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Measurement of Thermal Expansivity of Low-Expansion Glasses by Interferometric Methods: Results of an Interlaboratory Comparison

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An interlaboratory testing program on the measurement of the linear thermal expansion coefficient of low-expansion glasses has been carried out. Three different types of interferometric dilatometers, each located at three different organizations, and two kinds of low-expansion glass materials were selected for the experiments. As a result of the comparison, a reasonable agreement among the different measuring instruments was confirmed, and it was determined that the thermal expansion coefficient for low-expansion glasses can be measured with an accuracy of $\pm 4 \times 10^{-8}\,K^{-1}$ by using commercially available interferometric dilatometers.

KEY WORDS: coefficient of linear thermal expansion; dilatometry; interferometric dilatometer; low-expansion glasses; reference method; thermal expansion; standardization.

1. INTRODUCTION

In the new glass industry, several kinds of materials which have extremely low linear thermal expansion coefficients have been developed. These are used for the construction of precision instruments, optical components, etc. The accurate determination of the linear thermal expansion coefficient is one of the most important requirements for the development of new material industries.

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The most accurate method of measuring linear thermal expansion is considered to be the application of laser interferometry. Therefore, many kinds of new interferometric dilatometers have been developed and used in laboratory experiments [1–5]. Moreover, some commercially available measuring instruments which utilize laser interferometry have also appeared recently.

In this situation, an interlaboratory testing program suported by the Association of New Glass Industry (New Glass Forum), Japan, was conducted to compare the results obtained by different types of interferometric dilatometers. Two kinds of low-expansion glasses were selected for the test specimens and were supplied by their manufacturers. Three organizations, each having a different type of laser interferometer, participated in the interlaboratory comparison program and measured the expansivity in accordance with the same measuring conditions. The results of the present intercomparison are analyzed statistically and the possibility of standardization of the measuring method for low-expansion glasses by means of laser interferometric dilatometers is suggested.

2. MEASURING APPARATUS

A survey on the need for measurements of the linear thermal expansion coefficient (or thermal expansivity) of low-expansion materials was carried out for both manufacturers and users of measuring instruments [6]. From the analysis of this survey, it was determined that there is an urgent requirement for the standardization of a measuring method for linear thermal expansion coefficient in the new glass industry. From among many new material industries, a steering committee on this requirement has been organized in the New Glass Forum under the supervision of the Agency of Industrial Science and Technology, the Ministry of International Trade and Industry, Japan.

Although there exist many kinds of measuring methods which involve laser interferometry, most of them are adopted only for laboratory experiments. Two types of commercially available interferometric dilatometers have been developed and are commonly used in both factories and laboratories in Japan. Therefore, an interlaboratory comparison program has been planned involving different types of measuring apparatus and organizations.

In addition, an interferometric method which utilizes both double-path interferometry and optical heterodyne detection was chosen for the present intercomparison as a reference method. This method has been developed previously at the National Research Laboratory of Metrology, Japan. The accuracy of this method has been established by measuring the linear

thermal expansion coefficients of several standard reference materials and low-expansion materials [5, 7]. As a result of a preliminary survey, three different types of interferometric dilatometers were selected for the present interlaboratory comparison. Each dilatometer consisted of a He–Ne laser interferometer, a specimen holder, a temperature enclosure with controller, a vacuum system, temperature measuring sensors, and a recorder for both length and temperature changes.

The first type of dilatometer is a kind of Fizeau interferometer (Type I dilatometer), as shown in Fig. 1. The two mirrors are positioned at the ends of three specimens (their length is the same) and the interference fringes are detected by a TV camera. The measurement is carried out with the specimen under vacuum.

Figure 2 shows the principle of the second type of interferometer (Type II dilatometer). This is a so-called double-path interferometer whose merit is that the sensitivity is twice of that of a single-path interferometer. Another merit of this dilatometer is that stringent parallelism is not required for the specimen ends because of the utilization of parallel spring movement in the specimen holder. In this system, the shift in fringes is detected by an image sensor and the length change of the specimen is calculated with a resolution of 0.02 μm by a microcomputer [8]. The temperature of the specimen is measured by chromel–alumel thermocouples in both type I and type II dilatometers.

Figure 3 shows the principle of the third type of interferometer (Type III dilatometer), which is introduced as a reference method. In this

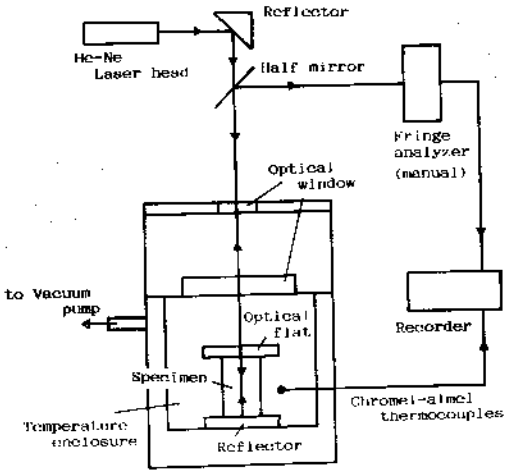

Fig. 1. Schematic diagram of the Type I dilatometer.

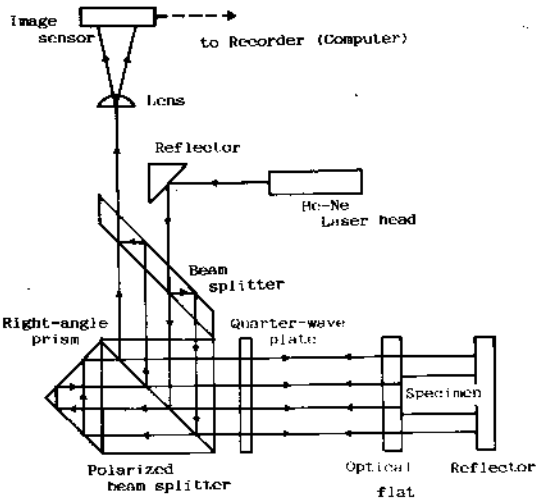

Fig. 2. Optical arrangement of the Type II dilatometer.

interferometer, a double optical path and an optical heterodyne detection system are adopted. Another merit is the introduction of symmetrical layout of the optical components [7] as is seen in Fig. 3. Therefore, fine sensitivity and self-compensation for optical misalignment caused during the sequential measurement can be achieved. Platinum resistance thermometers are used for both controlling and measuring the specimen

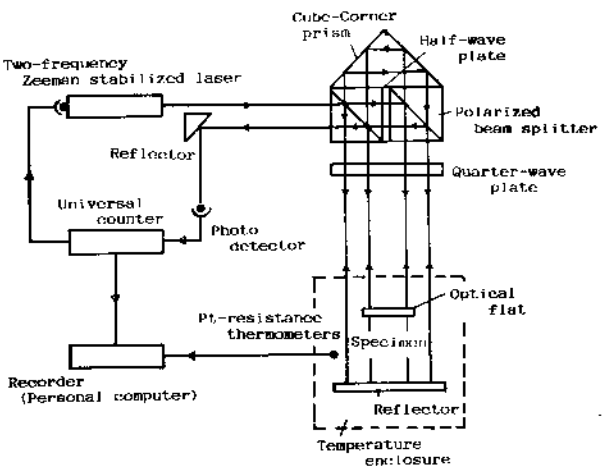

Fig. 3. Schematic diagram of the Type III dilatometer.

temperature in this system. The measurement of thermal expansion is carried out under vacuum as in the case of the above mentioned two types of dilatometers. The long-term stability of this interferometer was confirmed to be better than 2 nm (10 hr), and zero-drift was also estimated to be less than $4 \times 10^{-9}\ K^{-1}$ for a 5-cm-long specimen [9].

3. EXPERIMENTAL DESIGN

3.1. Selection of Materials

In order to confirm the measurement uncertainty of the methods introduced in the present intercomparison, well-characterized and stable specimens have to be used. Therefore, special care was devoted to the selection and preparation of the specimens. Among the possible candidate materials, two kinds of low-expansion glasses were chosen, as shown in Table I. These materials were kindly supplied by their manufacturers. The characterization of these two materials has already been established and both materials are considered to be suitable for the present stringent requirements from the viewpoint of homogeneity, stability, and handling [10].

Specimens used for the present interlaboratory comparison were carefully treated according to the manufacturers' guidelines to eliminate such efects as hysteresis, drift, etc. The form and size of specimens were also selected in order to fit each dilatometer. Test specimens to be measured at each organization were cut from the same billet and were distributed after suitable treatment. The flatness and parallelism at both ends of each specimen to be measured were fabricated to better than a quarter-wavelength of the light source and 5 s of arc, respectively.

3.2. Measuring Conditions

The main objective of the present interlaboratory comparison is to confirm the agreement of the measurement results obtained by partici-

Table I. Materials Used for the Experiments

Material	Manufacturer	Nominal LTEC[a] at 20°C
ULE	Corning	$-5 \times 10^{-8}\ K^{-1}$
Zerodur	Schott	$(-4 \rightarrow -9) \times 10^{-8}\ K^{-1}$

[a] Linear thermal expansion coefficient.

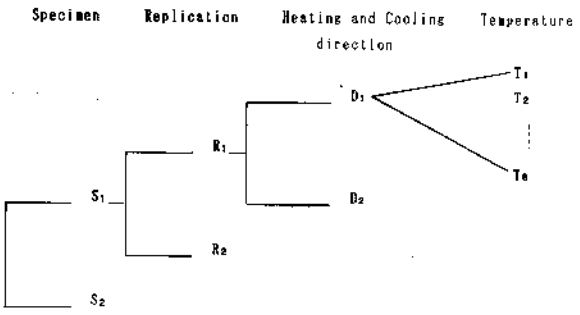

Fig. 4. Experimental design of the measurement.

pating organizations, each using a different interferometric dilatometer. The stability of the test specimens and the effect of measuring conditions are also to be examined. The measurement procedure was unified and the experiment was replicated twice at each organization for two specimens of each material.

Test temperatures were set up between −150 and 150°C at an interval of 50°C. The effect of heating and cooling rate of the specimen was also investigated. In all participating organizations, the heating rate of the specimen was controlled to be less than 0.5°C min $^{-1}$. Before each measurement, the specimen was kept at a given temperature for a time sufficient to achieve equilibrium.

Figure 4 shows the experimental layout of the present intercomparison. In the figure, S, R, D, and T represent the significance of the effects of specimen, replication of measurement, heating/cooling direction (hysteresis), and test temperature, respectively. By carrying out the measurement in accordance with this experimental design, not only the main effects but also the interactions among S, R, D, and T can be investigated.

4. EXPERIMENTAL RESULTS AND DISCUSSIONS

In order to investigate the variation of the measurement results, statistical analysis was introduced. Table II shows the result of an analysis of variance on the thermal expansion coefficient data obtained by the second organization (Type II dilatometer) when ULE specimens were measured. Table III is the result of an analysis of variance on similar data obtained by the third organization (reference method) when Zerodur specimens were measured. Although the significant effect of the test temperature is seen in both tables, no significant difference is detected for the

Table II. Analysis of Variance Table for Data on the Thermal Expansion Coefficient of ULE Specimens (Type II Dilatometer)

Source of variation	Sum of squares $(10^{-8}\,K^{-1})^2$	df	Mean square $(10^{-8}\,K^{-1})^2$
R: replication	11.6024	1	11.6204
D: H/C direction	0.0004	1	0.0004
E_1: 1st-order error	0.2604	1	0.2604
T: temperature	13010.1838	5	2602.0368
$R * T$	48.9371	5	9.7874
$D * T$	34.9371	5	6.9874
E_2: 2nd-order error	11.9671	5	2.3934
(E_2')	(95.8413)	(15)	(6.3894)

other factors including interactions among main effects. Similar analyses were also done for other combinations of materials and organizations.

Figure 5 shows the change of specimen length for the two kinds of glass materials in the temperature range between −150 and 150°C. The coefficients of the linear thermal expansion measured at the three organizations are shown in Fig. 6 (ULE) and Fig. 7 (Zerodur), respectively. For the comparison among different types of interferometric dilatometers (i.e., organizations), the deviations from the mean values are shown in Figs. 8 and 9, respectively. From these figures, no significant difference among dilatometers is seen for both glasses if the confidence interval of the measurement error is taken into consideration (see below). The results shown in Figs. 8 and 9 indicate that the thermal expansion coefficients measured by the three interferometric dilatometers are in good agreement.

Table III. Analysis of Variance Table for Data on the Thermal Expansion Coefficient of Zerodur Specimens (Type III Dilatometer)

Source of variation	Sum of squares $(10^{-8}\,K^{-1})^2$	df	Mean square $(10^{-8}\,K^{-1})^2$
R: replication	0.1504	1	0.1504
D: H/C direction	0.0004	1	0.0004
E_1: 1st-order error	0.3038	1	0.3038
T: temperature	1252.2471	5	250.4494
$R * T$	2.4471	5	0.4894
$D * T$	3.9171	5	0.7834
E_2: 2nd-order error	0.8838	5	0.1768
(E_2')	(7.2480)	(15)	(0.4832)

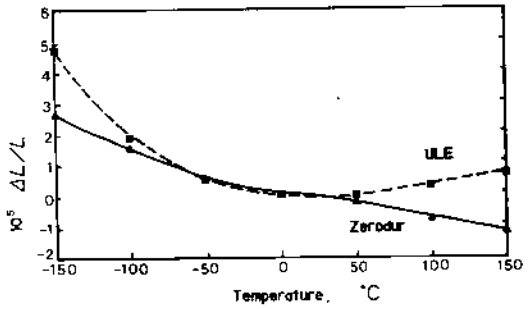

Fig. 5. Linear thermal expansion $\Delta L/L$ of ULE and Zerodur. The symbols ■ and ▲ are these average values from three different dilatometers. The curves —— and – – – are merely a guide to the eye.

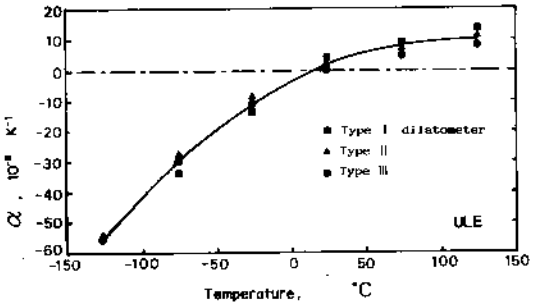

Fig. 6. Linear thermal expansion coefficient α of ULE. The curve —— is merely a guide to the eye.

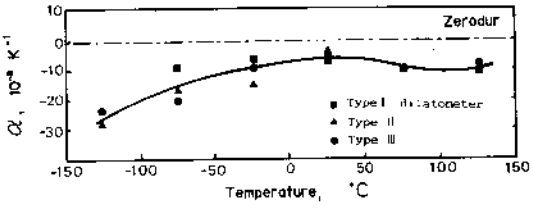

Fig. 7. Linear thermal expansion coefficient α of Zerodur. The curve — · — is merely a guide to the eye.

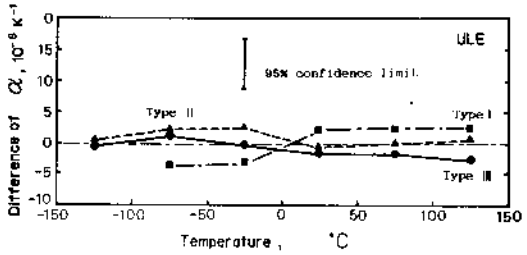

Fig. 8. Difference of the thermal expansion coefficient obtained by different types of dilatometers for ULE specimens from the mean value.

In the present intercomparison, a reference method was introduced and this is considered to be an absolute measuring method which does not give significant bias for the measurement of the linear thermal expansion coefficient. Furthermore, the results obtained by two types of commercially available interferometric dilatometers show no significant difference with respect to those obtained by the above reference method.

Usually the measurement uncertainty can be represented as a combination of bias from the true value and the precision expressed by a dispersion under the reproducibility condition. According to the above investigation, the measurement uncertainty of the thermal expansion coefficient in the present intercomparison may be calculated from Tables II and III as a 95% confidence interval for the effect of replication of measurement. For example, in Table II, as there is no significant interaction, the second-order error $V_{E2'}$ is calculated from the sum of squares of the variations, $S_{R\times T}$, $S_{D\times T}$, and $S_{R\times D\times T}$, by the following procedure:

$$V_{E2'}=(S_{R\times T}+S_{D\times T}+S_{R\times D\times T})/(\phi_{R\times T}+\phi_{D\times T}+\phi_{R\times D\times T})$$

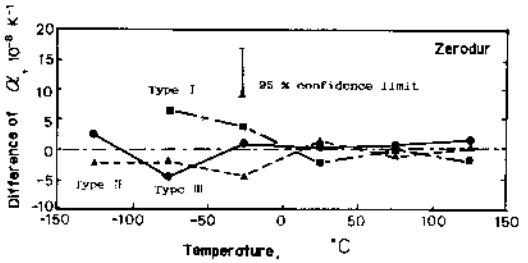

Fig. 9. Difference of the thermal expansion coefficient obtained by different types of dilatometers for Zerodur specimens from the mean value.

Table IV. Reproducibility (95% Confidence Limit) of Measurement

Dilatometer	Reproducibility (10^{-8} K^{-1})	
	ULE	Zerodur
I	4.0	3.8
II	3.8	3.0
III	1.0	1.0

where, $\phi_{R\times T}$, $\phi_{D\times T}$, and $\phi_{R\times D\times T}$ are the degrees of freedom for interactions $R\times T$, $D\times T$, and $R\times D\times T$. Finally, a 95% confidence interval is obtained as follows:

$$t(\phi_{E2'}; 0.05)\sqrt{V_{E2'}/n_e}$$

where t is the value of 5% significance level in the Student's t table for degrees of freedom of $\phi_{E2'}$, and n_e is the effective number of replications. In the present experiment, as the measurement was carried out by both heating and cooling the specimen at each test temperature, n_e is equal to 2. Following the above procedure, the 95% confidence limits are obtained for Types II and III dilatometers, by using the results in Tables II and III, as follows:

$$\text{Type II:} \quad t(15; 0.05)\sqrt{V_{E2'}/n_e} = 2.131\sqrt{6.3894/2} = 3.8\ (\times 10^{-8}\,\text{K}^{-1})$$

$$\text{Type III:} \quad t(15; 0.05)\sqrt{V_{E2'}/n_e} = 2.131\sqrt{0.4832/2} = 1.0\ (\times 10^{-8}\,\text{K}^{-1})$$

The results of the calculation are represented in Table IV for each type of dilatometer (organization) and material; it may be seen that the measurement uncertainty for commercially available interferometric dilatometers is less than or equal to $\pm 4\times 10^{-8}\,\text{K}^{-1}$. This measurement accuracy is not necessarily sufficient for precise measurements on materials whose linear thermal expansion coefficient is nearly equal to zero. However, it may be adequate for the urgent requirements of many industrial applications of new glasses.

5. CONCLUSION

An interlaboratory comparison on the measurement of the linear thermal expansion coefficients for low-expansion glasses has been carried out

using three different types of interferometric dilatometers. The measurement results for two kinds of materials showed a good agreement among participating organizations, and it was determined that measurements can be performed to within an uncertainty of about $\pm 4 \times 10^{-8}\,K^{-1}$, with commercially available interferometric dilatometers.

From the above investigation, the possibility of standardization of the measurement method for linear thermal expansion coefficient by means of interferometric dilatometers has been suggested.

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