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V-2

The Precise Measurement of the Thermal Expansion Coefficient of Glass by Interferometric Dilatometer

T. KISHII (*)

Tokyo Shibaura Electric Co., Tokyo (Japan)

La mesure précise du coefficient de dilatation des verres au dilatomètre interférentiel.

SOMMAIRE

En vue d'une mesure précise du coefficient de dilatation thermique des verres, l'auteur a utilisé un dilatomètre interférentiel et a essayé d'améliorer l'appareil ainsi que le mode opératoire.

La mesure préliminaire de la longueur de l'éprouvette est faite en utilisant une éprouvette auxiliaire et l'on observe moins de trois franges d'interférence, lorsque les deux éprouvettes sont introduites entre les surfaces optiques. Cette façon de faire permet de déterminer la longueur de l'éprouvette avec une précision de 2-3 μ lorsque la longueur de l'éprouvette auxiliaire est mesurée à l'aide d'un palmer à vis.

La mesure exacte de l'éprouvette est faite après son introduction dans l'interféromètre en utilisant simultanément les raies Hg (V) 5 460, 7 436 et Cd (R) 6 436, 4 696 Å.

L'ordre de la frange peut être déterminé exactement en mesurant au micromètre oculaire les distances entre deux franges voisines données par les deux raies.

On trouve qu'il existe entre l'ordre de la frange, n, et la longueur d'onde λ relatives aux deux raies utilisées une relation telle que :

$$a_{RV} = \frac{n_R \lambda_R - n_V \lambda_V}{\lambda_V} = 0{,}55.$$

Les ordres des franges peuvent être déterminés sans ambiguïté si l'on recherche les valeurs appropriées de n par tâtonnement jusqu'à ce que la relation ci-dessus soit satisfaite. Si l'on multiplie la moitié de la longueur d'onde par l'ordre de la frange déterminé de cette manière, il est possible de calculer la longueur de l'éprouvette avec une précision de 0,015 μ.

Le déplacement des franges aux températures T_1 et T_2 a été observé et on en a déduit la modification de la longueur de l'éprouvette, puis la valeur du coefficient de dilatation. La reproductibilité a été trouvée égale à $\pm 0{,}2 \times 10^{-7}$ par degré C.

Cette méthode, qui offre la possibilité de mesurer la dilatation avec une grande précision dans un grand domaine de température, a permis de déceler l'existence de quelques anomalies nouvelles du coefficient de dilatation des verres à des températures inférieures à 300 °C.

Genaue Messung des Ausdehnungskoeffizienten des Glases mit Hilfe des Interferenz Dilatometers.

ZUSAMMENFASSUNG

Der Autor beschreibt Verbesserungen in Konstruktion und Anwendung eines Interferenz-Dilatometers.

Die Probenlänge wurde einleitend mit Hilfe einer Hilfsprobe ermittelt, derart dass weniger als drei Interferenzstreifen beobachtet wurden, wenn man beide Proben zwischen die optischen Flächen legte. Auf diese Weise kann man die Probenlänge mit einer Genauigkeit von 2-3 μ bestimmen, wenn die Länge der Hilfsprobe durch ein Schraubenmikrometer gemessen ist.

Nach Einführung in das Interfereometer wurde die exakte Probenlänge durch gleichzeitige Beobachtung der Spektrallinien Hg (G) 5 460, 7 430 und Cd (R) 6 438, 4 696 Å gemessen. Die Ordnung der Streifen kann genau ermittelt werden, wenn die Abstände zwischen zwei benachbarten Streifen für beide Spektrallinien durch ein Ocularmikrometer gemessen werden.

Da zwischen der Streifenbreite a und der Wellenlänge λ der entsprechenden Spektrallinien die Beziehung

$$a_{RG} = \frac{n_R \lambda_R - n_G \lambda_G}{\lambda_G} = 0{,}55$$

besteht, können die Streifenbreiten genau bestimmt werden, wenn die n-Werte durch Probieren richtig gewählt sind, sodass sie der angegebenen Beziehung genügen. Wenn die halbe Wellenlänge mit der so bestimmten Streifenbreite multipliziert wird, soll es möglich sein, die Probenlänge mit einer Genauigkeit von 0,015 μ zu berechnen.

Bei zwei Temperaturen, T_1 und T_2, wurde die Verschiebung der Interferenzstreifen beobachtet und daraus die Änderung der Probenlänge und schliesslich der Ausdehnungskoeffizient berechnet. Die Reproduzierbarkeit betrug $\pm 0{,}2 \times 10^{-7}$ °C.

Die Methode erlaubt die Präzisionsmessung der Ausdehnung in einem ziemlich grossen Temperaturbereich. Es wurden einige bisher unbekannte Anomalien im Verlauf des Ausdehnungskoeffizienten von Gläsern bei Temperaturen unterhalb 300 °C entdeckt

(*) B. Sc. - Staff member of Glass Engineering Section, Lamp and Tube Manufacturing Department, Tokyo Shibaura Electric Co.

INTRODUCTION

Since the invention of an interferometric dilatometer the instrument as well as the procedure have successively been improved to obtain better accuracy, and now the automatic recorders are quite usual [1, 14].

Being an important property especially for the process of sealing, the expansion of glass is measured in various ways and is used in many glass factories as a routine test which provides a key for carrying on the quality control.

Owing to the ever increasing demand for constant glass quality the control limit is becoming narrower and narrower, and it is now necessary to check frequently the conventional forms of apparatus for accuracy by comparing with the results obtained by a more exact instrument. Although it was found that an interferometric dilatometer provides sufficiently accurate results to meet the requirement, the current procedures would have to be improved by improving the method of measuring the sample length and by securing a more uniform temperature distribution throughout the sample. In most cases one would expect the errors to be caused by surface films, condition of setting as well as the shape of samples.

A series of systematic studies have been carried out in order to establish methods which will reduce the expected errors to a minimum.

Having developed the new method successfully the author measured the expansion of fused silica, borosilicate, potassium-soda-lead, and of soda-lime-magnesia glass. By virtue of high accuracy it was possible to provide a positive proof of the existence of anomalies in the expansion coefficients whose behaviou depends on the thermal history of the glass. Although the transition points below 300 °C have already been suggested by some authors the present results would be of some service for considering the glass structure in the light of recent theories.

1. — APPARATUS

The apparatus consists essentially of two interferometer plates made of polished Pyrex brand glass placed either side of a tubular test piece whose longitudinal dimentional change was determined by the optical path difference between two pencils of light ray each reflected at the surface of the interferometric plates. The optical path difference gives rise to interference fringes at the ocular micrometer by which the distance between two fringes of subsequent order is measured. The change of this distance is directly related to the change of the length of the specimen. The use of this principle generally eneables the measurement of length to be made to a degree of accuracy higher than that attained by any other method. The general arrangement is reproduced schematically in *figure* 1 which in many points works in the same way as the MERRITT's instrument [9] except some minor changes, being introduced for using, successively, the different light sources, and for minimizing the temperature difference throughout the test piece.

Monochromatic light was obtained by the combination of some special lamps combined with two filters whose transmission curves are reproduced in *figure* 2. The lines of Hg-green 5 461 Å (g), Hg-violet 4 358 Å (v), Cd-red 6 438 Å (R), and Cd-green 5086 Å (G) were used (1).

An electric tubular furnace in which the sample rests in a thick aluminium cup at the centre was provided with a thermostat which, with the aid of a subsidiary thermo-junction, made it possible to heat up to the desired temperature very quickly, and to keep constant at that temperature for a long time.

Aluminium was found to be an appropriate material for the specimen container [10] since its thermal conductivity, being about ten times of that of stainless steel, makes the temperature distribution sufficiently uniform, while the rate of oxidation is very low as long as it is used below 400 °C.

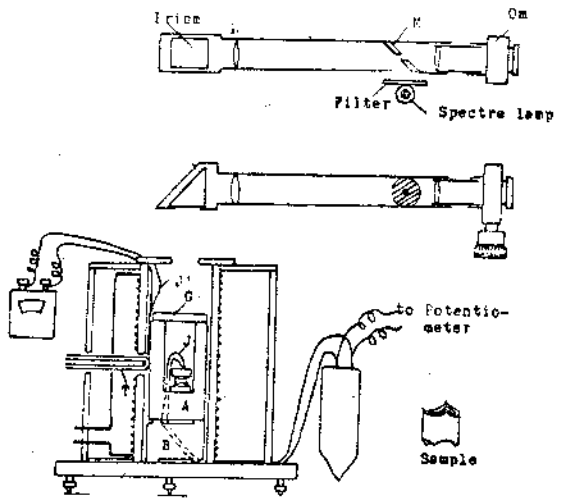

FIG. 1.—Apparatus.

L: Lens f = 34 cm.
Om: Ocular micrometer.
M: Mirror with slit and diaphragm.
JJ': Pt-Pt 13 % Rh thermo-junction.
G: Glass plate.
B: Platform (refractory).
A: Aluminium cup 70 ø × 100 (hole 40 ø × 60).
T: Thermistor.

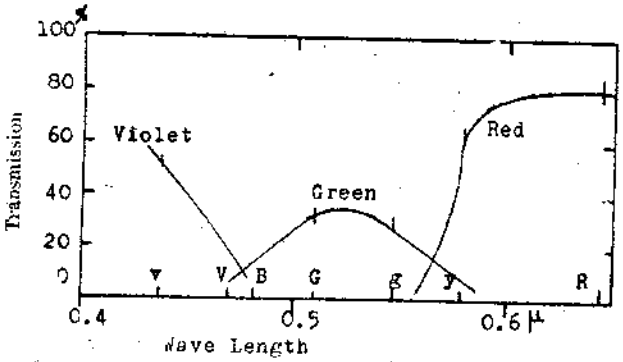

FIG. 2.—Transmission Curves of Filters.

The maximum temperature difference measured by a differential thermo-junction at the sample's surface has proved to be less than 0.5 °C when the temperature was kept constant at 300 °C, and less than 1.5 °C when it was raised at a rate of 1.5 °C/mn. The temperature fluctuation was found to be less than ± 0.2 °C during an hour's observation, which increased to about ± 0.6 °C if the observation was prolonged for several hours.

(1) The spectral lines are abbreviated respectively as g, v, R and G.

2. — PREPARATION OF TEST PIECE

Samples were prepared from tubing drawn, by hand or by machine, having a diameter of 10-15 mm. The tube was 10 mm long, and was polished at both ends by an oil stone until the interference fringes were observed when placed between the interferometer plates. It was then washed in a hot aqueous solution of sodium carbonate followed, successively, by water cleansing, drying at about 100 °C, and polishing with dry, fine alundum powder until the interference fringes of sufficiently wide pitch was observed. As a finishing touch the polished tube was worked so that it has three points at each end.

This rather complicated method of preparing the sample has proved to be necessary to eliminate the effect of surface films [2, 10]. It was also confirmed that no measurable change occured in the length of specimen by drying at about 100 °C.

3. — METHOD OF MEASUREMENT

The present method consists of simply measuring the sample length at two temperatures, say, T_1 and T_2 with the highest accuracy so that the mean thermal expansion may be calculated. Therefore it is of prime importance to know the exact values of temperature and of the length of the specimen.

a) Measurement of temperature.

The combination of Pt-Pt 13 % Rh thermo-junction with a potentiometer made it possible to read the temperature to 0.05 °C. The precision, however, might be regarded as 0.6 °C at 300 °C and 0.1 °C at room temperature taking into account of the slightly uneven temperature distribution at the surface.

b) Measurement of sample length.

In order to form the basis of the measurement the sample length has to be measured with sufficient accuracy. For this purpose the author used a subsidiary piece whose length was proved to agree with that of the sample within three wave lengths when both were brought together between the interferometer plates. The arrangement is shown in *figure* 3.

For the exact evaluation of the sample length with known accuracy the author used the coincidence method [15, 17] originally introduced by SAUNDERS in the measurement of the interference fringes of equal thickness [13], with some modifications.

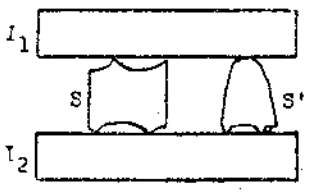

FIG. 3.—Comparison of the sample length with that of subsidiary piece.
I_1, I_2: Interferometer plates.
S: Sample.
S': Subsidiary piece.

In the first place the sample was placed between two interferometer plates as shown in *figure* 1, and the distance between the arbitrary selected dark parts of the interference fringes of several spectral lines (g 5 461 Å, G 5 086 Å, R 6 438 Å, v 4 358 Å) was measured by the ocular micrometer. In order to know the order of the interference fringes a point

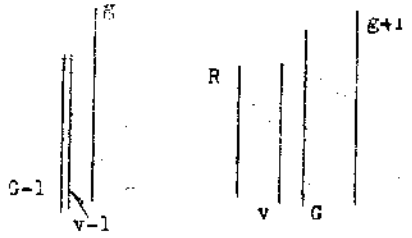

FIG. 4.—The determination of the relative position of interference fringes by R (Cd Red), G (Cd Green), g (Hg Green), v (Hg Violet) lines.

Fringe order.
$g = 40\ 100$
$R = 34\ 011$
$G = 43\ 057$
$v = 50\ 244$

$$a_{Rg} = \frac{\text{distance } (R, g)}{\text{distance } (g+1, g)} = 0{,}55 \qquad a_{RG} = \frac{\text{distance } (R, G-1)}{\text{distance } (G, G-1)} = 0{,}72$$

$a_{Gg} = 0{,}81$ $\qquad a_{vG} = 0{,}91$

$a_{vg} = 0{,}71$

was marked either on the sample or on the upper interferometer plate, and the order was referred as zero when the distance between two plates was zero so that the integral order always corresponds to the dark part of the fringes. *Figure* 4 illustrates schematically how the relative positions of the spectral lines were evaluated.

A dark part of the interference fringe by R (Cd red 6 438 Å), the order of which is K*, was used as a standard and the distance between R and g (Hg green 5 461 Å) as well as that between the first and the second order of g ($g + 1$, g) were measured. From the results a constant a_{Rg}

$$a_{Rg} = \frac{\text{Distance } (R, g)}{\text{Distance } (g+1, g)}$$

(which corresponds to the fraction of the value $K^* \frac{\lambda R}{\lambda g}$, was calculated for the value, say, $a_{Rg} = 0.55$. The same procedure was repeated for the other combinations of the spectral lines in order to obtain a_{Gg}, a_{vg}, a_{RG} and a_{vG}.

The second step is to determine the order of interference fringes. The length of the subsidiary piece was measured with a screw micrometer with known accuracy. As this value can be presumed as that of the sample length measured by the screw micrometer, the range within which the order of the fringes by R 6 438 Å, K*, exists may be estimated.

For each value of K* being included in this region values of a_{Rg} were calculated in order to choose the K* value which made the observed and calculated values of a_{Rg} to coincide.

The same process was repeated for a_{Gg}, a_{vg}, a_{RG}, and a_{vG} for the purpose of finding out one set of the orders of the fringes K*, L*, etc., each corresponding to the spectral lines R, G, etc.

When the sample length is known with the accuracy of 3 μ, the appropriate value of K*, L*, etc. may be fixed if a_{Rg}, etc., can be measured with the accuracy of 0.10. When the sample length is known with the accuracy of 5 μ the values of the orders may be fixed if a_{Rg} can be measured with the accuracy of 0.06, and those of a_{Gg}, a_{vg}, a_{RG}, a_{vG} with that of 0.10.

As those values may be estimated with accuracies higher than those in above examples K*, L*, etc., may generally be determined even when the calculations are performed under the assumption that the accuracies so low as 10 μ and 0.10 for the sample length and for a_{lig}, etc. respectively. The author's experience has shown that the accuracy of the measurement of a_{lig}, a_{Gig}, a_{RG} is 0.05.

The third and the last step is the evaluation of the exact sample length. As the order of the interference fringes in the field of the ocular micrometer can thus be determined the order of the interference fringes at the marked point, or the order at the marked point, for example by G 5 086 Å L, can be known with the precision of 0,05. If the half wave length of G (the lines of Cd green) be multiplied by the corresponding order L at the marked point, the sample length may be calculated with the precision of $\pm$ 0.015 μ, although the wave length shold be corrected because of the influence of temperature and atmospheric pressure. This point will be referred below.

c) Air correction.

The wave length of light is affected by the temperature and pressure of air, and consequently it is necessary to make some corrections for these influences. The air correction was calculated by assuming that the wave length of light and the refractive indices of air in the standard state (15 °C, 760 mmHg) have the values as given in Table 1 [18], and those of "refractive index - 1" are proportional to absolute temperature, and are inversely proportional to the atmospheric pressure.

TABLE 1

Source	Wave length	(Refractive index - 1) 10^7
	Å	
Cd (R)	6438.4690	2763.8
Cd (G)	5085.8212	2787.2
Hg (g)	5460.7430	2778.8
Hg (v)	4358.325	2810.5

If λ_0 represents the wave length in standard state and $\Delta_{T,p;15}$ the air correction at the temperature T under the atmospheric pressure p, the wave length at T, p will be given by

$$\lambda_{T,p} = \lambda_0 \, (1 + \Delta_{T,p;15}).$$

The calculated air correction seems to be correct with the precision of 2×10^{-6} at room temperature, and 2×10^{-7} in the temperature range between 0 °C and 300 °C.

The refractive index of air and its variation due to the change of temperature and pressure at room temperature have already been thoroughly studied, so that it may be calculated with the help of the following three experimental formulae.

$$(n-1)\,10^6 = \left\{ \left(288.02 + \frac{1.478}{\lambda^2_{air}} + \frac{0.0316}{\lambda^4_{air}}\right) \frac{h\,(1+\beta h)}{760\,(1+760\,\beta)} \frac{1}{1+0.003710} \right\}$$

$$\beta = 2.4 \times 10^{-6} \qquad (a)\ [20]$$

$$(n_{t/R} - 1) = \left(268.036 + \frac{1.476}{\lambda^2_{vac}} + \frac{0.01803}{\lambda^4_{vac}}\right) \frac{1+\alpha 20}{1+\alpha t} \frac{B}{760}\, 10^{-6}$$

$$\alpha = 0.00367 \qquad (b)\ [21, 22]$$

$$(n_{t,p} - 1) = (n_{20;760} - 1) \frac{p(1+\beta p)}{760(1+760\beta)} \frac{1+20\alpha}{1+T\alpha}$$

$$\alpha = 0.003674; \qquad \beta = 0.37 \times 10^{-6}$$

$$(c)\ [23, 24]$$

Although it is not clear how well these equations can be applied to higher temperatures the author tried to calculate the air corrections at higher temperatures by mere extrapolations. The results of the calculation is shown in table 2.

TABLE 2. — *The air correction for* He (Y) 5876 A
$\Delta_{Y,t,760;15}$ unit 10^{-6}

Temp.	By equation			By MERRITT [9] (1)	By the author's assumption
	(a)	(b)	(c)		
20 °C	0.0479	0.0474	0.0474	0.05	0.0473
100 °C	0.6382	0.6326	0.6329	0.635	0.6313
300 °C	1.388	1.380	1.380	1.38	1.378
1 000 °C	2.151	2.146	2.146	2.14	2.144

(1) $\Delta_{t,23}$ in the original table is converted to $\Delta_{t,15}$.

d) Measurement of Expansion.

Having measured the sample length very exactly at room temperature we now reach the stage of measuring the dilatation due to the temperature elevation as well as the change in length during prolonged observations.

For calculation of the thermal expansion coefficient it is necessary to know the order of the interference fringes at the marked point at any set of temperatures T_1 and T_2.

The usual method for determining the order at high temperature is to count or to record with an automatic recorder the number of fringes which pass through the marked point during the heating of the specimen.

It was confirmed that the coincidence method can also be used for this purpose. Let 2D represent the optical path difference at a dark part of the interference fringes by R (Cd red 6 438 Å) at the temperature T_2, K* its order, and L the order of the fringes by G (Cd green 5 086 Å) at the sample place we have the following relation which may be used for evaluating the exact value of L.

$$2D = \lambda_{R_2} K^* = \lambda_{G_2} L$$

$$\lambda_{R_2}(1+\Delta_{R_2})\, K^* = \lambda_{G_2}(1+\Delta_{G_2})\, L$$

$$L = K^* \lambda_{R_2}/\lambda_{G_2} + L(\Delta_{R_2} - \Delta_{G_2})$$

$$(\because K^* \lambda_{R_2} = L \lambda_{G_2})$$

If L is known previously with the accuracy of $\pm$ 2 to 4 the exact value of L may be determined by calculating L for every K* and comparing the fraction of it with that of measured value. Furthermore L can be determined by using g (Hg green 5 461 Å) together with R and G when L is previously known with the accuracy of $\pm$ 6 to $\pm$ 13. The accuracy being necessary for measuring the fraction of L is $\pm$ 0.10.

The same procedure may be used for the measurement of the dimensional change of a specimen while its temperature

is kept constant for a long time. It may also be used for checking the errors which may occur in counting the number of moving fringes during the heating-up process, or for the comparison of the length of specimen before and after the experiment.

4. — THE REPRODUCIBILITY OF THE THERMAL EXPANSION COEFFICIENT

As the sample length may thus be determined at different temperatures, say, T_1 and T_2, the mean value of the thermal expansion coefficient between these temperatures may be estimated as follows.

Let l be the sample length and N the order of the fringes at the marked point. The wave length and the values of N should be known at three different temperatures, namely, at 15°, T_1, and T_2 °C, each being distinguished below by adding the suffixes 0, 1, and 2 to the corresponding notations.

Then there exists the relations.

$$l_1 = N_1\lambda_1/2 = (1 + \Delta_1)N_1\lambda_0/2 \qquad (\text{at } T_1),$$
$$l_2 = (1 + \Delta_2)N_2\lambda_0/2 \qquad (\text{at } T_2),$$

and the mean thermal expansion coefficient is given by:

$$\alpha_{T_1, T_2} = l_2 - l_1/l_0(T_2 - T_1).$$

As the wave length at 15 °C was used as a standard it is convenient to use the sample length at 15 °C to calculate the thermal expansion coefficient.

When $l_0 = 10$ mm,
$\lambda_0 = 0.5\ \mu$,
$T_1 =$ room temperature,
$T_2 = 300$ °C,

and the expansion coefficient of the sample,

$$\alpha_{0,300} = 100 \times 10^{-7}/°C$$

then N_1 will be equal to 4×10^4.

1° As the precision of measuring $T_1 = 0.1$ °C, and that of measuring $T_2 = 0.6$ °C, $T_2 - T_1$ will approximately be equal to 300 °C with the precision of 0.7 °C, whose relative error amounts to 0.23 %.

2° As the precision of measuring N_1 and $N_2 = 0.05$, and that of calculating $\Delta_1 = 1 \times 10^{-7}$, and that of calculating $\Delta_2 = 4 \times 10^{-7}$, taking into account the error caused by that of measuring temperature and atmospheric pressure, the precision of l_1 and l_2 becomes 0.015 μ and 0.020 μ, respectively. Therefore, $l_2 - l_1$ will approximately be equal to 30μ, with the accuracy and the relative error of 0.04 μ and 0.13 %.

3° The precision of l_0 is 0.02 μ, and its relative error is 0.0002 %.

Consequently, the repeatability of the measurement of the thermal expansion coefficient is expected to be about ± 0.4 %, namely $\pm 0.4 \times 10^{-7}$/°C.

5. — THE SURFACE FILM EFFECT AND ERRORS CAUSED BY THE DIFFERENCE IN EXPANSION BETWEEN THE SAMPLE AND THE INTERFEROMETER PLATES

The surface film adsorbed on the sample is known to be one of the principal sources of errors [2, 10]. As already stated the method of preparing the sample was improved in order to minimize this effect with the result that the error coming from this source was made negligibly small. An example of checking the surface film effect is shown below.

The sample was prepared from a tube of vitreous silica (1) by the method described in III. The fringe order, M, by g (5 461 Å) at the mark point was measured with the following result.

Sample No. 1	Sample No. 2
Transparent fused silica (as received).	Opaque fused silica (preheated at 1 350 °C for 2 hrs).
Immediately after preparation: 12.2 °C, 765.0 mm Hg.	Immediately after preparation: 16.0 °C, 756,1 mm Hg.
M = 47851.73 (13.06524 mm).	(M = 36147.59 (9.86966 mm).
	After preheating up to 280 °C 14.4 °C, 762.4 mm Hg M = 36147.70 (9.86965 mm).
After heating to 327.7 °C at the rate 1.5 °C/mn: 327.7 °C, 763.3 mm Hg.	After heating to 349.0 °C at the rate 1.5 °C/mn: 349.0 °C, 761.7 mm Hg.
M = 47853.65 (13.06771 mm).	M = 36149.71 (9.87168 mm).
Then after holding at 327.7 °C for 3.5 hrs.: 327.4 °C, 763.3 mm Hg.	Then after holding at 349.0 °C for 3.5 hrs.: 348.5 °C, 762.3 mm Hg.
M = 47853.70 13.06772 mm).	M = 36149.73 (9.87169 mm).
After cooling: 14.0 °C, 762.9 mm Hg.	After cooling: 13.3 °C, 769.6 mm Hg.
M = 47851.77 (13.06528 mm).	M = 36148.00 (9.86970 mm).

The above figures prove that the dimensional change is within the experimental error. If we assume that the silica glass does not change its length when heated to about 350 °C, it can be concluded that the surface film effect can be neglected as long as the sample is prepared by the improved method.

It will be expected that the difference in expansion between the sample and the interferometer plates affects the set up condition, because the contact points would move around and the contact pressure would change the direction during the experiment, or during heating up, so that some errors might be introduced [2, 10].

As in the author's measurement the temperature was kept constant for sufficient time to equalize the distribution around the sample and the plates, the condition of contact will be about the same as at the beginning and consequently the possibility of introducing the errors would be reduced to the minimum.

Actually, when the thermal expansion coefficient was measured, keeping the temperature constant, a better reproducibility was obtained. In fact it has been proved that when the expansion was measured in the continuous heating up process, it was impossible to avoid the dispersion of the data due to the difference in expansion described above and the uneven temperature distribution around the sample.

6. — CHANGE OF SAMPLE LENGTH DURING MEASUREMENT

Recently it has been revealed that glass changes its volume when it is kept at a constant temperature. This can be observed by measuring its density change when glass is kept

(1) Manufactured by Nihon Denko Co.

at a temperature in the transformation range [25, 28], and by observing the ice point drift of the glass thermometer at comparatively low temperature for a long time [29, 31].

It is usually considered, however, that the dimensional change occurs very slowly at low temperatures, namely, at temperatures more than 150 °C lower than the strain point, and the time effect would hardly be observed by heating only a few hours as is the case of expansion measurement. But this assumption is not valid if the sample be held at a constant temperature for some longer time, or when the heating up rate is small in order to get better accuracy in measuring the expansion.

A distinct volume change could be observed even at comparatively low temperature, for example, lower than 250 °C. This effect was observed in the following experiments.

The sample was kept at constant temperature within $\pm$ 0.6 °C and the shift of the interference fringes was observed. To eliminate this time effect, therefore, the author took the following steps; the measured change of length was converted to the value at the mean temperature assuming a proper value of expansion coefficient; and when the change of length during the measurement was small the total contraction was calculated by comparing the sample length before and after the measurement.

1. Fused silica glass.

The result of measurement is shown in VI and no change in length was observed when heated up to 350 °C.

2. Pyrex chemical resistant type borosilicate glass (*fig.* 5 and 6).

The chilled sample contracted considerably at 301 °C. The roughly annealed sample (heated at 650 °C for a short time, then cooled in the furnace rather quickly) showed less tendency to contract. The expansion curves differ according to the thermal history, and even with the heat treatment at 337 °C a decrease in expansion was observed [32].

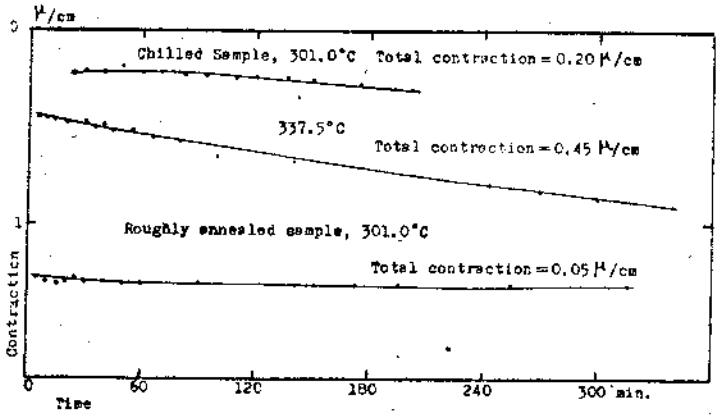

FIG. 5.—The contraction of a Pyrex chemical resistant type borosilicate glass.

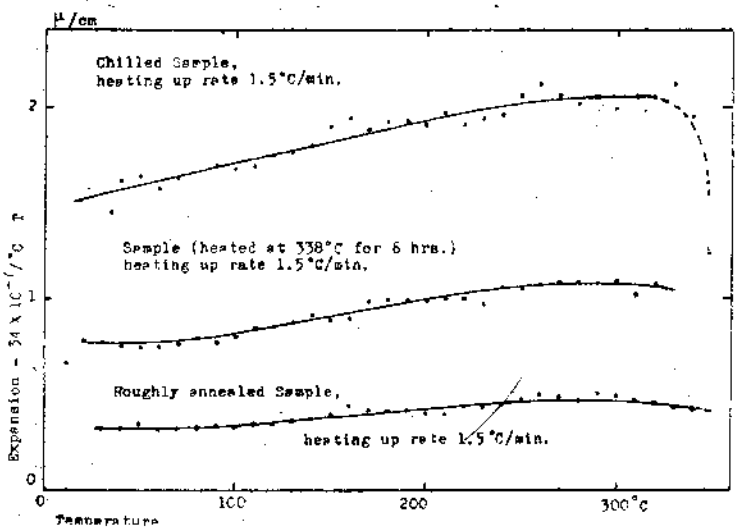

FIG. 6.—The expansion curves of a Pyrex chemical resistant type borosilicate glass.

3. Potassium-soda-lead glass (stem glass for vacuum valves) (*fig.* 7 and 8).

The chilled sample contracted appreciably at 226 °C, while the roughly annealed one (heated at 550 °C for some short time, then cooled in the furnace) and another one, heated at 420 $\pm$ 5 °C for an hour, contracted at 300 °C. When the chilled sample was heated at the rate of 1.5 °C/mn the expan-

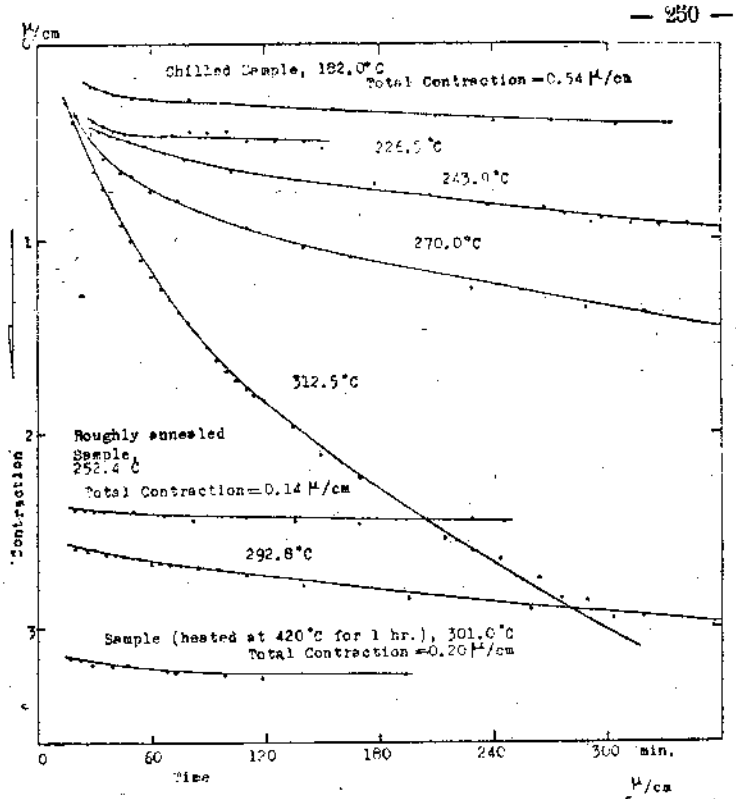

FIG. 7.—The contraction of a potassium-soda-lead glass.

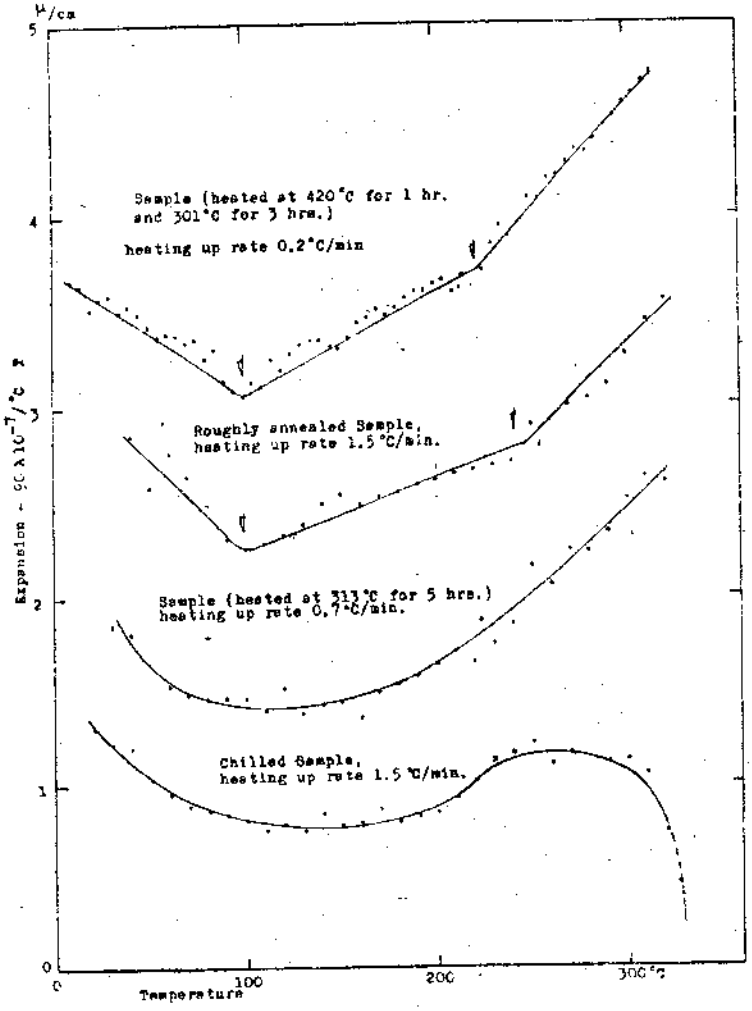

FIG. 8.—The expansion curves of a potassium-soda-lead glass.

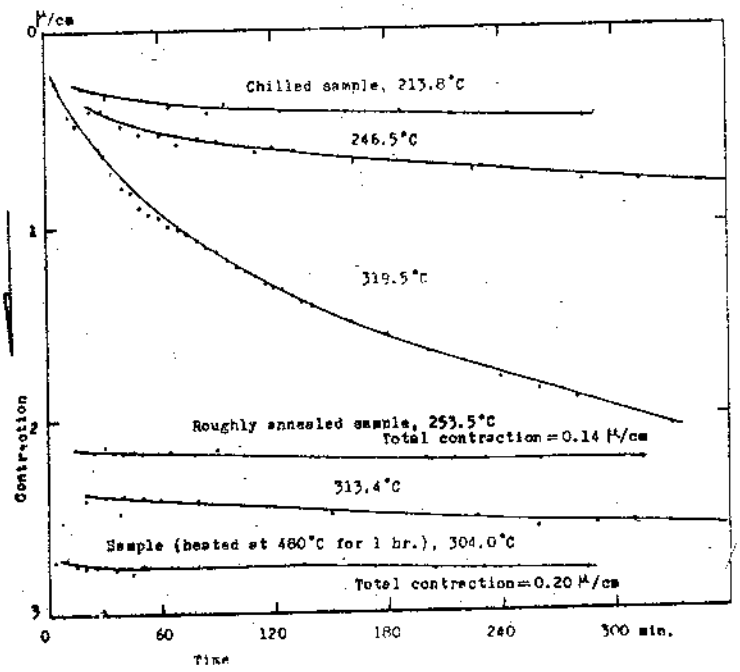

FIG. 9.—The contraction of a soda-lime-magnesia glass.

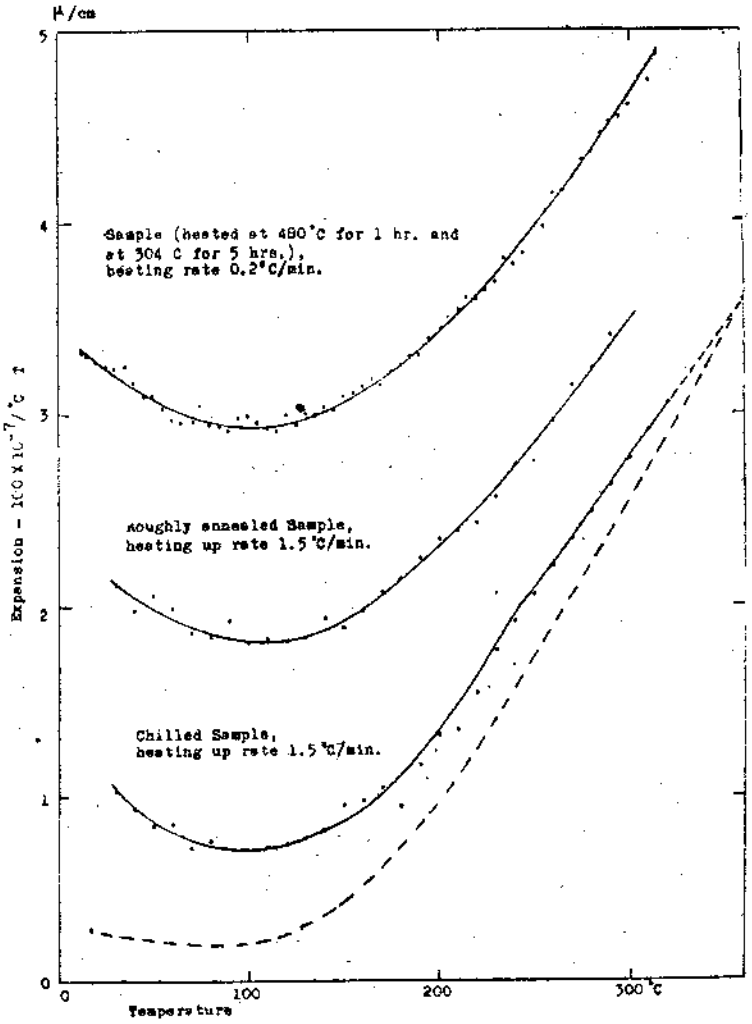

FIG. 10.—The expansion curves of a soda-lime-magnesia glass.

TABLE 3. — *The result of measuring the thermal expansion.*
Coefficient of the soda-lime-magnesia glass.

0 Right after preparation	1	2	3	4	5
12.2 °C 765.4 mmHg 47210.99G[1] 12.00528 mm	15.6 °C 766.3 mm Hg 47211.18G 12.00536 mm	15.8 °C 765.7 mm Hg 47211.45G 12.00543 mm	14.3 °C 756.3 mm Hg 47210.66G 12.00526 mm	12.2 °C 765.6 mm Hg 47210.31G 12.00510 mm	11.6 °C 765.4 mm Hg 47209.86G 12.00498 mm
	117.0 °C 765.3 mm Hg 47254.01 G 12.01713 mm	88.7 °C 763.2 mm Hg 47241.53 G 12.01377 mm	99.0 °C 756.1 mm Hg 47246.05 G 12.01502 mm	98.7 °C 764.7 mm Hg 47245.94 G 12.01496 mm	98.9 °C 764.1 mm Hg 47246.21 G 12.01503 mm
	185.5 °C 765.7 mm Hg 47286.17 G 12.02568 mm	198.9 °C 760.3 mm Hg 47293.10 G 12.02752 mm	199.7 °C 758.2 mm Hg 47293.61 G 12.02766 mm	196.7 °C 763.8 mm Hg 47292.21 G 12.02727 mm	195.2 °C 764.4 mm Hg 47291.27 G 12.02702 mm
304.6 °C 762.9 mm Hg 47348.36 G 12.04194 mm	301.4 °C 765.7 mm Hg 47346.32 G 12.04141 mm	286.7 °C 760.3 mm Hg 47338.30 G 12.03933 mm	293.5 °C 759.0 mm Hg 47342.12 G 12.04033 mm	295.0 °C 763.2 mm Hg 47342.82 G 12.04050 mm	293.2 °C 764.8 mm Hg 47342.00 G 12.04028 mm
After 3.5 hr 303.5 °C 762.1 mm Hg 47347.33 G 12.04168 mm	$\alpha_{15\ 301}$[3] = 105.05×10^{-7}/°C	$\alpha_{15\ 287}$ = 104.24×10^{-7}/°C	$\alpha_{14\ 291}$ = 104.63×10^{-7}/°C	$\alpha_{13\ 295}$ = 104.27×10^{-7}/°C	$\alpha_{15\ 293}$ = 104.43×10^{-7}/°C
	$\alpha_{0\ 300}$[2 3] = 104.0×10^{-7}/°C	$\alpha_{0\ 290}$ = 103.7×10^{-7}/°C	$\alpha_{0\ 300}$ = 103.9×10^{-7}/°C	$\alpha_{0\ 300}$ = 103.7×10^{-7}/°C	$\alpha_{0\ 300}$ = 103.9×10^{-7}/°C

[1]: order of the interference fringes by G.
[2]: l^0 = 12.00530 mm which is the sample length at 15.0 °C.
[3]: This value is calculated assuming that

$$\alpha_{0\ 15} = 85 \times 10^{-7}/°C$$
$$\alpha_{280\ 305} = 115 \times 10^{-7}/°C$$

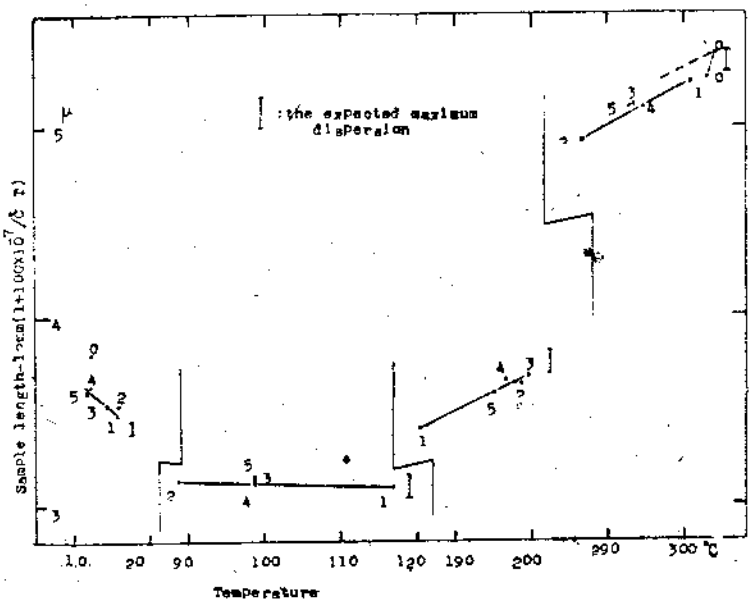

FIG. 11.—The result of a repeated measurement of the thermal expansion coefficient of a soda-lime-magnesia glass.

sion curve indicated that the decrease of its expansion coefficient has already had an effect.

Those samples which were heated at a temperature in annealing range change their expansion coefficient abruptly at around 100 °C and 230 °C. These temperatures seem to correspond to the transition points described by TURNER and his coworkers [33, 36, 37].

4. **Soda-lime-magnesia glass** (bulb glass for lamps and vacum valves) (*fig.* 9 and 10).

The chilled sample contracted considerably at 214 °C. The roughly annealed sample (heated at 500 °C for a short time then cooled in the furnace) and another one, heated at 480 ± 5 °C for an hour also contracted at 300 °C.

When the chilled sample was heated at the rate of 1.5 °C/mn, the expansion curve indicated that a decrease of the expansion coefficient at 240 °C had occured.

Those samples which were heated up to the annealing range showed a considerable change of the expansion coefficient between the room temperature and 300 °C, particularly around 100 °C and 240 °C [33, 37].

Perhaps, these are the transition points, but the abruptness of the change in the expansion coefficient differs from that of the potassium-soda-lead glass. This is seen in the figures.

7. — REPEATED MEASUREMENT OF THE THERMAL EXPANSION COEFFICIENT

The attention should be paid to those causes of errors which were mentioned above if we wish to measure the thermal expansion coefficient as precisely as possible. An example of the reproducibility when the coefficient was measured, taking these effects into account, is as follows.

A sample was made of soda-lime-magnesia glass which was preheated at 480 ± 5 °C for an hour. The contraction was observed at 340 °C for about 5 hours and the expansion coefficient was measured repeatedly. The contraction during the measurement was expected to be very small. The result of the measurement is shown in Table 3 and in *figure* 11.

The dispersion of date was smaller than expected from the accuracy of the temperature and the fringe order measurement, and the uneven temperature distribution around the sample. The value of $\alpha_{0\text{-}300}$ was measured with the repeatability whose variation is less than $\pm\ 0.2 \times 10^{-7}$/°C. No contraction was observed during the measurement.

8. — CONCLUSION

1) It is shown that the thermal expansion coefficient can be measured by the interferometric dilatometer with a repeatability of $\pm\ 0.2 \times 10^{-7}$/°C. And it is expected that the accuracy of $\pm\ 0.4 \times 10^{-7}$/°C can be obtained by using a calibrated thermo-junction.

2) It has been shown that the contraction of the sample during the measurement should not be neglected. The rate of contraction depends upon its thermal history.

3) Potassium-soda-lead glass preheated up to annealing range showed distinct breaks on its expansion curve. As for soda-lime-magnesia glass preheated up to annealing range, the breaks in the expansion curve were also observed, but the changes of the expansion coefficient were not so distinct as in the case of potassium-soda-lead glass.

4) It is demonstrated that the coincidence method which has been used at room temperature can also be used at higher temperatures at least up to about 300 °C.

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