High temperature tolerant transducer for Ultrasonic Assisted Hydrothermal Method
超音波アシスト水熱合成法のための高温対応振動子の開発

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1. Introduction

High quality piezoelectric materials can be synthesized with a hydrothermal method from high-temperature (140 ~ 210 deg.) aqueous solutions at high vapor pressures. However, the reaction time is very long. To overcome this disadvantage, we had proposed to irradiate an ultrasonic to the solution during the hydrothermal method. We named it “ultrasonic assisted hydrothermal method (UAHTM)”[1-3] (Fig. 1).

In spite of UAHTM effect, some problems still remain. For the ultrasonic irradiation, PZT rings are used as a high-power ultrasonic transducer. The piezoelectric properties of PZT start to getting worse from about 150 deg. Thereby, ultrasonic irradiation during UAHTM became unstable and its repeatability was insufficient.

To solve this problem, we have developed high-power ultrasonic transducer using LiNbO₃ (LN) single crystal as a source of vibration instead of PZT. LN has high Curie point (1210 deg.) and its piezoelectric property is expected to be kept at high temperature.

<table>
<thead>
<tr>
<th>LiNbO₃ (36°Y-cut)</th>
<th>PZT-8</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_{33}$</td>
<td>0.57</td>
</tr>
<tr>
<td>$e_{33}/e_{0}$</td>
<td>40</td>
</tr>
<tr>
<td>$e_{33}$ [C/m²]</td>
<td>4.7</td>
</tr>
<tr>
<td>$d_{33}$ [pC/N]</td>
<td>38.8</td>
</tr>
<tr>
<td>$T_c$ [°C]</td>
<td>1210</td>
</tr>
</tbody>
</table>

Table 1 Property of LiNbO₃ and PZT [4-6]

Piezoelectric constant $d$ of LN is smaller than that of PZT, so the vibration velocity of LN transducer is lower than that of PZT transducer at the same input voltage and the same quality factor. However, critical stress limitation of LN was reported to be higher than that of PZT [7]. Therefore, by applying high input voltage and by using large number of multi layered rings, we tried to realize higher vibration velocity of LN transducer.

3. Construction of the transducer

Left of Fig. 2 shows the conventional PZT transducer used to UAHTM. It is a bolted Langevin transducer (BLT) using 4 PZT rings. Horn and bolt part is made of Hastelloy which has resistance to high alkaline chemicals. Aluminum washers are settled both side of PZT to keep the preload by thermal expansion. Nodal point was flange at the center, and vibration was magnified by a factor of five by the horn.

Right of Fig. 2 shows the LN transducer developed in this research. To prevent the cracks at the LN rings, the rings and the electrodes were adhered by electro-conductive glue DOTITE XA-874 (FUJIKUEA KASEI CO., LTD.).
4. Evaluation of the transducer

To evaluate the temperature dependence of the transducer property, the admittance curve was measured from 25 to 250 deg. celsius. Fig. 3 shows the temperature dependence of the quality factor of LN and PZT transducer.

![Quality Factor vs Temperature Graph](image)

**Fig. 3** Temperature dependence of quality factor

At the higher temperature, the quality factor of PZT transducer was decreased. On the other hand, that of LN transducer wasn’t. This result verified that the LN transducer is effective at high temperature.

To examine the ultrasonic irradiation by the LN transducer on UAHTM, a PZT thin film was synthesized. A titanium substrate was put in the solution, and PZT was synthesized for 24 hours. The transducer was driven at 900 V\textsubscript{p-p} input voltages. Left image of Fig. 4 shows the surface of the PZT film. Right image is the surface of a PZT film synthesized without ultrasonic assist in the same synthesis condition.

![Surface of PZT Film](image)

**Fig. 4** Surface of PZT film
(Left: UA by LN transducer  Right: without UA)

Particle size of PZT synthesized with UAHTM was smaller than that without ultrasonic assist. It is known that high power ultrasound can make the PZT particle smaller. This result indicates that the LN transducer can irradiate adequate ultrasound power.

5. Conclusion

In this study, the ultrasonic transducer using LN single crystal rings was developed to apply high temperature condition of UAHTM. By comparing the property of the developed the LN transducer and conventional PZT transducer, it was verified that the LN transducer was suitable for high temperature conditions.

Now, we are trying to synthesize lead-free piezoelectric materials by UAHTM using the LN transducer.

Acknowledgment

This work is supported by the New Energy and Industrial Technology Development Organization (NEDO). We appreciate kind supports from Furuuchi Chemical Corporation, and Taiatsu Techno Corporation.

References

7. Seiji Hirose, Sadayuki Takahashi, “Measuring methods of high power characteristics on piezoelectric transducer and design method of piezoelectric transformer considering these results” *The Transactions of the Institute of Electronics, Information and Communication Engineers. A*, 380-A(10), 1997, pp.1621-1636