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Investigation of the thermal expansion of lithium orthosilicate

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Within the EU, the current grade of advanced ceramic breeder pebbles is composed of a mixture of Li4SiO4 (LOS) and Li2TiO3 (LMT). These pebbles are fabricated at KIT by the melt-based process "KALOS". The addition of LMT is beneficial for two aspects: the mechanical strength of the pebbles is considerably increased and the long-term stability at high temperatures is improved. Nevertheless, the advanced ceramic breeder pebbles generally show a number of defects that decrease the ideally achievable mechanical strength. In this work, high-temperature X-ray diffraction (HT-XRD) experiments will show that the well-known displacive phase transformation of LOS at temperatures between 665 °C and 723 °C is strongly anisotropic. The formation and growth of the defects of the pebbles may thus not be exclusively attributed to thermal stresses during the fabrication of the pebbles but also to this phase transformation. To address this issue, principally two ways exist. Either the intensity of the phase transformation is attenuated, or the phase transformation is shifted to unobjectionably low temperatures. In this study both ways as well as their combination are demonstrated. It will be shown that the solid solution of germanium in the LOS lattice is an effective way of attenuating the phase transformation while the partial substitution of lithium by magnesium shifts the phase transformation to lower temperatures. Furthermore, it is also demonstrated that by adding magnesium the thermal expansion coefficient of LOS can be aligned with that of LMT, so that intrinsic stresses are reduced as additional benefit. HT-XRD, dilatometry as well as differential scanning calorimetry are used for the analysis of different compositions to evaluate the individual effects of the added elements. Eventually, the impact on the pebble strength is demonstrated for selected promising compositions.

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