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Effects of Al substitution for $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ piezoelectric single crystals

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ABSTRACT

$Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ [CTGAS] material is a piezoelectric material belonging to the Langasite-type group (space group: P321).and the CTGAS single crystals (x=0, 0.25, 0.50, 0.75) with a diameter of 1 in. were grown by a Czochralski [Cz] method using an Ir crucible under Ar+2%O_2. The CTGAS single crystals without any cracks could be grown using a CTGAS seed crystal in the x range of $0 \le x \le 0.75$. Cell parameters, a and c, decrease while the a/c ratio increased with the Al concentration. Piezoelectric constant d_{11} and electromechanical coupling factor k_{12} for the X-cut specimens of the CTGAS single crystals were increased by the Al substitution.

1. Introduction

$Ca_3BGa_3Si_2O_{14}$ (B=Nb, Ta) materials belong to the Langasite group with the P321 space group and have been investigated as a piezoelectric material due to their high piezoelectric constant and electromechanical coupling factor at high temperature [1–5]. The $Ca_3NbGa_3Si_2O_{14}$ [CNGS] and $Ca_3TaGa_3Si_2O_{14}$ [CTGS] single crystals have been expected to be applied for some sensor devices which are used under high temperature like in the case of combustion pressure sensor. In addition, the CNGS and CTGS single crystals have comparable temperature stability of frequency [TCF] to quartz [6] and a small-size oscillator can be achieved using the CNGS and CTGS piezoelectric elements.

Ga sites in the CNGS and CTGS can be substituted by Al^{3+} ion and we reported the crystal growth of the Al doped CNGS, $Ca_3Nb(Ga_{1-x}Al_x)_3Si_2O_{14}$, [CNGAS] and Al doped CTGS, $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$, [CTGAS] single crystals by a micro-pulling-down (μ-PD) method [7,8]. In the studies, the CNGAS single crystals could be grown in the x range of $0 \le x \le 0.6$ and the CTGAS single crystals in the x range of $0 \le x \le 1$. In addition, we grew the CNGAS bulk single crystals by a Czochralski [Cz] method and the piezoelectric properties were measured [9]. The results of the piezoelectric properties measurements revealed that the piezoelectric constant, d_{11}, and electromechanical coupling factor, k_{12}, for the X-cut specimens increased by the Al substitution. However, it is difficult to obtain the CNGAS single crystals whit high quality in the x range of $0.8 \le x \le 1$ by the melt-growth method while there are some reports about $Ca_3NbAl_3Si_2O_{14}$ (x=1) [CNAS] single crystal [10].

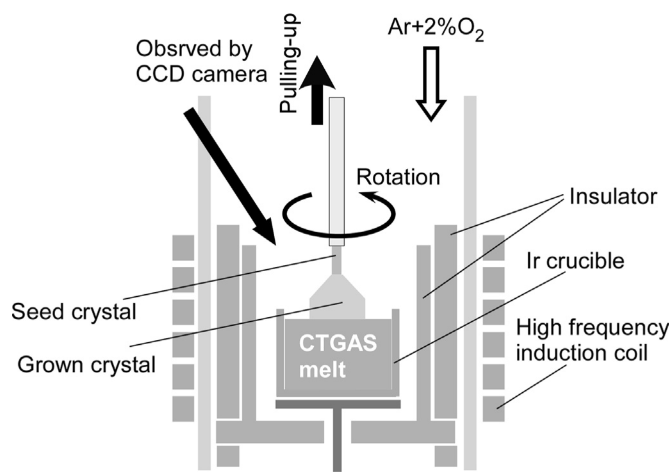

Fig. 1. Schematic of the Cz method for the crystal growth of CTGAS single crystals.

On the other hand, $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$, [CTGAS] single crystals can be grown by the melt-growth method in the all x range and improvements of piezoelectric properties are expected by the change of

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the Al concentration. However, there are only reports about crystal growth and piezoelectric properties of CTGS and $Ca_3TaAl_3Si_2O_{14}$ (x=1) [CTAS] single crystals. Therefore, in this study, we grew the CTGAS single crystals and investigated the phases, cell parameters, densities, crystallinities, chemical compositions and piezoelectric properties of the CTGAS single crystals to reveal the effects of Al substitution on the crystal growth, crystal structure and piezoelectric properties.

2. Experimental

$Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$ single crystals with a diameter of 1 in. were grown by the Cz method using a ϕ50 mm Ir crucible. Fig. 1 is the schematic of the Cz method for the growth of the CNGAS single crystals. Mixed powders with nominal compositions of $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$, x=0, 0.25, 0.50, 0.75 were prepared using starting materials, $CaCO_3$, β-Ga_2O_3, α-Al_2O_3, SiO_2 (> 4N purity) and

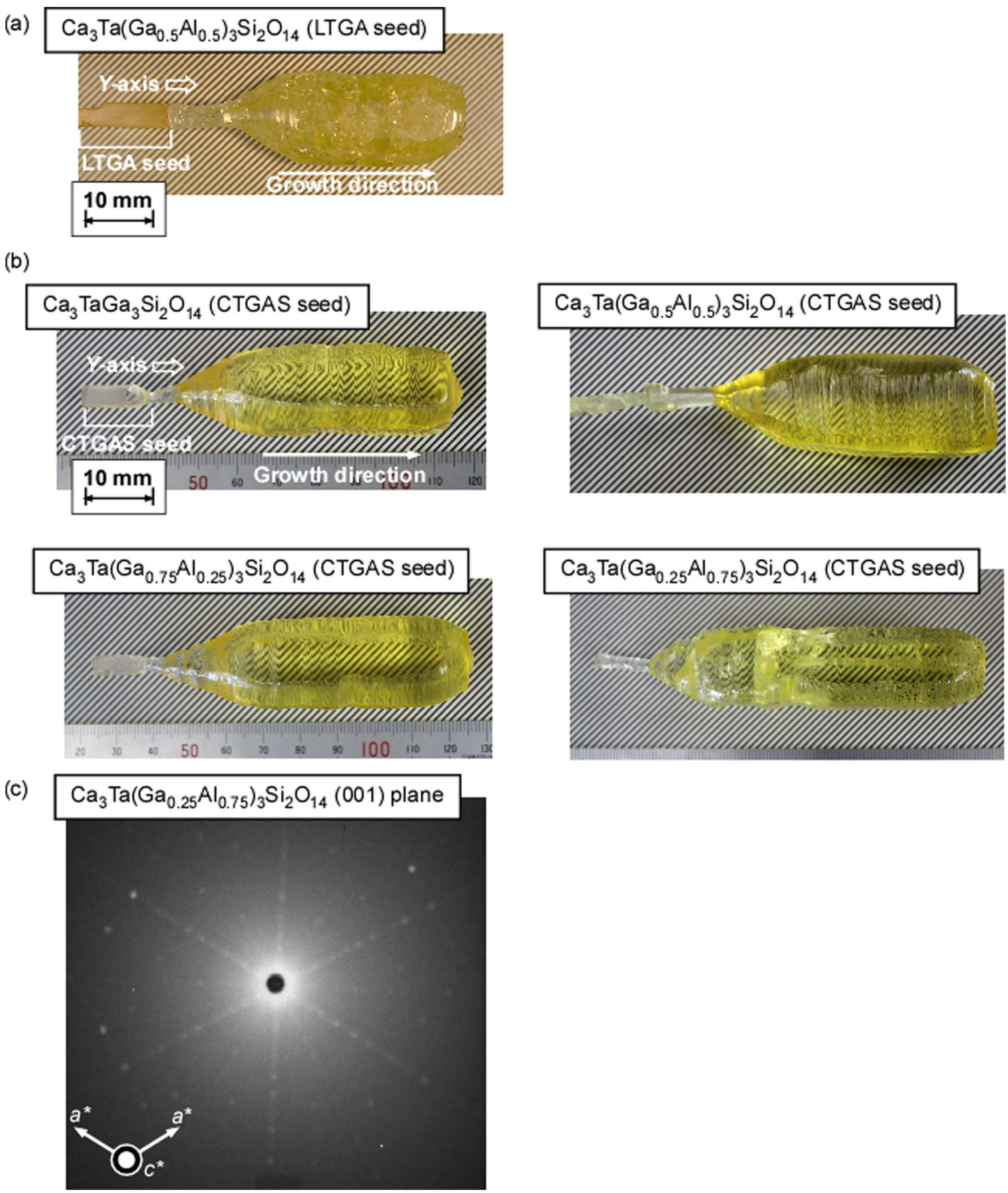

Fig. 2. (a) As-grown CTGAS single crystals grown using LTGA and (b) CTGAS seed crystals. (c) Back-reflection Laue image on the crystalline facet. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)