

Evaluation of Mass-Produced Commercial LiTaO₃ Single Crystals Using the LFB Ultrasonic Material Characterization System

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Abstract—A mass-production line of lithium tantalate (LiTaO₃) crystals with a maximum charge number of 60 for surface acoustic wave (SAW) devices was evaluated with the line-focus-beam (LFB) ultrasonic material characterization system. Some serious problems associated with chemical compositions were observed and resolved by measuring the velocities of Rayleigh-type leaky surface acoustic waves (LSAWs), V_{LSAW} , for two groups of LiTaO₃ wafers: 21 36°YX-LiTaO₃ wafers selected randomly from crystal ingots grown with different charge numbers in different furnaces, and 14 42°YX-LiTaO₃ wafers obtained at the top, middle, and bottom parts from 5 crystals selected from 39 crystals grown successively in the same furnace and crucible. Using the measured V_{LSAW} and the predetermined relationship between V_{LSAW} and Li₂O concentrations, $M(\text{Li}_2\text{O})$, we estimated the average $M(\text{Li}_2\text{O})$ controlled in the current mass-production line to be about 48.77 mol% with a maximum difference of 0.75 mol%. The composition for each crystal ingot increased linearly about 0.04 mol% from the top to the bottom, and no dependence on the charge number was observed, as the melt composition used for the mass production was controlled through Curie temperature (T_C) measurements. A nearly true congruent composition of 48.49 Li₂O-mol% was obtained through the precise V_{LSAW} data for the 42°YX-LiTaO₃ wafers, that was about 0.3 mol% less than the melt composition in the production line. It was also pointed out that the T_C measurement conditions, including room temperatures surrounding the measurement systems, should be re-examined for reliable production control. A guideline for more efficient mass production of the crystals has been established concerning the true congruent composition as the starting material.

I. INTRODUCTION

THE LiTaO₃ single crystals were successfully grown using the Czochralski method in 1965 [1], and the crystals were first put into practical use as substrates of X-cut 112.2°-rotated-Y-propagating (X-112.2°Y) LiTaO₃ for surface acoustic wave (SAW) devices in 1977 [2]. Since then, demand for the SAW device wafers has been increasing, especially with the rapid spread of recent mobile communications [3]–[5]. For that reason, many manufacturers recently entered the highly competitive field of crystal production. In mass production of SAW devices, it is neces-

sary to effectively grow homogeneous crystals to meet the requirements for high performance and low cost.

Based on the results of basic research associated with crystal growth conditions and physical and chemical properties [6]–[8], great efforts have been made in industry to establish the proper crystal growth conditions for mass production, in order to ensure homogeneities in chemical compositions within and among the crystal ingots [9]–[12]. The optimum growth conditions for growing commercial crystals with the same chemical composition have not yet been established [13]–[16]. Curie temperature (T_C) measurements are most commonly used to evaluate the chemical compositions. Ideally, each grown crystal ingot should have the same T_C values along the pulling-axis direction as well as along the diameter-axis direction within the crystal. At present, the actual homogeneity of the crystals is not sufficiently known because the real growth conditions for large-diameter, homogeneous crystals have not yet been established, and the accuracy of T_C measurements for evaluation is insufficient [17].

We have been studying the development and application of a high-precision material evaluation technology, the line-focus-beam ultrasonic material characterization (LFB-UMC) system, by measuring the velocities of Rayleigh-type leaky surface acoustic waves (LSAWs), V_{LSAW} , on a water-loaded specimen surface [18], [19]. So far, we have improved the system [17], [19] and have developed the system calibration method [20], [21] for highly precise measurements. We also have established evaluation procedures for lithium niobate (LiNbO₃) and LiTaO₃ single crystals and wafers [13]–[17], [22]–[26]. In addition, we have used the system to evaluate the measurement conditions for T_C , which is one of the crystal evaluation parameters for analyzing the chemical compositions, by setting guidelines for evaluating the T_C measurement conditions using the standardized scale of V_{LSAW} [27]. Furthermore, by evaluating detailed chemical composition distributions within optical-grade LiTaO₃ single crystals with the V_{LSAW} measurements, we have improved the growth conditions of the crystals and have succeeded in growing more homogeneous single crystals [15], [26].

In this paper, we use the LFB-UMC system to evaluate LiTaO₃ single crystals mass produced by a manufacturer that grows crystals successively up to 60 times in charged cycles. We also investigate some of the industrial and scientific problems involved in the mass-production line by

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examining the distributions of the chemical compositions within and among the crystals.

II. MASS-PRODUCTION PROCESS

The LiTaO₃ crystals are grown by the Czochralski method by putting a seed crystal rod into the material melt within an iridium crucible and pulling it up while rotating. The growth usually is stopped when about half of the melt in the crucible is crystallized and the reduced material is added to the crucible so that the next crystal can be pulled up. The material remaining at the top and bottom of crystals after the process of forming cylindrical crystal boules in the wafer fabrication line as well as crystals with defects, such as cracks, are recycled as the material source for crystal production because the material is so expensive. Attention should be paid to the fact that, if the composition for crystal growth differs from the real congruent composition, the melt composition changes as the growths are repeated. Distributions in chemical composition within and among grown crystals then are inevitable [11]–[13], [16]. Furthermore, as the number of crystal growth cycles is increased, the chemical composition ratio varies more or less, and there could be a problem if some of the impurities are incorporated within the crucible during the crystal growth process. Because of this, the crucible is emptied and washed after several crystal growths before starting a new set with the same charge numbers. The demerits of this process are the decreased effectiveness in mass production and the extra cost. Recently, some manufacturers have strongly endeavored to improve the growth conditions and to increase the charge number over 10 in order to reduce production costs, based on the industrial experiences of evaluating crystals with T_C . However, there has been no technical data reporting the relationship between the crystal quality and the charge number increased so far. Therefore, we investigate crystals grown with a large charge number of 60 by the manufacturer used as an example here to determine whether the chemical compositions are carefully controlled or whether there are any technical problems in the process.

III. LFB-UMC SYSTEM

The principle of the $V(z)$ curve measurements to obtain V_{LSAW} using the LFB-UMC system was presented in detail in the literature [18]. The newest measurement system [17], [19] installs the whole mechanical system, including the ultrasonic device and specimen, in a temperature-controlled chamber to stabilize the measurement environment. The system includes a specimen transfer mechanism, a temperature-controlled pure-water supply and drain system, and an automatic tilting stage so that specimens can be changed and aligned easily by remote control so effective measurements can be made without disturbing the stabilized measurement environment.

The system achieves the highest accuracy among the LFB-UMC systems that we have developed. The accuracy

in V_{LSAW} measurements was estimated to be $\pm 0.0013\%$ ($\pm 2\sigma$ where σ is the standard deviation) at an arbitrary single chosen point on the surface of the specimen and $\pm 0.003\%$ within a two-dimensional, continuous-scanning area of $75 \text{ mm} \times 75 \text{ mm}$ [17]. The absolute accuracy of the LSAW velocities after system calibration [20] using the standard specimens of LiTaO₃ [28], [29] was about $\pm 0.01\%$.

As all wafers used in the following experiments were thinner than 0.5 mm, the influence of the waves reflected from the back surface of the specimen should be observed in the V_{LSAW} measurements. Therefore, this influence can be eliminated by using the moving average processing for the measured values of frequency dependences of V_{LSAW} [30].

IV. EXPERIMENTS

A. Specimens

Two groups of LiTaO₃ single crystal wafers produced by a manufacturer were taken as specimens.

In the first group, a total of 21 36°-rotated- Y -cut X -propagating (36° YX -) LiTaO₃ [31] wafers was prepared. These wafers were individually extracted from each of 21 ingots randomly chosen from crystal ingots with a charge number of 1 to 60 regardless of furnaces. However, the positions of the wafers within the ingots cannot be defined. The wafers are 100 mm in diameter, 0.35-mm thick, and optically polished on only one side.

In the second group, a total of 14 42° YX -LiTaO₃ [32] wafers was prepared. Five 84 to 86 mm long crystal ingots with charge numbers 1, 10, 21, 30, and 39 were selected from the 39 crystals successively grown in one furnace system. Three wafers were individually taken at three positions (top, middle, and bottom parts) of each crystal ingot boule. However, one wafer from the top of the charge number 21 ingot was lost because of a failure in the wafer preparation process. All the wafers were 100 mm in diameter, 0.5-mm thick, and optically polished on both sides. The T_C values for the top and bottom ends of each crystal ingot boule usually were measured using the differential thermal analysis (DTA) method [33] to evaluate the chemical compositions of crystals from this manufacturer.

B. LSAW Velocities

Velocities of LSAWs propagating along the crystallographic X axis for the 36° YX -LiTaO₃ wafers were measured using the LFB-UMC system at five measurement points positioned at intervals of 10 mm over a range of ± 20 mm in the diameter direction parallel to the X axis. The V_{LSAW} variations measured in each wafer ranged from 0.06 to 0.31 m/s. The relationship between the average values of the V_{LSAW} for each wafer and the charge numbers is shown in Fig. 1. The average V_{LSAW} values vary from 3126.14 to 3128.33 m/s, and thus it seems that the

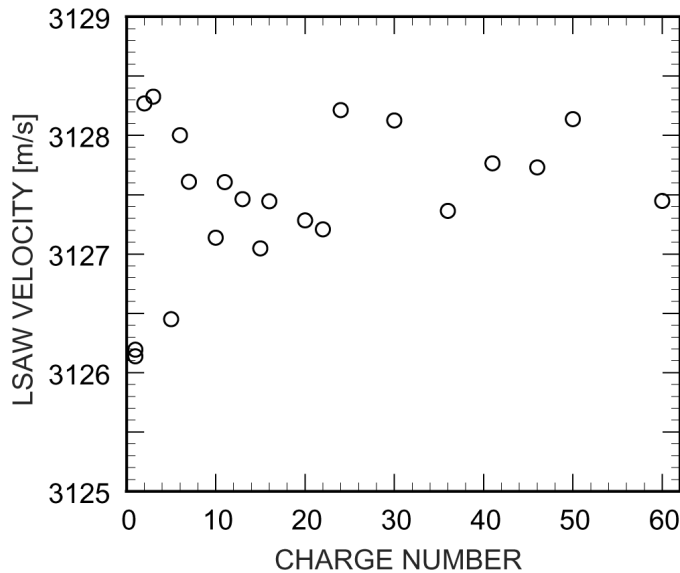


Fig. 1. LSAW velocity distributions with charge number for $36^\circ YX$ - LiTaO_3 wafers prepared from 21 crystals randomly selected in the mass-production line.

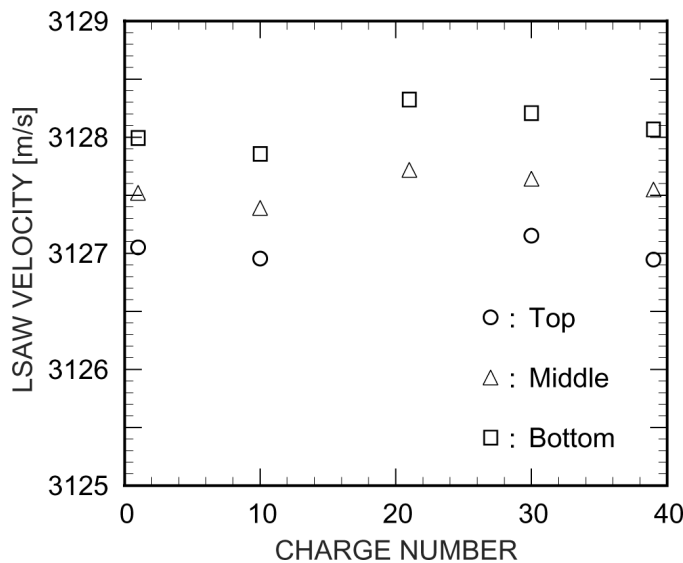


Fig. 2. LSAW velocity distributions with charge number for $42^\circ YX$ - LiTaO_3 wafers prepared from five crystals of a series of 39 crystals grown successively in one furnace.

charge numbers and V_{LSAW} might not be particularly related. The average V_{LSAW} for all the wafers is 3127.47 m/s with a standard deviation of 0.62 m/s and a maximum difference of 2.19 m/s.

Similarly, V_{LSAW} measurements for the $42^\circ YX$ - LiTaO_3 wafers along the X axis were measured at five measurement points. The V_{LSAW} variations measured on each of the wafers ranged from 0.06 m/s to 0.26 m/s. Fig. 2 depicts the relationship between the average V_{LSAW} values and the charge numbers. The averaged V_{LSAW} values for all wafers ranged from 3126.95 m/s to 3128.32 m/s. About a 1 m/s increase in velocity was observed from the top to the bottom of all the ingots. Fig. 2 reveals almost no dependence

TABLE I
MEASUREMENT SYSTEM AND CONDITIONS OF CURIE TEMPERATURES FROM THE MANUFACTURER.

Instrument	Rigaku TG-8120
Heating rate	40°C/min
Atmosphere	Air
Weight of sample (LiTaO_3)	112 mg
Particle diameter of sample	As powdered (<400 μm)
Weight of reference (Al_2O_3)	51 mg

of V_{LSAW} on the charge numbers, even for the crystals grown successively in one crucible. The average V_{LSAW} for all the wafers is 3127.60 m/s with a standard deviation of 0.44 m/s and a maximum difference of 1.38 m/s.

As seen from the above results, no particular changes in velocity and no significant velocity variations were observed, even though the examined crystals include crystals grown with very large charge numbers (maximum 60), as compared to the conventional crystal growth with the smaller charge numbers. This will be discussed further in a later section, but this is because the chemical compositions of the grown crystals were evaluated using T_C measurements of powdered samples obtained with a grinding machine from the tail of as-grown crystals; and, using these results, the chemical composition ratio was adjusted for the next crystal growth.

C. Curie Temperatures

The T_C was measured for powdered samples taken from the central part (20 mm ϕ) of each $36^\circ YX$ - LiTaO_3 wafer and each $42^\circ YX$ - LiTaO_3 wafer by the DTA method under the conditions shown in Table I. Fig. 3 illustrates the relationships between V_{LSAW} and T_C for each set of wafers. Squares and circles represent the results for the 36° (42°) YX - LiTaO_3 wafers. The solid lines represent approximated lines obtained by the least-squares method for each set of results. Dotted and dash-dotted lines represent the relationships calculated with the constants as a function of chemical composition in the literature [29] for 36° (42°) YX - LiTaO_3 , according to the analytical procedure of the LSAW propagation characteristics [34]. An ideal V_{LSAW} difference between $36^\circ YX$ - LiTaO_3 and $42^\circ YX$ - LiTaO_3 is about 0.08 m/s, which is almost as small as the reproducibility in the V_{LSAW} measurement. In Fig. 3, V_{LSAW} and T_C are proportional to each other. When considering the measurement accuracy in V_{LSAW} of better than ± 0.1 m/s, the deviations from the approximated lines are mostly due to the measurement accuracy of T_C . Most of the deviations of the measured values from the approximated lines are within $\pm 0.7^\circ\text{C}$. Also in Fig. 3, both the approximated lines for the measured values of $36^\circ YX$ - LiTaO_3 and $42^\circ YX$ - LiTaO_3 are about 0.3–0.8°C below in T_C from the calculated lines. The differences are thought to be caused by problems associated with the measurement conditions of T_C used during manufacture [27].

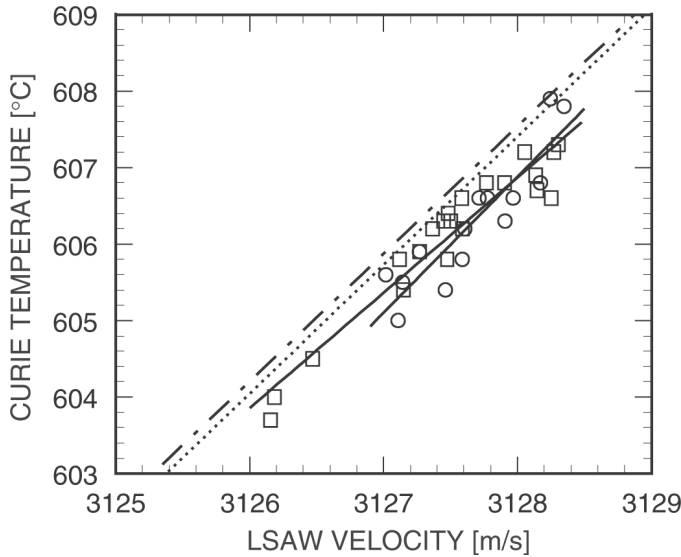


Fig. 3. Relationships between LSAW velocities and Curie temperatures. Squares: Measured for $36^\circ YX$ -LiTaO₃. Circles: Measured for $42^\circ YX$ -LiTaO₃. Solid lines: Approximated lines for each set of the measured values. Dotted line: Calculated for $36^\circ YX$ -LiTaO₃. Dash-dotted line: Calculated for $42^\circ YX$ -LiTaO₃.

V. DISCUSSION

A. Melt Composition

To estimate the melt compositions in mass production, we also used the accumulated T_C data for 1701 LiTaO₃ crystal ingots grown by the manufacturer for the past 4 years (from September 1998 to April 2001), in addition to the results shown in Figs. 1 and 2. The crystal ingots had different pulling axes such as X and $36^\circ Y$ different diameters of 3 and 4 in., and different crystal lengths ranging from 40 to 126 mm. The T_C was measured for powdered samples at the central area, 20 mm ϕ , of the top and bottom parts of each crystal ingot boule. In the estimate, T_C values at the middle position (43 mm from the top of crystals; average length of the crystals was about 86 mm) were obtained from the straight lines drawn between two T_C values at the top and bottom parts of each ingot, and they were converted into the Li₂O concentration $M(\text{Li}_2\text{O})$ in the melt, with V_{LSAW} as a universal scale using the predetermined relationships among the V_{LSAW} , T_C , and $M(\text{Li}_2\text{O})$ [15], [29] as:

$$M(\text{Li}_2\text{O}) = 0.14353 \times V_{\text{LSAW}} - 400.10, \quad (1)$$

$$M(\text{Li}_2\text{O}) = 0.08606 \times T_C - 3.43, \quad (2)$$

where V_{LSAW} is the value for $42^\circ YX$ -LiTaO₃ and both V_{LSAW} and T_C increase linearly with $M(\text{Li}_2\text{O})$. Fig. 4 shows the results. When the data are fitted to Gaussian distributions, the average melt composition is 48.77 Li₂O-mol% with a standard deviation $\sigma = 0.09$ Li₂O-mol% and a difference between the maximum and minimum of 0.75 Li₂O-mol%. Even though the melt composition ratios are adjusted using T_C results, the melt compositions as a whole are well distributed, including the recycled LiTaO₃

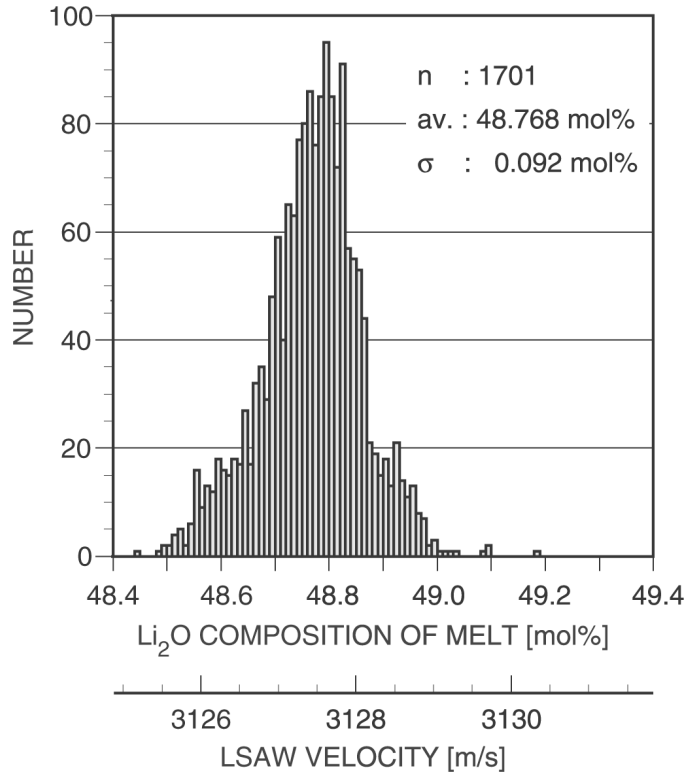


Fig. 4. Statistical distributions of melt compositions estimated from Curie temperatures that are related to LSAW velocities.

material and some Li₂CO₃ or Ta₂O₅ material that was added to the remaining material in the crucible to adjust the chemical composition ratio just after the previous crystal growth. This result coincides well with that obtained in Fig. 1.

B. Congruent Composition

The congruent composition should be defined for a melt from which crystals can be grown homogeneously along the pulling axis with the same chemical composition as the melt [6], [7]. Here, we try to estimate a congruent composition using the V_{LSAW} data obtained more accurately for the series of $42^\circ YX$ -LiTaO₃ crystals grown recently in Fig. 2, and to compare that composition with the result using the T_C data. Figs. 5(a) and (b) present the relationships among $M(\text{Li}_2\text{O})$, V_{LSAW} , and T_C at the top and bottom (normalized at 86 mm from the top) parts. In Figs. 5(a) and (b), straight lines fitted by the least-squares method to each set of experimental data were drawn with $M(\text{Li}_2\text{O})$ deviations of -0.007 to 0.013 mol% for the V_{LSAW} data and with $M(\text{Li}_2\text{O})$ deviations of -0.014 to 0.017 mol% for the T_C data. In Fig. 5(a), the melt composition at the intersection of two straight lines is 48.49 Li₂O-mol% corresponding to 3125.40 m/s from (1). In Fig. 5(b), it is 48.55 Li₂O-mol% to 604.0°C from (2). When considering the accuracies for V_{LSAW} and T_C , we can see from these results that the V_{LSAW} data obtained with higher accuracy can be useful for more accurately estimating the congruent composition. The estimated composition of 48.49 Li₂O-mol% is quite close to

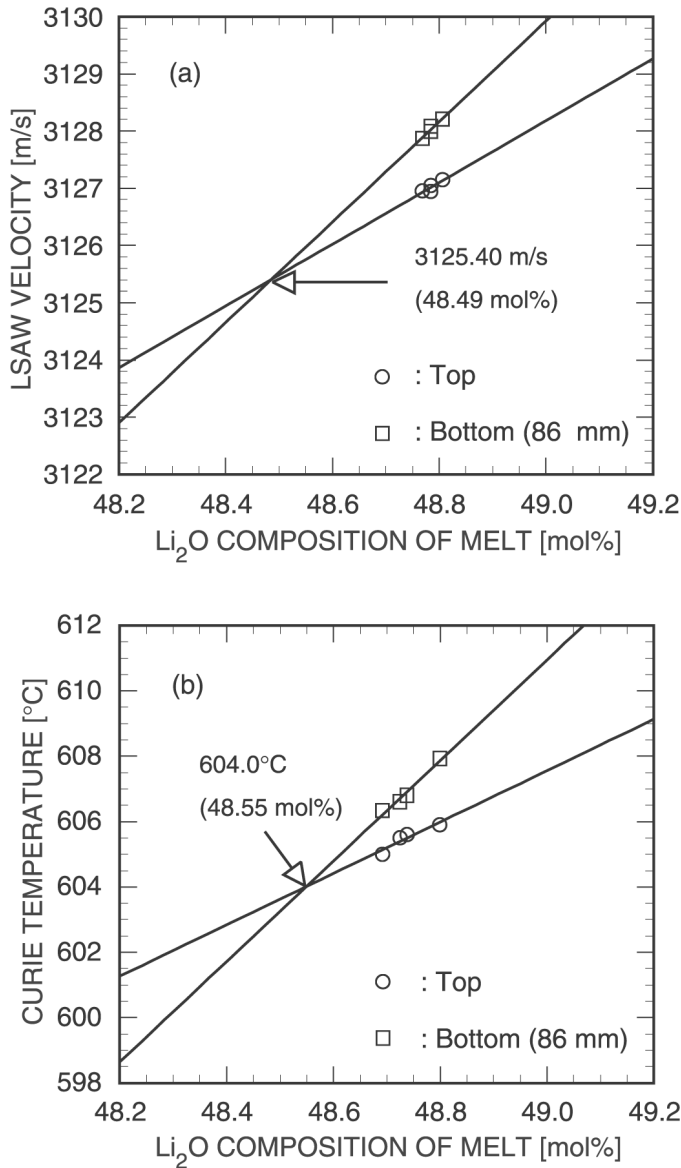


Fig. 5. Estimates of congruent compositions from LSAW velocities (a) and Curie temperatures (b) measured for $42^\circ YX$ -LiTaO₃ wafers.

48.47 Li₂O-mol% obtained by estimating the congruent composition for optical-grade LiTaO₃ [15], [26]. A more accurate estimation of the congruent composition could be made by repeating the same experimental procedure in crystal growth and V_{LSAW} evaluation around the 48.49 Li₂O-mol%. The manufacturer then will be able to produce crystals and wafers with excellent homogeneities in chemical composition using the starting material with the congruent composition determined and the recycled material. The corresponding T_C is about 602°C. Consequently, successive crystal growths with charge numbers exceeding 60 might be possible, if impurities are prevented from entering and mixing with the melt.

C. Control of Chemical Composition

According to the definition of a congruent composition, the composition of the grown crystal should be identical

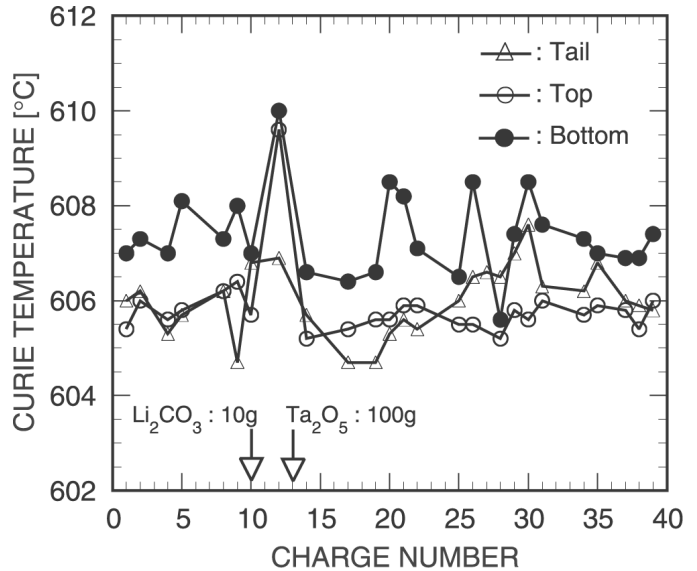


Fig. 6. Records of Curie temperatures obtained for a series of 39 $42^\circ YX$ -LiTaO₃ crystals and notes.

to that of the melt. So, the congruent composition estimated in Fig. 5 (about 48.49 Li₂O-mol%) must be used for growing crystals with higher homogeneity in chemical composition. However, the manufacturer was doing to control the melt compositions at about the 48.77 Li₂O-mol% analyzed in Fig. 4, which corresponds to a T_C of 606.5°C. The manufacturer might refer to the data of 48.75 Li₂O-mol% reported as a congruent composition in the literature [7]. It could be very useful to feed back the measured T_C in order to adjust the material composition for the next crystal growth, thus to some degree controlling changes in the chemical composition of grown crystals. However, the controlled T_C of 606.5°C is about 3°C higher than the T_C (603.2°C) for the congruent composition (48.49 Li₂O-mol%) estimated in Fig. 5. As a result, as shown in Fig. 2, the composition increases linearly from the top to the bottom with a velocity increase of about 1 m/s (corresponding to a chemical composition increase of 0.04 Li₂O-mol%) for each crystal ingot, because the melt composition has a slightly Li₂O-rich composition of about 0.3 mol%.

To study the results in Fig. 2, we examined the crystal production records. Fig. 6 presents the T_C data measured for a series of 39 grown crystals. There are two types of T_C data: powdered samples (particle size less than 20 μm) obtained from the tails of the 26 grown crystal ingots using a grinding machine and indicated by triangles, and powdered samples (particle size less than 400 μm) in the central area of 20 mm ϕ at the top (circles) and bottom (dots) parts for the crystal ingot boules processed for wafer fabrication. Thirteen crystals with no T_C data had serious defects, such as cracks, so the crystals were recycled directly for the next crystal growth. The T_C results for powder samples obtained from the ingot tails do not always coincide with those for powder samples obtained at the bottom parts; deviations range from -0.9°C to 3.3°C . It is very interesting in the notes that the T_C datum for the powdered

sample ground from the tail of the ninth crystal ingot was used to adjust the composition with an additional 10 g of Li_2CO_3 for the next crystal growth of charge number 10, because the measured T_C was about 1.5°C lower than that for the previous crystal ingot. Right after the addition of Li_2CO_3 , T_C increased rather more than expected (over 2°C). Therefore, the data was fed back again to adjust the composition with an additional 100 g of Ta_2O_5 for the thirteenth crystal growth in order to decrease T_C to about 606°C . After that, there was no control in composition, although the T_C results in the tails exhibited some variations with the charge numbers in this series of the crystals. This means that there is no assured guideline to control the chemical composition. The T_C values were 1 or 2°C higher at the bottom parts, corresponding to the fact that the melts were not really congruent but were Li_2O -rich. It seems that variations in T_C were caused by the lower accuracy of T_C than of V_{LSAW} . The differences between the T_C data at the ingot tails and at the bottom parts could be related to the effect of the different particle sizes as one of the error factors for T_C measurements. From the V_{LSAW} variations observed in Fig. 2, we can understand the practical implications of production control by T_C measurements.

D. T_C Measurement Conditions

Measured T_C values are influenced by the measurement conditions, such as the heating rate, and the particle diameter and weight of powdered specimen and reference materials [27]. It was evident that the differences in T_C observed were due to the difference in particle sizes between the two kinds of powdered samples from the tail of as-grown crystal ingots obtained with a grinding machine and from the bottom parts by grinding them with a pestle and mortar. We also examined another factor associated with measurement errors, the measurement environment of room temperatures. Fortunately, we have records of room temperatures (T_R) and $T_C(\alpha\text{-SiO}_2)$ data for synthetic quartz for checking the DTA systems made at the first measurement every working day. The phase transition temperature of the crystal is 573°C [35], very close to the T_C of congruent LiTaO_3 single crystals (about 600°C). The results of our study on evaluating previous piezoelectric materials [14], [26], [36] have proven that synthetic quartz used for SAW devices is especially remarkable in its homogeneity and can be expected to be a material for a stable standard specimen. Fig. 7 presents the results of 537 data collected over 27 months (from January 2000 to March 2002), with a solid line, $T_C(\alpha\text{-SiO}_2) = -0.1346T_R + 578.2$, fitted to the data. The room temperatures in winter were a bit lower than 20°C , and slightly higher than 25°C in summer. The average room temperature was about 22.45°C , with a maximum deviation of 18.2°C . The corresponding measured T_C variations were manifested as higher T_C in winter and lower T_C in summer, with an average T_C of 575.18°C and a maximum deviation of 2.3°C . This could be because the T_C measurement conditions produced T_C values, a few

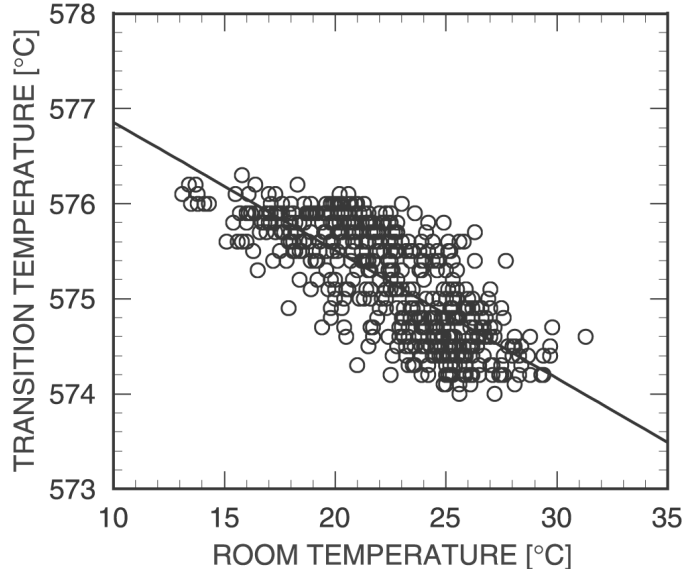


Fig. 7. Records of phase transition temperatures for synthetic quartz and room temperatures.

degrees higher than the well-known phase transition temperature of 573°C for quartz. We can estimate from this figure that the T_C measurement errors introduced by the manufacturer might be $\pm 1^\circ\text{C}$ at an average T_R of 22.45°C with additional errors in maximum deviations of $\pm 1.2^\circ\text{C}$, depending on the T_R .

This suggests a guideline for precise measurements of T_C . The measurement environment is also a very important factor, and the room temperatures should be controlled within $\pm 2^\circ\text{C}$ to reduce the T_R dependence to less than $\pm 0.3^\circ\text{C}$ in the T_C measurements. The use of synthetic quartz as a reference is also a good idea for checking T_C measurement systems that can be easily included in the measurement routine, although we have already proposed a more accurate calibration method using V_{LSAW} [27]. Thus, careful attention is required in the measurement, and the measurement conditions must be investigated in any further discussions about accuracy.

VI. CONCLUSIONS

In this study, we applied the LFB-UMC system to evaluate a mass-production line of LiTaO_3 single crystals for SAW devices in a manufacturer that uses a maximum charge number of 60 to grow crystals. We measured Rayleigh-type V_{LSAW} for $36^\circ\text{YX-LiTaO}_3$ and $42^\circ\text{YX-LiTaO}_3$ wafers, as well as T_C , then compared them. This study revealed that the average V_{LSAW} was 3127.52 m/s with a maximum difference of 2.2 m/s, that each crystal ingot had a velocity increase of about 1 m/s from the top to the bottom, and that there was no dependence on the charge number. Because the average velocity corresponded to the chemical composition of 48.77 Li_2O -mol%, we could understand that the manufacturer was producing the crystals grown from the melt composition controlled around

that value through T_C measurements. We also estimated the congruent composition to be 48.49 Li₂O-mol% from V_{LSAW} distributions along the crystal pulling-axis direction obtained for a series of the 42°YX-LiTaO₃ crystal ingots. The congruent composition was about 0.3 Li₂O-mol% less than the melt composition controlled for the current mass production by the manufacturer. A more accurate estimate of the congruent composition will be made by further experiments using V_{LSAW} measurements in a similar way. This investigation revealed that the measurement conditions associated with T_C measurements mainly used for chemical composition analysis in industry, including the measurement environment of room temperatures, must be reconsidered for reliable production control.

Once we obtain a true congruent composition using this ultrasonic method, it is easy to grow homogeneous crystals from the starting material with a true congruent composition and their recycled material (such as failed crystals and material remaining at the top and bottom parts of the crystal ingots), easily satisfying the required condition of a tolerance of $\pm 0.04\%$ in SAW velocity [11] because the corresponding tolerance for T_C is $\pm 1.6^\circ\text{C}$ [17]. We suggest that in the near future it will be possible to simplify the evaluation procedure after the establishment of crystal growth conditions for each furnace, probably eliminating the need to measure V_{LSAW} and T_C . Also, from the technical point of view in the acoustic properties, it may be possible to grow homogeneous crystals with charge numbers of over 60, leading to lower cost and improvement in productivity, especially if there is no problem of impurities being incorporated during mass production. In general, these results also suggest to other manufacturers that the industrial conditions (especially chemical composition) for growth of LiTaO₃ and LiNbO₃ crystals should be re-examined for more efficient production of crystals with better homogeneity.

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