Investigation of rotated X-cut $Ca_3TaGa_3Si_2O_{14}$ single crystals operating in FS mode in the temperature range up to 900°C

A. Medvedev, S. Sakharov, O. Buzanov, A. Zabelen, V. Alenkov
OAO «Fomos-Materials»
Moscow, Russia
medvedev@newpiezo.com

T. Kudo, Y. Yokota, K. Kamada, A. Yoshikawa
Institute for Materials Research,
Tohoku University
Sendai, Japan

Abstract— **Crystal elements manufactured made of calcium tantalum gallium silicate single crystals (CTGS) operating in face shear mode were investigated. The frequency temperature characteristics of the rotated X-cut in high temperature range (up to 900°) were measured. The largest coupling factor close to 21.1% was found for (XYt) 9°-cut. It was shown that frequency temperature characteristic has a departure from a quadratic parabola which is atypical for langasite family crystals. The maximum value of the turnover temperature point 578° corresponds to (XYt) 9°-cut.**

Keywords— $Ca_3TaGa_3Si_2O_{14}$, resonator; single crystal; bulk acoustic waves; temperature coefficient of frequency (TCF)

I. INTRODUCTION

Langasite family crystals are promising piezoelectric materials for the new generation of wide bandwidth acoustic wave filters, frequency control, and high-temperature sensors, as they can successfully replace the traditionally used ceramics and quartz. The crystals belong to the trigonal system (space group P321, point group 32) and have high acoustic Q-factors, zero temperature coefficients of frequency of the first-order, high coupling factors. Moreover, there is no phase transition up to pre-melting temperature.

$Ca_3TaGa_3Si_2O_{14}$ (CTGS) single crystal belongs to langasite family group and possesses a number of unique piezoelectric characteristics. The large magnitude of the piezoelectric modulus d_{14} and the small frequency deviation of thickness shear mode in a wide temperature range (the temperature coefficient of frequency of the second order (TCF(2)) is close to −22.6 ppb/°C^2 [1]) enable its application in the development and manufacture of piezoelectric devices operating at high temperature. On the other hand, there is a great demand for resonators operating in a face shear mode. These resonators can be used in force or acceleration sensors operating at high temperature range.

In this work, the main piezoelectric parameters and frequency versus temperature characteristics (FTC) for rotated X-cuts operating in a face shear mode were investigated.

II. EXPERIMENTAL

CGTS single crystals were grown by using the Czochralski method with RF-heating of crucible. The original charge was synthesized from high-purity tantalum pentaoxide, gallium oxide and calcium carbonate with a purity not worse than 4N. The growth process was carried out in Ar atmosphere. Argon was used as a protective gas medium. The crystals were grown along the crystallographic <100> direction. The pulling rate was 0.5 − 2 mm/h and the rotation speed was 10 - 15 rpm.

The orientations of CTGS crystals and cuts notation were made according to the negative value of piezoelectric modulus d_{11} for right-handed crystals. The plate's cuts were denoted by (XYt) Ψ. In this work, we used the plates of (XYt)–21°, (XYt)–9°, (XYt)–15°, (XY), (XYt)9°, and (XYt)30° cuts. The obtained crystals and plates were oriented using X-ray diffraction. The plates were square in shape and were lapped with 10μm Al_2O_3. The roughness of plates was Ra<0.24 μm. The thickness of plates was about 0.8 mm. Ir electrodes were deposited on parallel main faces of the samples. The thickness of electrodes was close to 300 nm. The impedance characteristics of the piezoelectric vibrators were measured using Agilent HP 4294A impedance phase-gain analyzer.

Fig.1. CTGS single crystal

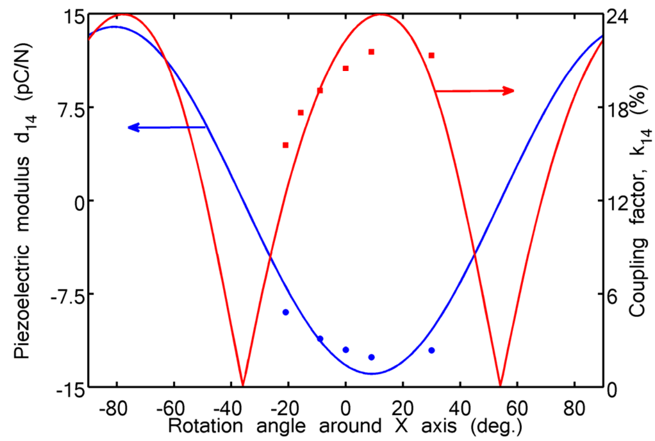

Fig.2. Piezoelectric modulus d_{14} and coupling factor k_{14} as a function of rotation angle Ψ. The measured values of the piezoelectric moduluses are marked by circles and the coupling factors are indicated by squares.

The coupling factor k_{ij} was determined using resonance anti-resonance method.

III. Results and Discussion

A. Piezoelectric modulus d'_{14} and coupling factor k'_{14}

Based on the piezoelectric moduluses obtained in [2] piezoelectric modulus d'_{14} was calculated, following the equation,

$$d'_{14} = d_{11}\sin(2\Psi) + d_{14}\cos(2\Psi) \quad (1)$$

The coupling factor k'_{14} was determined, using the following text,

$$k'^2_{14} = d'^2_{14} / s'_{44}\varepsilon_{11} \quad (2)$$

Using values of elastic constants [3] elastic compliance coefficients were calculated. These results are given in Table 1.

TABLE I. Elastic compliance coefficients s_{IJ} (10^{-12} m^2/N) of CTGS single crystal

Compliance coefficients	Value of quantity
s_{11}	8.702
s_{12}	-3.546
s_{13}	-1.571
s_{14}	-0.1573
s_{33}	5.706
s_{44}	23.928
s_{66}	24.496

The values of rotated elastic compliance coefficients s'_{44} were calculated, following the equation,

$$s'_{44} = s_{44}\cos^2(2\Psi) + (s_{11} + s_{33} - 2s_{13})\sin^2(2\Psi) + s_{14}\sin(4\Psi) \quad (3)$$

The value of dielectric constant $\varepsilon_{11}/\varepsilon_{11}$ =16.6 was taken.

Fig. 2 gives the variation of piezoelectric modulus d_{14} and coupling factor k_{14} as a function of rotation angle Ψ around crystallographic X-axis from -90° to 90°. The experimental values of coupling factor and piezoelectric modulus are also presented. The maximum value of coupling factor k_{14} is close to 23.96 %. The corresponding magnitude of the piezoelectric modulus d_{14} equals 13.87 pC/N observed at cut angle Ψ=12°10´. On the other hand, the coupling factor is zero at cut angle Ψ=54°10´.

B. Frequency temperature characteristics

Deviations of resonance frequency for rotated (XYt) Ψ -cuts in the temperature range from 25°C up to 900°C are presented in Fig.3. As observed, the frequency deviation for the temperature range from room temperature up to 900°C has the maximum value of about 20 000 ppm for (XYt)9°-cut and is close to 7 500 for (XYt)-21°-cut. Corresponding measured coupling factors for those cuts are 21.04% and 15.53%, respectively.

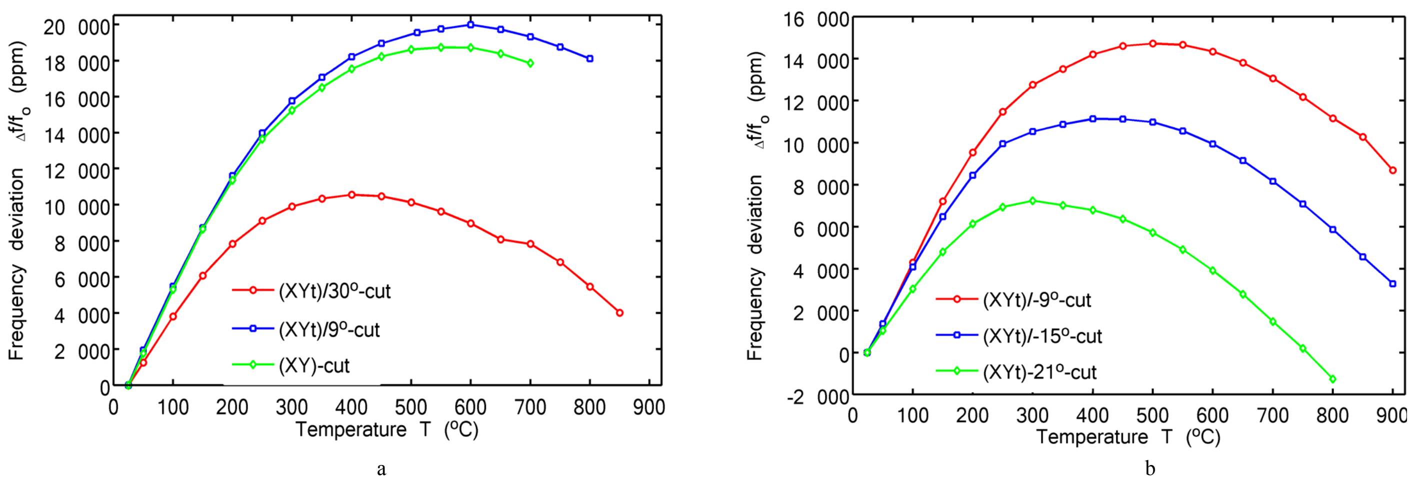

Fig.3. Frequency temperature characteristics of a) (XY), (XYt)9°, and (XYt)30° cuts ; b) (XYt)–21°, (XYt)–15°, and (XYt)–9° cuts. The measured values are marked by circles, squares, and diamonds.

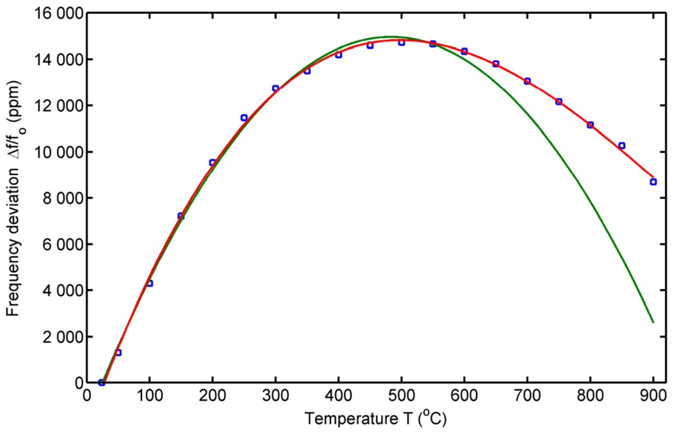

Fig.4. Frequency temperature characteristic of (XYt)-9°-cut.

As shown in fig.4, the frequency temperature characteristic has a departure from the quadratic parabola (green curve) which is atypical for langasite family crystals. The frequency temperature characteristic may be approximated using polynomial function of the second-order. In this case, the values of temperature coefficient of frequency of the second order for lower and upper parabola branches should be different. For example, for (XYt)-9°-cut the TCF(2) of the lower branch is close to −71.2 ppb/°C^2, and it is close to −30.9 ppb/°C^2 for the upper parabola branch.

All six cuts have turnover temperature points. Fig. 5 gives the variation of turnover temperature as a function of rotation angles around X-axis for the angles range from -21° to 30°.The maximum value of the turnover temperature is about 578°C for (XYt)9°-cut.

CONCLUSION

$Ca_3TaGa_3Si_2O_{14}$ single crystals operating in face shear mode in the temperature range up to 900°C were investigated. It was shown that the largest coupling factor of $Ca_3TaGa_3Si_2O_{14}$ single crystals operating in FS mode is close to 21.1% for (XYt)9°- cut. The frequency versus temperature characteristics of rotated X-cuts have extremum points in the temperature range from 330 to 578 °C.

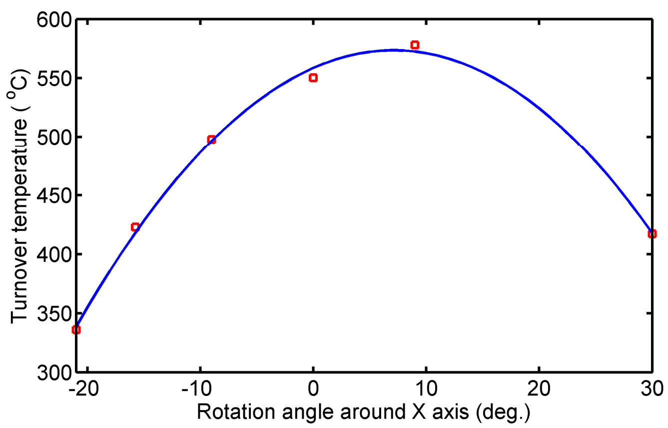

Fig.5. Turnover temperature as a function of rotation angle around X-axis.

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