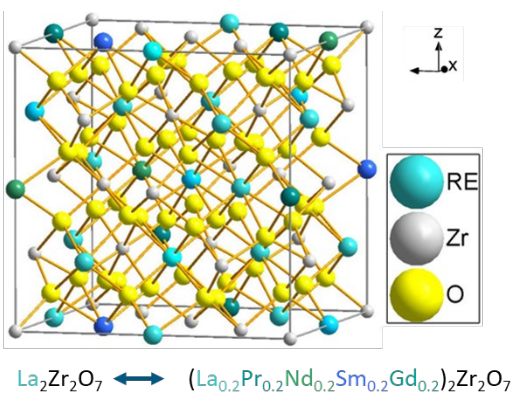

Figure 1: Multicomponent equiatomic pyrochlore oxide.

In this work, multicomponent equiatomic pyrochlore oxides with the general formula $A_2Zr_2O_7$ and 5 different cations on the A-site, as depicted in Figure 1, have been successfully synthesized by using reverse co-precipitation. The composition has been varied systematically regarding ion size and valency to evaluate its influence on crystal structure, phase stability and material properties. For A, transition metals such as La, Nd, Sm, Gd, Y and Zr were used. The chemical and structural characterization was performed using X-ray diffraction with Rietveld refinement, Raman spectroscopy, scanning electron microscopy equipped with electron-dispersive X-ray spectroscopy, electron backscatter diffraction and electron microprobe. Dilatometry, differential scanning calorimetry, high temperature XRD and long-term annealing experiments were conducted to assess the thermal stability as well as thermodynamic and thermophysical characterization of the compounds.

The calcined materials were obtained either in single phase pyrochlore or defective fluorite structure, depending on the cationic radius ratio. For the first time, an easy indicator for the prediction of single phase, multicomponent equiatomic pyrochlore oxides is presented. Important properties for application as TBC material, such as sintering behavior, grain growth rate, coefficients of thermal expansion and heat capacity are promising and show comparable or beneficial behavior compared to the state-of-the-art material 8YSZ.
The results of this work underline the great potential of these materials for future thermal barrier coatings.