Phase Formation, Crystal Growth, Crystal Structure and Piezoelectric Properties of $Ca_3TaAl_3Si_2O_{14}$ Single Crystal

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$Ca_3TaGa_3Si_2O_{14}$ (CTGS) single crystal with the langasite-type structure has been investigated as a piezoelectric material due to the high piezoelectric constant and electromechanical coupling factor at high temperature. In addition, the stable temperature coefficient of frequency (TCF) around room temperature of the CTGS is comparable to the quartz and it can be expected to be applied for the next-generation oscillator with small size and low power consumption. We have developed Al doped CTGS, $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$, (CTGAS) single crystals [1] and their piezoelectric properties were investigated [2,3]. The piezoelectric constants d_{11} and electromechanical coupling factor k_{12} were systematically increased with increasing Al concentration. However, the $Ca_3TaAl_3Si_2O_{14}$ (CTAS) single crystal could not be grown due to the generations of the impurity phases. Therefore, in this study, we investigated the phase formation of the CTAS powder sintered at various temperature and grew the CTAS single crystals using the sintered powders. In addition, the crystal structure and piezoelectric properties of the grown CTAS single crystal were measured.

CTAS mixed powders were prepared from a nominal composition of $Ca_3TaAl_3Si_2O_{14}$ using $CaCO_3$, Ta_2O_5, α-Al_2O_3 and SiO_2 powders (> 4N) and the mixed powders were sintered at 1100~1350°C in air several times. Phases of the sintered powders were identified by the powder X-ray diffraction (XRD) measurement. The crystal growth was performed using the sintered powders by the Czochralski (Cz) method using an Iridium crucible in N_2+2%O_2 atmosphere. Phases of the grown CTAS crystals were investigated by the powder XRD measurement and scanning electron microscopy (SEM). Chemical compositions of grown crystals were analyzed by the Energy Dispersive X-ray spectroscopy (EDX). Laue image was obtained on the facet surface of the crystal and the crystal orientation was identified to prepare the X-cut specimen for the measurements of piezoelectric properties. Piezoelectric properties of the X-cut specimen were measured by the impedance analyzer.

The CTAS sintered powder were prepared under various sintering temperature (1100~1350°C). Main phases were impurity phases in the powder XRD patterns of the powders sintered at 1100~1300°C and there was a small amount of the CTAS phase. On the other hand, the impurity phases were dramatically decreased by the sintering at 1350°C and main phase was the CTAS phase in the powder XRD pattern of the powder sintered at 1350°C. In addition, the CTAS single crystals were grown using the CTAS sintered powders. The CTAS single crystal grown from the powder sintered at 1350°C indicated a single phase of the langasite phase while the CTAS crystals grown from the powder sintered below 1300°C included many impurity phases. Clear Laue pattern could be obtained from the facet surface of the CTAS single crystal grown from the powder sintered at 1350°C and there was no impurity phase in the SEM image. Details of the phase formation and crystal growth, and crystal structure and piezoelectric properties of the grown CTAS single crystal will be reported.

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