Growth of $La_2Zr_2O_7$, $La_2Hf_2O_7$ and Lu_3TaO_7 single crystals with high melting point by micro-pulling-down method and their optical properties

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Introduction

Iridium (Ir) and Platinum (Pt) crucibles have been generally used in the micro-pulling-down (μ-PD) method for crystal growth of functional oxide single crystals. However, the melting point (m.p.) of Ir is about 2450°C, and materials research of single crystals by the μ-PD method has basically been limited to materials with m.p. below 2200°C. While Rhenium (Re) crucible enables crystal growths of sesquioxide, it is not currently used due to the rarity of the element. On the other hand, Tungsten (W) and Molybdenum (Mo) with higher m.p. than Ir have been used for crystal growth of sapphire single crystals. W and Mo crucibles have not been used in a wide range of materials due to concerns about reaction with oxides. In this study, we tried to establish growth technique of oxide single crystals with over 2200°C using W and Mo crucibles, and material research in high temperature range were performed for $La_2Zr_2O_7$ (m.p.: 2283°C), $La_2Hf_2O_7$ (m.p.: 2418°C) and Lu_3TaO_7 (m.p.: 2380°C) single crystals.

Experimental

ZrO_2, HfO_2, Ta_2O_5 (>3N), La_2O_3, CeO_2, Eu_2O_3, and Er_2O_3 (>4N), powders were mixed as nominal compositions of $La_2Zr_2O_7$, $La_2Hf_2O_7$ and Lu_3TaO_7, and mixed powders were sintered at 1600°C for 24 hours in air. In addition, sintered powders of specimens including dopant (Ce, Eu or Er) were also prepared. The sintered powder was set in W or Mo crucibles with a die at the bottom, and the crucible and deoxygenated ZrO_2 insulators were set in the center of a high-frequency induction coil. Deoxygenated ZrO_2 insulators were prepared by sintering the commercial ZrO_2 insulators at 2000°C in Ar using a carbon electrical furnace. The crucible was heated up to the m.p. of the target material, and single crystal was grown using a W rod as a seed. Phase identifications and crystallinity evaluations of obtained crystals were performed by measurements of powder X-ray diffraction (XRD), Laue camera, pole figure and X-ray rocking curve (XRC). Optical and scintillation properties of crystals with dopant were also evaluated.

Results and discussions

Crystal growth of $La_2Zr_2O_7$ single crystals were grown by the μ-PD method using Mo or W crucibles. While the Mo crucible deteriorated after crystal growth, W crucible could be used stable without the deterioration. In addition, deoxygenated ZrO_2 insulators could suppress oxidation of W crucible, and $La_2Zr_2O_7$ single crystal could be obtained (Fig.1). $La_2Hf_2O_7$ and Lu_3TaO_7 single crystals were also able to be grown under the similar conditions.[1] After post-annealing in air, grown crystals indicated high transparency. Eu or Er-doped $La_2Zr_2O_7$ and $La_2Hf_2O_7$ single crystals indicated emission light originating from dopants. Details of crystal growths and optical properties will be reported.

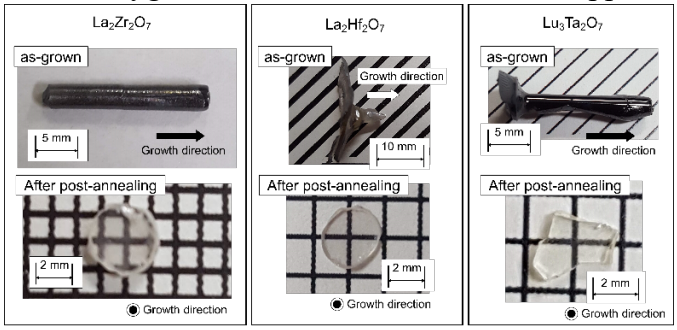

Fig. 1 As-grown $La_2Zr_2O_7$, $La_2Hf_2O_7$ and Lu_3TaO_7 single crystals grown by μ-PD method using W crucible and polished specimens after post-annealing.

References

[1] T. Suda, Y. Yokota, et al., Crystal growth of $La_2Hf_2O_7$ by micro-pulling-down method using W crucible. J. Cryst. Growth. 2022;583:126547.