3P1-3

Consideration of relationship between crystal structure and coefficient of thermal expansion on Langasite-type piezoelectric single crystals

ランガサイト系圧電単結晶における結晶構造と線膨張係数の関係に関する検討

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1. Introduction

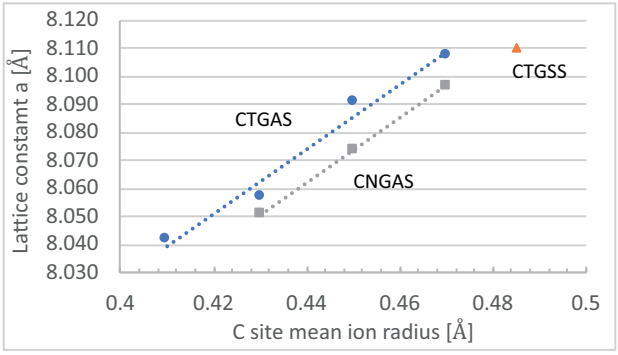
Langasite-type piezoelectric single crystals have the chemical composition of $A_3BC_3D_2O_{14}$ and a trigonal structure with a 32-point group as same as α-quartz. Langasite-type piezoelectric single crystals have two to three times higher electromechanical coupling factors than that of quartz and stable piezoelectric properties at high temperatures owing to the absence of phase transition up to the melting point. We have developed langasite-type single crystals such as $Ca_3Ta(Ga_{1-x}Al_x)_3Si_2O_{14}$[CTGAS], $Ca_3Ta(Ga_{1-x}Sc_x)_3Si_2O_{14}$[CTGSS], $Ca_3Nb(Ga_{1-x}Al_x)Si_2O_{14}$ [CNGAS] in which Ga (C-site element) are substituted with Al or Sc in $Ca_3TaGa_3Si_2O_{14}$[CTGS], $Ca_3NbGa_3Si_2O_{14}$ [CNGS], and have found out its possibility of application to small and low power consumption resonator [1-3]. We also revealed that material constants (elastic constants, piezoelectric constants, and so on) and their thermal coefficients varied linearly with substitution amount by substituting Ga with other elements with different ion radius [4, 5]. Controlling acoustic velocity propagating in the piezoelectric substrate and thickness of the substrate are necessary to control the resonant frequency of the resonator. The acoustic velocity can be obtained by getting the elastic constants and piezoelectric constants. However, behavior of the coefficient of thermal expansion (CTE), which relates directly to thickness of the substrate, were not always obvious.

In this study, we investigate the relationship between CTE and crystal structure of langasite-type single crystal.

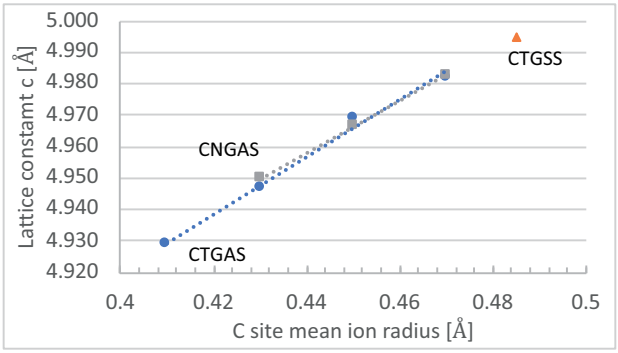
2. Relationships among parameters

Taking CTGS、CTGAS、CTGSS、CNGS、CNGAS as specimen, lattice parameters are plotted as a function of C-site mean ion radius. The results are shown in **Fig. 1**. Lattice constant c increases in proportion to the C-site ion radius regardless of B-site element (Ta or Nb). On the other hand, lattice constant a increases in proportion to the C-site ion radius revealing difference between the results of CTGAS and CNGAS depending on B-site element. The plot of CTGSS is not on the fitting line of CTGAS, and its lattice constant a does not change by increasing C-site ion radius.

Fig. 2 shows relationship between C-site ion



(a) Lattice constant a



(b) Lattice constant c

Fig. 1 Relationship between lattice parameters and C site mean ion radius for CTGAS, CTGSS, CNGAS.

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radius and first order CTE. α_{11} (CTE along a-axis direction) is in direct proportion to C-site ion radius and there is the difference due to B-site element. However, the result for CTGSS is not on the fitting line for CTGAS, α_{11} does not increase with increase of C-site ion radius. On the other hand, α_{33} (CTE along c-axis direction) decreases with C-site ion radius and there is the difference due to B-site element. The result for CTGSS is not on the fitting line for CTGAS as same as α_{11}. Besides the result of CTGS (the largest C-site mean ion radius among CTGAS) is also not on the fitting line, it seems that decrease of α_{33} stops at the range over 0.46 Å of C-site ion radius.

3. Discussion

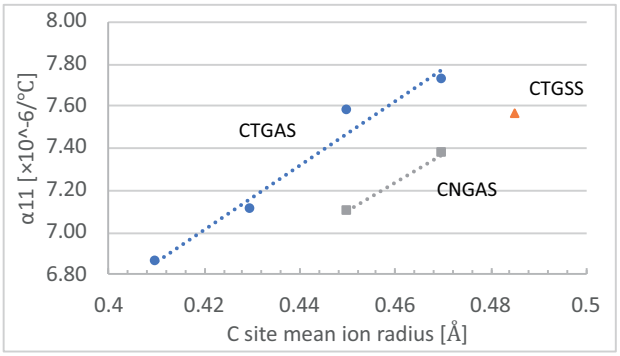
Taking $La_3Ga_5SiO_{14}$ (LGS), $La_3Ta_{0.5}Ga_{5.5}O_{14}$ (LTG), $La_3Nb_{0.5}Ga_{5.5}O_{14}$ (LNG) addition to CTGAS, CTGSS, and CNGAS, relationship between first order CTE and volume of unit cell obtained from lattice constants was shown in **Fig. 3**. The result of Fig. 3 reveals that CTE becomes smaller with increase of volume of unit cell. For the case of ultra-low expansion SiO_2 glass, it is well known that CTE can be reduced to ppb order by doping TiO_2. The reason is interspace for absorbing thermal expansion increases by doping TiO_2 because bond length of Ti-O is longer than that of Si-O and bond angle of Si-O-Ti is larger than that of Si-O-Si. In the same manner as the TiO_2-SiO_2 ultra-low expansion glass, the result of Fig. 3 suggests that bond length and bond angle become larger by element substitution resulting in smaller CTE.

Although B-site ion radius of Ta and Nb does not differ from each other, there are some difference between CTEs for CTGAS and CNGAS as shown in Fig. 2. The reason is that internal structure changes by varying bonding angles such as O-Ta-O and O-Nb-O although B-site ion radius does not change [6].

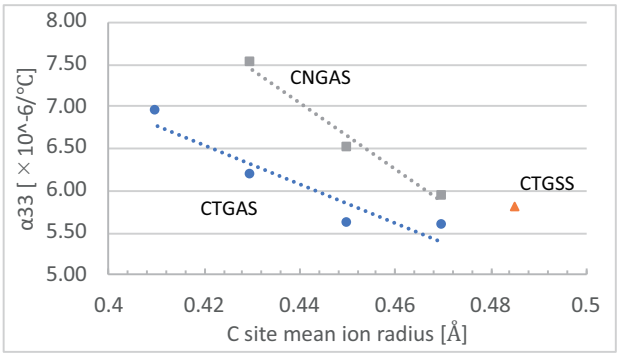
CTE becomes smaller by increasing volume of unit cell. As shown in Fig. 2, however, relationship between C-site ion radius and CTE ∪ shows reverse trend (increase for α_{11}, decrease for α_{33}). Therefore, it is necessary to investigate about not only local structure but also whole of the crystal structure.

4. Conclusion

Relationship between CTE and crystal structure was investigated for langasite-type single crystals. We revealed that CTE becomes smaller with increase of volume of unit cell. Hereafter, it is necessary to investigate relationship between element substitution and crystal structure (bond length and bond angle).



(a) Coefficient of thermal expansion α_{11}.



(b) Coefficient of thermal expansion α_{33}.

Fig. 2 Relationship between coefficient of thermal expansion and C site mean ion radius for CTGAS, CTGSS, CNGAS.

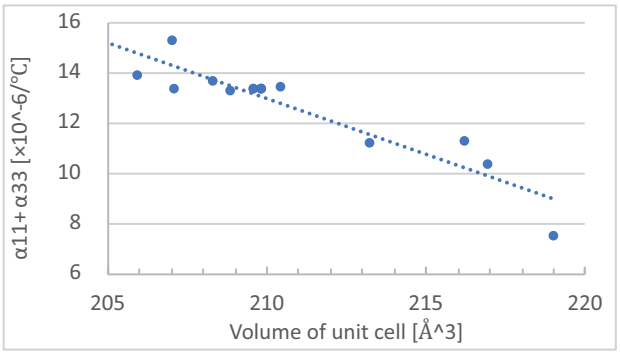


Fig. 3 Relationship between volume of unit cell and coefficient of thermal expansion ($\alpha_{11}+\alpha_{33}$).

References

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