

# Crystal growth and evaluation of langasite type crystal grown by micro pulling down method

ランガサイト型圧電単結晶のマイクロ引き下げ法による育成および評価

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## 1. Back ground

To solve the environmental problem, energy harvesting has been widely researched. For the energy harvesting sun light, wind, are widely used. On the other hand, vibration are not widely used for the energy harvesting though it is useful energy because it's continuously supplied energy.

Vibration energy are used as electricity with piezoelectric material, magnetostrictive material, electret and magnet devices. For the piezoelectric generator, Ferroelectric ceramics like PZT are famous. Though, it is frangible and contains lead in the device, we tried to make a generator with piezoelectric single crystal. As the material, we choose langasite type crystal.

Langasite type crystal are investigated as a piezoelectric material for applications such as piezoelectric sensor, oscillator due to no phase transition up to melting point, and high electromechanical coupling coefficient.

Langasite type crystals were first developed as  $\text{La}_3\text{Ga}_5\text{SiO}_{14}$  (LGS),  $\text{La}_3\text{Ta}_{0.5}\text{Ga}_{5.5}\text{O}_{14}$  (LTG), and  $\text{La}_3\text{Nb}_{0.5}\text{Ga}_{5.5}\text{O}_{14}$  (LNG) which are called disordered structure crystal and in 2000s ordered crystals such as  $\text{Ca}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$  (CNGS),  $\text{Ca}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$  (CTGS),  $\text{Sr}_3\text{NbGa}_3\text{Si}_2\text{O}_{14}$  (SNGS),  $\text{Sr}_3\text{TaGa}_3\text{Si}_2\text{O}_{14}$  (STGS) were developed. Ordered crystal has higher electrical resistivity and lower impedance.

In this report, we will grow the langasite type crystal for oscillator operate in low frequency and evaluate the properties to consider it for the application to structures like building and bridge.

## 2. Experiment

We used micro-pulling-down ( $\mu$ -PD) method for the growth technique. In this method crystals are grown by pulling down the melt from the die at the bottom of the crucible. And the crystal shape can be controlled by the shape of die which is useful to grow the material in particular form with

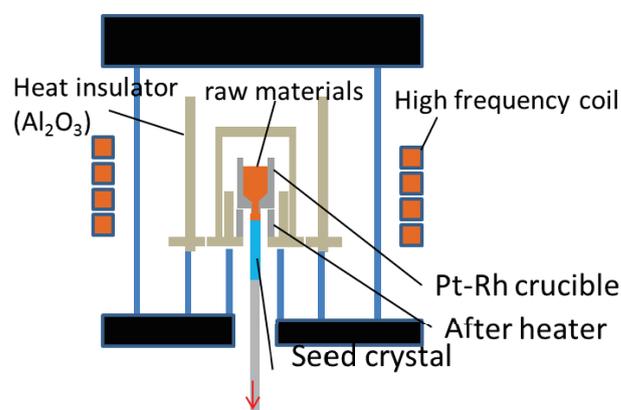


Fig. 1 Pattern diagram of  $\mu$ -PD method

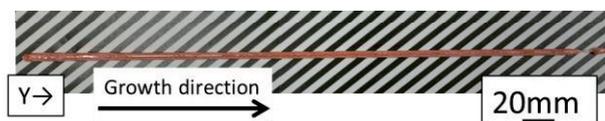


Fig. 2 CNGS crystal grown by the  $\mu$ -PD method.

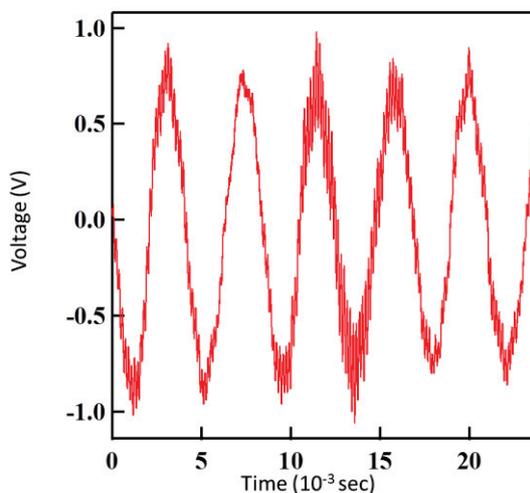


Fig. 3 Time vs. voltage diagram of CNGS crystal generate with the sound from speaker.

high growth rate. Fig. 1 shows the Pattern diagram of  $\mu$ -PD method. For starting materials, high purity powder (>4N) or bulk crystal were used to grow the crystal. Powder were mixed and sintered at 1000 to 1200°C for several times. Starting materials were entered in Pt crucible with the die at the bottom. Crucible was heat up to melting point and crystal was grown by using seed crystal. Growth rate was 0.03~0.2 mm/min. In order to evaluate low frequency operation, electrode was put on to the crystal. Oscilloscope and were used for the measurement. circuit contains resistance (1~2M  $\Omega$ ) and also evaluate with rectifier circuit. Vibration was given by hitting the sample or the speaker vibrating at lower frequency.

### 3. Result

Crystals with composition CNGS and CTGS were grown by  $\mu$ -PD method. Crystal was succeed to grow as the same shape of die. Meniscus was about 0.5~1mm and the grown crystal was longer than 200mm with  $\phi$ 3mm and 1 $\times$ 3 mm shape. Fig. 1 shows a typical crystal of CNGS grown by the  $\mu$ -PD method.

With the generation test, voltage from the crystal was observed. Output voltage is shown in Fig. 2. From the result frequency of the crystal has several modes. And its lowest frequency mode can be explained as a bending vibration mode. Though crystal frequency has inversely proportional to the square of the length. Crystal was vibrated with both hitting the sample and speaker. Maximum workload was higher than 200 $\mu$ W.

### 4. Conclusion

We succeed to grow the CNGS, CTGS type crystal with  $\mu$ -PD method. The crystals were longer than 200mm with  $\phi$ 3mm and 1 $\times$ 3mm shapes.

Maximum power generated by single crystal was higher than 200 $\mu$ W and it was possible to generate with vibration. Though langasite type crystal can be consider as a useful material for oscillator.

### References

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