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Dependence of acoustic property on Al substitution for $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ single crystal

$Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$単結晶の音響特性における Al 置換量依存性

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1. Introduction

$Ca_3TaGa_3Si_2O_{14}$ (CTGS) [1, 2], one of the langasite-type single crystals, is a promising material for pressure sensors operating under high temperature environment as well as high stability oscillators for future communication applications. Although CTGS has no rare-earth elements unlike previous langasite-type crystals such as $La_3Ga_5SiO_{14}$ (LGS) and $La_3Ta_{0.5}Ga_{5.5}O_{14}$ (LTG), they include expensive element of Ga. In previous work, we have successfully grown Al-substituted CTGS single crystal, that is $Ca_3Ta(Ga_{0.5}Al_{0.5})_3Si_2O_{14}$ (CTGAS), to reduce Ga[3]. However, physical performance of the mixed crystals formed as a result of partial substitution of Ga^{3+} in CTGS with Al^{3+} is not clear. Therefore, examination of acoustic properties of the crystals having intermediate composition of $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ (CTGAS*x*) was considered to be important for further understanding of nature of LGS-type materials.

In this paper, we examined dependences of acoustical properties on Al substitution for CTGAS*x* single crystals using the ultrasonic micro-spectroscopy (UMS) technology [4-6].

2. Specimens

We grew CTGAS*x* single crystals with different compositions of x=0, 0.25, and 0.50 by Czochralski method pulling along *Y*-axis[3]. Crystals are about 1 inch$^{\Phi}$ × 60 mm^{L}. We prepared five specimens of *X*-, *Y*-, *Z*-, 35°*Y*-, and 140°*Y*-cut with 2-mm thickness from each crystal ingot.

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3. Experiments

Using the plane wave ultrasonic material characterization (PW-UMC) system[6], which is one of the main system of the UMS technology, we measured longitudinal wave velocities at 50-450 MHz and shear wave velocities at 40-200 MHz at 23°C. We could not observe any velocity dispersions in these frequency ranges for all results. Figs. 1 and 2 show measured results of longitudinal and shear waves velocities for each specimen as a function of Al substitution content. Al contents were measured by electron probe microanalyzer (EMPA). Bulk wave velocities for all propagation directions linearly increase with Al content.

Density was also measured at 23°C based on the Archimedes method. Results are shown in Fig. 3. Density linearly decreases with Al content.

4. Discussion

From the results in Fig. 1 and Fig. 2, we found that bulk waves linearly increase with Al content. However, their sensitivities to Al content (gradient of the lines in Figs. 1 and 2) are different depending on propagation direction. Especially for Z-propagation, velocity variations due to Al substitution are significantly great. This suggests that sensitivities for c_{33} and c_{44} to Al content are relatively larger among elastic constants.

Because Ga is heavier than Al, density of CTGAS decreases as compared with CTGS due to the effect of Al substitution. We estimated the increase of velocities due to the decrease of density according to the relationship of $V=(c'/\rho)^{1/2}$. As a result, we found that increase of velocities in Figs. 1 and 2 are due to not only decrease of density but also increase of effective elastic constants.

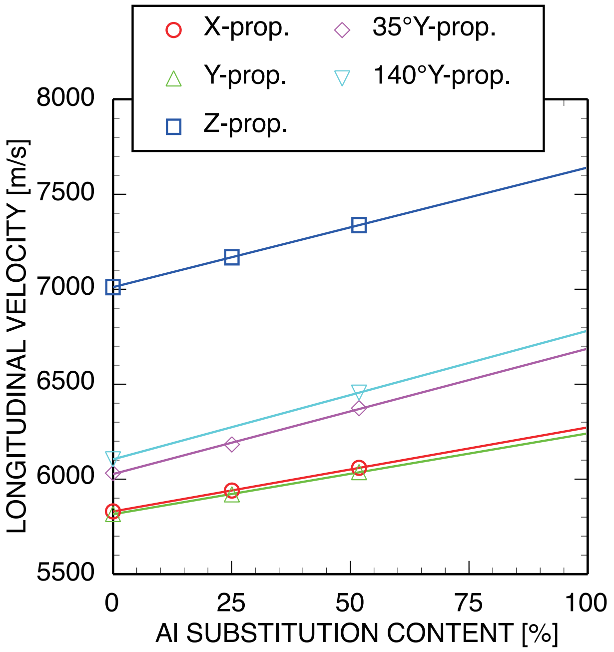

Fig. 1 Al substitution dependences of longitudinal wave velocities for CTGAS.

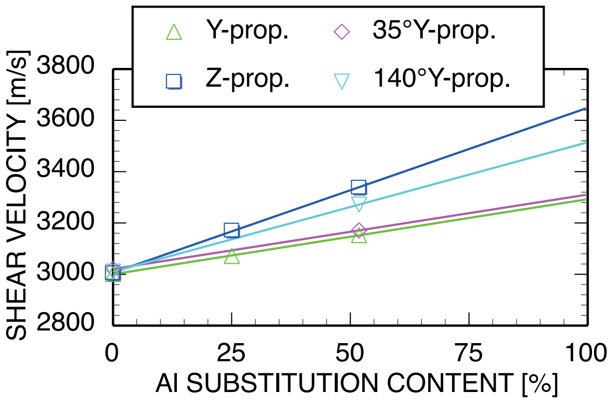

Fig. 2 Al substitution dependences of shear wave velocities for CTGAS.

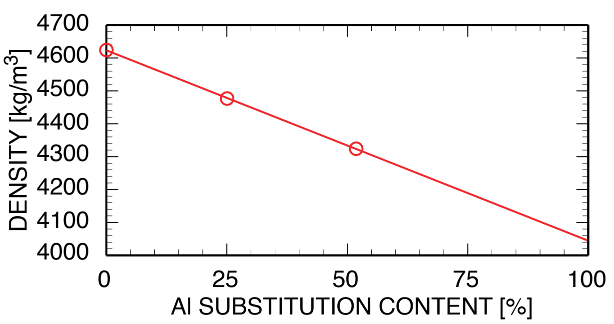

Fig. 3 Al substitution dependences of densities for CTGAS.

5. Conclusion

We examined dependence of acoustic properties for CTGAS on Al substitution content. Through measurements of bulk wave velocities for *X*-, *Y*-, *Z*-, 35°*Y*-, and 140°*Y*-prop. by using the PW-UMC system, longitudinal and shear waves velocities for CTGAS linearly increase with Al content for all propagation directions. Density linearly decreases with Al content. From these results, we suggested that effective elastic constants for CTGAS linearly increase due to the effect of Al substitution.

Acknowledgment
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