

## Some problems of precise measurements of heat transfer coefficients in glass melts

### Part 1. Measurements of effective conductivity

Sergei V. Tarakanov, Victor K. Leko and Oleg V. Mazurin  
Joint-Stock Company Thermex, St. Petersburg (Russia)

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In the paper the method of measurements of the so-called effective conductivity of glass melts basing on characteristic of temperature gradient in a molten glass inside a refractory tank is analyzed. For this purpose a computer model based on the solution of the radiative transfer equation in a semitransparent medium was used. Among the papers on effective conductivity measurements published so far, only two contained enough numerical information on the details of measurement which permitted to perform the corresponding calculations. The authors of the present paper show what particular defects of experimental procedure could be the reason of surprising results reported in these publications, namely that the measured effective conductivity coefficient of a number of glasses was found to be lower than the radiative conductivity alone.

#### Einige Probleme bei der Messung von Wärmeübertragungskoeffizienten in Glasschmelzen Teil 1. Messungen der effektiven Leitfähigkeit

In der vorliegenden Arbeit wird ein Verfahren zur Messung der sogenannten effektiven Leitfähigkeit von Glasschmelzen analysiert, das auf dem Temperaturgradienten als Kennwert der Glasschmelze in der Schmelzwanne beruht. Das zu diesem Zweck angewendete Computermodell basiert auf der Lösung der Gleichung für die Strahlungswärmeübertragung in einem halbdurchlässigen Material. Von den bisher veröffentlichten Arbeiten über Messungen der effektiven Leitfähigkeit enthalten nur zwei ausreichende numerische Informationen über Einzelheiten der Messung, um damit entsprechende Berechnungen durchzuführen. Die Verfasser der vorliegenden Arbeit zeigen die wesentlichen Mängel der in den genannten Veröffentlichungen beschriebenen experimentellen Vorgehensweise auf, die der Grund für die darin beschriebenen überraschenden Ergebnisse sein könnten; so wurde z. B. festgestellt, daß die gemessene effektive Leitfähigkeit einer Anzahl von Gläsern niedriger war als die Strahlungsleitfähigkeit allein.

### 1. Introduction

An impressive progress in computer technique permits to develop software for precise computer simulation of various technological processes. It is possible now to increase substantially the effectiveness of such processes of glass technology as glass melting, glass forming, and glass annealing or tempering by using computer simulation approaches. These possibilities, however, may only be realized if the specialists who are developing and using the corresponding computer programs are supplied with precise enough data on a number of glass melt properties. Among these properties are viscosity, density, heat transfer characteristics, specific heat, surface tension, and in some cases electrical conductivity. At present the problems of precise measurements of heat transfer characteristics of glass melts seem to be considerably less developed than those of any other of the enumerated properties of glass melts. Hence, this problem should attract a special attention of scientists.

Glasses belong to the so-called semitransparent substances. This means that one has to take into account

two processes of heat transfer in glasses: thermal conductivity, which is similar to the corresponding process in any opaque substance and radiative conductivity, which is determined by heat transfer resulting from radiation. It is known that the coefficient of thermal conductivity increases by factors 1.5 or 2 from an ambient temperature to 300 to 400 °C, and after that its changes with temperature are comparatively small. On the other hand, radiative conductivity at temperatures below 400 to 500 °C is negligible, but it increases intensively with temperature and at high enough temperatures its values surpass those of thermal conductivity by many times.

For calculation of heat transfer taking part during melting and annealing one has to consider both kinds of conductivity. For calculations of heat transfer in glass melting furnaces for comparatively transparent glasses, one can often use only radiative heat transfer data as a reasonable enough approximation. For colored glasses radiative conductivity coefficients are considerably lower than for practically colorless glasses, and in these cases it may be important to know values of thermal conductivity coefficients even at high temperatures.

In comparison with publications on most of the practically important properties of glass melts, the

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number of publications on heat transfer characteristics of glass melts is rather small. The authors of the majority of papers do not compare their results with results published for glasses of the same types in other papers although sometimes the difference between such results exceeds the reported errors by many times. Probably it was not so significant some time ago when semiquantitative information on values of heat transfer coefficients could satisfy most of the needs of glass technologists. Now the knowledge of precise heat transfer data is important. Thus, the critical analyses of the existing literature on measurements of heat transfer coefficients seem to be essential.

Two main methods of studies of heat transfer coefficients in semitransparent substances at high temperatures are known. One is similar to that which is used for measurements of thermal conductivity coefficients in opaque substances. The so-called effective conductivity coefficient is obtained in this case, which is the sum of the thermal conductivity coefficient and a certain additional value arising from the influence of the radiative heat transfer. Problems related to the proper use of this method will be discussed in the first part of the present paper. The second method is the measurement of absorption spectra of glass melts and calculation of radiative conductivity from these spectra. Analyses of studies using this method will be presented in the second part of this paper.

## 2. Two main methods of heat transfer studies of glass melts

In the following the two methods in question will be considered. The first method is based on the determination of temperature gradients in the melt and of the value of heat flux. If certain conditions are met (these conditions will be discussed later in detail), this method permits to measure the sum of thermal conductivity and radiative conductivity coefficients. Therefore, it will be called the method of determination of total conductivity coefficient,  $\lambda_s$ .

The other method is based on the measurements of the absorption spectrum of the melt within a broad enough range of wavelengths (the higher the temperature of measurements, the higher should be the corresponding range). Evaluation of this spectrum permits to calculate the radiative conductivity coefficient,  $\lambda_r$ .

The difference in these two coefficients is equal to the thermal conductivity coefficient,  $\lambda_c$ :

$$\lambda_c = \lambda_s - \lambda_r.$$

If the values of  $\lambda_c$  and  $\lambda_r$  are of the same order of magnitude, such a way of determining  $\lambda_c$  is quite practicable. However, at temperatures of glass melting it is possible only for colored glasses with high absorption coefficients in the red and near-infrared regions of spectra. Usually,

in this temperature range  $\lambda_r \gg \lambda_c$ . In this case the value of  $\lambda_c$  can be considerably smaller than the sum of errors of  $\lambda_s$  and  $\lambda_r$  measurements.

It seems that in most cases scientists working on the determination of thermal conductivity measured not the total conductivity but only part of it, that is the sum of  $\lambda_c$  and a certain fraction of  $\lambda_r$ . Such a sum can be called the effective conductivity coefficient,  $\lambda_{\text{eff}}$ :

$$\lambda_{\text{eff}} = \lambda_c + x \lambda_r$$

where  $x < 1$ . If  $x = 1$ , then instead of  $\lambda_{\text{eff}}$  the value of  $\lambda_s$  is obtained by the corresponding measurements. Very often the real value of  $x$  remains unknown.

It is very difficult if possible at all to try to determine the probable level of errors and sources of such errors for most of the published studies on thermal, radiative, and total conductivity of glass melts: The papers usually contain too little information on this subject. However, there are found two exceptions to the rule. Fortunately, the papers by Henderson et al. [1] and Endrýs et al. [2] contain both a detailed enough description of effective conductivity measurements and data on radiative conductivity obtained by evaluation of absorption spectra. Accordingly, the present authors were able to use these two papers as a base for discussions on the problems of precise measurements of heat transfer characteristics in glass melts.

In the analysis a well-known theory of heat transfer in semitransparent substances (see for example [3 and 4]) was used.

## 3. Analyses of experimental data presented in [1 and 2]

The density of heat flux correlated with the thermal conductivity process is expressed by the Fourier law:

$$\vec{q}_c = -\lambda_c \nabla T$$

where  $\vec{q}_c$  is the vector of conductive heat flux density,  $T$  the absolute temperature,  $\nabla$  the operator of the gradient.

Unlike the conductive heat flux the density of which is determined by the value of thermal conductivity coefficient and temperature gradient in the given point, i. e. is a local quantity, the radiative heat flux is an integral quantity, depending on the state of media in a certain volume. The radius of this volume depends on the frequency of radiation and is equal, by the order of magnitude, to the length of the free path of the radiation which is determined by the well-known Rosseland formula. In a particular case of the dense optical medium, when a free path radiation length is considerably smaller than the specific linear scale of the studied object ( $l \ll L$ ), the expression for a heat flux density takes a

form of Fourier law. In this case it appears possible to introduce notions of radiative conductivity and total conductivity coefficient:

$$\vec{q}_s = -(\lambda_c + \lambda_r) \nabla T \quad (1)$$

where  $\vec{q}_s$  is the vector of total heat flux density.

Description of the radiative heat flux density using the approximation of radiative conductivity (equation (1)) is quite tempting. However, one should remember that it is possible only for the case of an optically thick medium. If this condition is not met, then the notions of radiative and total conductivity do not have any physical meaning, although formally the values of the corresponding coefficients can be derived from the experimental results.

As far as it is known, in practically all the papers published so far where the method of determination of effective conductivity coefficients for glass melts by measurements of the temperature gradient and heat flux density was used [1, 2, and 5 to 7], the evaluation of experimental data was performed on the assumption that the approximation of radiative conductivity is correct. Probably, the only exception are the publications by Tyutyunnik (e. g. [8]).

Among the glasses investigated in the mentioned studies [1 and 2], the authors selected the following glasses for their analyses: green bottle glass, white bottle glass, float glass, and TV-screen glass from [2], and also light green glass and two colorless glasses from [1].

Using temperature dependencies of radiative conductivity presented in [1 and 2] (which were determined by evaluation of the absorption spectra of the corresponding glasses) the authors calculated temperature dependencies of free paths of radiations for all the enumerated glasses (figure 1). The thicknesses of glass layers in experimental units were as follows: in a study described in [1] about 150 mm, and in that described in [2] about 200 mm. This means that the approximation of radiative conductivity can be considered as definitely correct at least for two of the studied glasses, namely for the green bottle glass and the light green glass.

For white bottle glass, float glass, and colorless glasses the free path of radiation is of the same order of magnitude as the thickness of the studied glass. In these cases one can expect that the use of the radiative conductivity approximation should lead to the qualitatively correct result for the inner areas of the glass melt. However, in those areas of the melt which are spaced from the surface by a distance less than the free path of radiation the errors of calculations could be quite significant.

For TV-screen glass the free path of radiation surpasses half of the thickness of the melt, and therefore the use of the approximation of radiative conductivity is impossible here.

Thus, in principle the radiative conductivity approximation could be correctly used for the analysis of experiments with green bottle glass [2] and light green glass

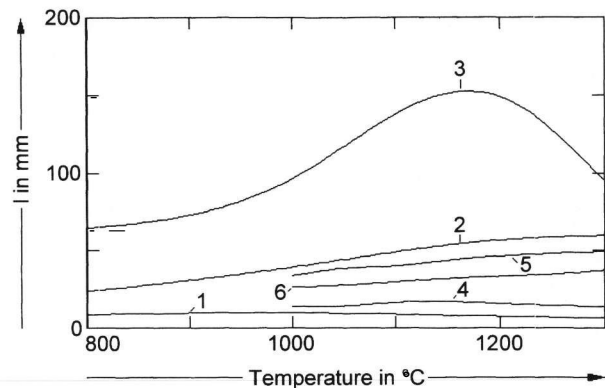


Figure 1. Free path of radiation as a function of temperature determined by Rosseland formula for various glasses; curve 1: green bottle glass [2]; curve 2: float and white bottle glasses [2]; curve 3: TV-screen glass [2]; curve 4: light green glass [1]; curve 5: colorless glass (0.035%  $\text{Fe}_2\text{O}_3$ ) [1]; curve 6: colorless glass (0.051%  $\text{Fe}_2\text{O}_3$ ) [1].

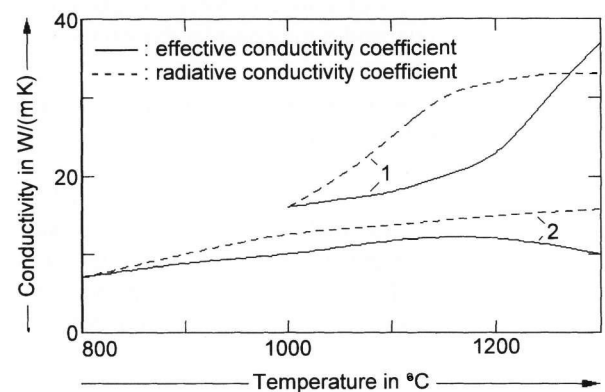


Figure 2. Effective and radiative conductivity coefficients for two glasses; curve 1: light green glass [1]; curve 2: green bottle glass [2].

[1]. There was every reason to expect that the coefficients of effective conductivity obtained for these glasses are equal to the coefficients of total conductivity, i. e. to the sum of thermal and radiative conductivity coefficients. However, as it follows from the papers in question, it was not so.

Figure 2 demonstrates the temperature dependencies of  $\lambda_{\text{eff}}$  and  $\lambda_r$  for the two glasses, taken from [1 and 2]. It is seen from the figure that for green bottle glass  $\lambda_{\text{eff}}$  is smaller than  $\lambda_r$  in the whole investigated temperature range. At high temperatures even the character of the two dependencies is different:  $\lambda_r$  steadily increases with increasing temperature, but  $\lambda_{\text{eff}}$  has a maximum near 1200°C.

Another kind of dependencies is seen for light green glass. Here the coefficients of effective and radiative conductivity coincide with each other on the borders of the investigated temperature interval, but within this interval  $\lambda_{\text{eff}}$  is considerably smaller than  $\lambda_r$ . The shapes of the curves are also quite different.

The method of effective conductivity evaluation by measurements of temperature gradients in glass melts is still used broadly enough. Therefore, the authors tried to find reasons for these discrepancies using a mathematical model of radiation transfer.

#### 4. Computer simulation of temperature profiles in glass melts for experimental conditions of [1 and 2]

For the study of problems connected with experimental determination of heat transfer characteristics of glass, the numerical modelling of temperature distributions in glasses for conditions similar to those described in [1 and 2] was performed. As a subject for such modelling an infinite glass layer confined between two radiated planes with temperatures  $T_{\text{up}}$  and  $T_{\text{down}}$  was selected. The bottom plane (the bottom of the bath) is in contact with the glass, the upper plane (the crown of the furnace) is separated from the surface of the glass by a gas layer. The conditions of heat exchange between the glass, gas, and the crown can depend quite considerably on the way of the heating of the glass.

In the furnace described in [2] the glass was heated by a system of heating elements. In this case the temperature  $T_{\text{up}}$  is a certain average temperature of all radiative objects in the area above the glass surface including heating elements, the real crown of the furnace, side walls etc. Heat exchange between the glass and gas takes place in the form of free convection and presumably does not play an essential role in the heat transfer processes.

In the furnace used in [1] the glass was heated by a gas burner. In this case forced convection takes place. At the same time the exchange of radiation between the glass and the crown of the furnace should be also taken into account.

For calculation of temperature distribution in the glass a one-dimensional stationary form of heat transfer equation was used:

$$\lambda_c d_{xx}^2 T - d_x q_r = 0 \quad (2)$$

together with the following boundary conditions:

$$- \lambda_c d_x T + q_r^{\text{in}} = \alpha(T - T_{\text{gas}}) + q_r^{\text{ex}} \quad (3)$$

for  $x = 0$  and

$$T = T_{\text{down}} \quad (4)$$

for  $x = L$

where  $x$  is the coordinate counted from the upper surface of the glass ( $0 < x < L$ ),  $L$  is the height of the glass layer,  $q_r$  the density of the radiative flux of energy in the direction of the  $x$  axis above the surface (ex) and below the surface (in) of the glass,  $\alpha$  the coefficient of heat

transfer, which describes the exchange of heat between the glass and gas,  $T_{\text{gas}}$  the temperature of gas at the glass surface.

Boundary condition (3) expresses the continuity of the total heat flux on the upper surface of the glass, including conductive and radiative (internal) heat fluxes from the glass, and convective and radiative (external) heat fluxes to the glass. Condition (3) is similar to the conditions used in [9 and 10], although it differs somewhat from these conditions by its appearance. Condition (4) represents the equality of temperatures of the bottom plane of the glass and the bottom of the furnace in a point of their contact.

Values of radiative heat flux density were determined by the following equation:

$$q_r = 2\pi \int_0^\infty d\nu \int_{-1}^1 I_\nu \mu d\mu \quad (5)$$

where  $I$  is the spectral intensity of radiation;  $\nu$  the frequency of radiation;  $\mu$  the independent variable characterizing the direction of radiative flux propagation which is equal to the cosine of an angle between the direction of ray and the  $x$  axis.

For calculation of spectral intensity of radiation the equation of radiation transfer was used:

$$\mu \partial_x I_\nu = \kappa_\nu (n^2 I_{\nu e} - I_\nu) \quad (6)$$

for  $-1 < \mu < 1$

in combination with the following boundary conditions on the low and upper borders:

$\mu > 0, x = 0:$

$$I_\nu(\mu, 0) = r_{\text{down}} I_\nu(-\mu, 0) + (1 - r_{\text{down}}) n^2 I_{\nu e}(T_{\text{down}}), \quad (7)$$

$\mu < 0, x = L:$

$$I_\nu(\mu, 0) = r_{\text{up}} I_\nu(-\mu, L) + (1 - r_{\text{up}}) n^2 I_{\nu e}(T_{\text{up}}). \quad (8)$$

The dependence of coefficients of reflection  $r_{\text{down}}$  and  $r_{\text{up}}$  from the direction of propagation of radiation was determined by Fresnel's equations.

Unfortunately, in both publications [1 and 2] absorption spectra are not presented. The authors reported only values of radiative conductivity coefficients calculated by evaluations of these spectra. Thus, an inverse problem must be solved, i. e. the following integral equation obtained as a result of elimination of the radiative path length by the use of the Rosseland formula. It led to the following equation:

$$\lambda_r = \frac{16}{3} \sigma n^2 T^3 \frac{\int_0^\infty \kappa_\nu^{-1} \partial_T I_{\nu e} d\nu}{\int_0^\infty \partial_T I_{\nu e} d\nu} \quad (9)$$

where  $\lambda_r = \lambda_r(T)$  are the known temperature dependences.



The search for the solution of equation (9) was performed in the class of step functions (figure 3). It is to be noted that in spite of their simplicity, functions of these class describe the main features of spectral curves for many glasses reasonably well. That is why they are broadly used for approximation of absorption spectra of various glass compositions, see e.g. in [1 and 10]. Besides, due to the fact that the transfer of radiative energy is especially intensive in the range of spectra where the glass is the most transparent, in the solution of equation (9) the absorption coefficient was considered to be dependent on temperature only within this particular range of spectra (from 0.5 to 2.75  $\mu\text{m}$ ).

In figure 4 the profile of temperature distribution in the green bottle glass reported in [2] for experiment no. 1 is given. In the same figure the temperature distribution calculated on the base of the authors' model (curve 2) is also shown. In the course of calculations, the temperatures of radiated surfaces,  $T_{\text{up}}$  and  $T_{\text{down}}$ , were selected to ensure the correspondence of the calculated temperatures at upper and bottom surfaces of the glass melt to the experimental ones. The heat transfer between the upper surface of the glass and the gas was considered as negligible.

As follows from the figure, the main distinction between calculated and experimental curves is the difference between curvatures of these curves: The calculated curve is convex and the experimental one is concave. To find a possible reason for such a contradiction between theory and experiment, the sign of the second derivative of glass temperature is considered with respect to a spatial coordinate. Due to the validity approximation of radiative heat transfer for the green bottle glass the following equation can be used:

$$q_s = -\lambda_s d_x T$$

where  $q_s$  is the total heat flux density in the direction of  $x$  axis.

Differentiation of this expression with respect to  $x$  and assumption of independence of thermal conductivity from the temperature lead to the following equation:

$$d_{xx}^2 T = -\lambda_s^{-1} d_x q_s = q_s^2 \lambda_s^{-3} d_T \lambda_r. \quad (10)$$

In the case of one-dimensional and stationary temperature pattern the heat flux density does not depend on the coordinate, i.e.  $d_x q_s = 0$ . As it follows from figure 2, in the whole range from 800 to 1300  $^{\circ}\text{C}$  the temperature coefficient of radiative conductivity is positive. One can assume that it remains positive up to 1450  $^{\circ}\text{C}$  as well. Accordingly, as it follows from equation (10), the second derivative of temperature with respect to  $x$  should be negative, i.e. the calculated curve should be convex. From equation (10) follows that the mentioned second derivative can become positive in the discussed case only if the value  $d_x q_s$  becomes negative. This means that the

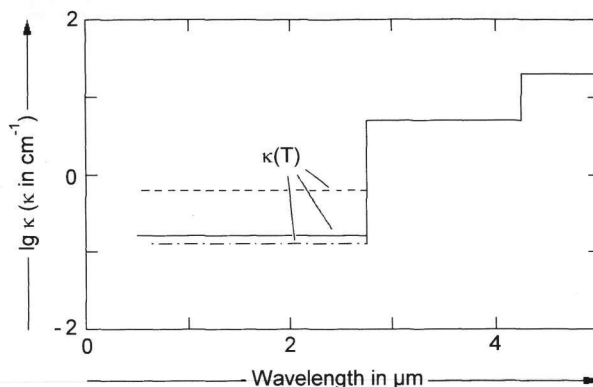


Figure 3. Approximation of absorption spectrum by means of a stepped function.

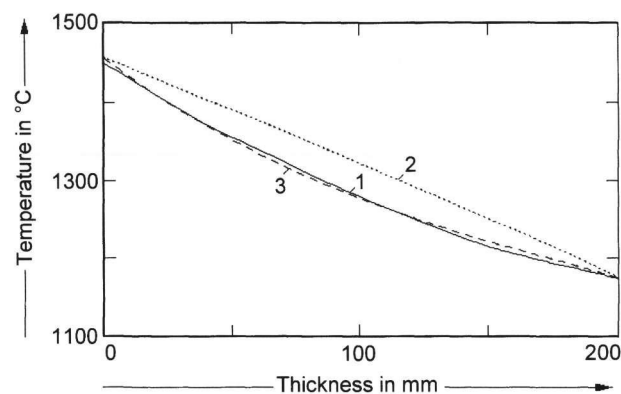


Figure 4. Distribution of glass temperature as a function of glass thickness for green bottle glass [2; figure 2, table 2, experiment no. 1]; curve 1: experiment; curve 2: theory, without heat losses ( $Q_{\text{max}} = 0$ ); curve 3: theory,  $Q_{\text{max}} = -200 \text{ kW m}^{-3}$ .

concavity of the experimental temperature profile can be explained by a decrease of the heat flux density through the glass in direction from the upper surface of the glass melt to the bottom, which is possible only in the case when the temperature profile in the glass is not one-dimensional.

Due to the expected violation of one-dimensionality of the heat field it could be considered reasonable to carry out the further analysis on the base of two-dimensional or three-dimensional approximations. However, due to the absence of information about temperature conditions on the side walls of the tank the authors had to keep within the one-dimensional case.

Within the frames of one-dimensional formulation of the problem one can model the change in heat flux along the depth of the glass layer if the possibility of heat losses or heat generation in equation (2) is introduced. These values can be found using the condition of the coincidence of experimental and calculated temperature profiles.

Good correspondence of the calculated and experimental curves, presented in figure 4 (curves 1 and 3), was reached at the following linear distribution of heat losses:

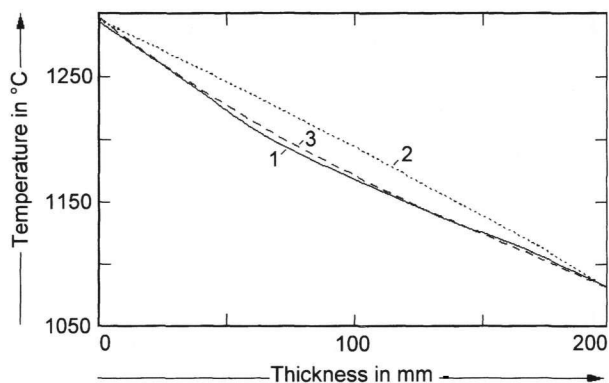


Figure 5. Distribution of glass temperature as a function of glass thickness for green bottle glass [2; figure 2, table 2, experiment no. 3]; curve 1: experiment; curve 2: theory, without heat losses ( $Q_{\max} = 0$ ); curve 3: theory,  $Q_{\max} = -100 \text{ kW m}^{-3}$ .

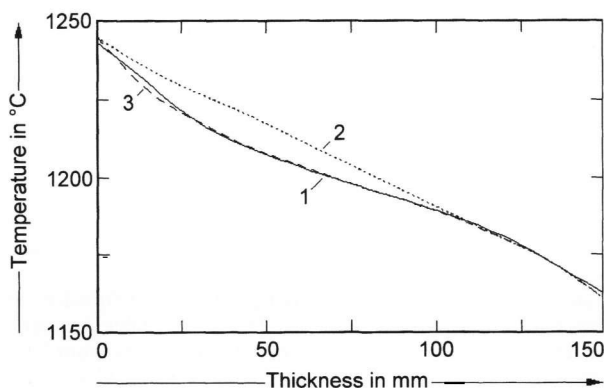


Figure 6. Distribution of glass temperature as a function of glass thickness for light green glass [1; table 1, figure 3, glass no. 4]; curve 1: experiment; curve 2: theory, without heat losses; curve 3: theory, with heat losses.

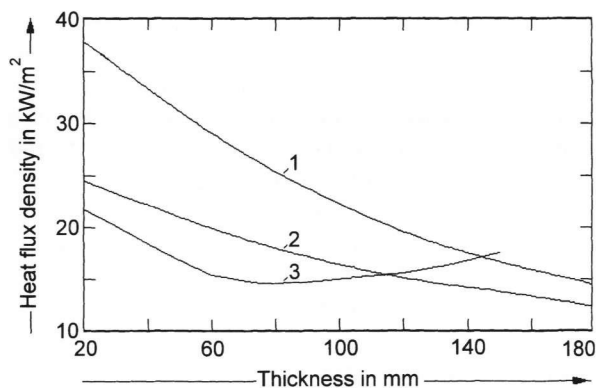


Figure 7. Calculated distribution of heat flux density as a function of glass thickness for three glasses (theory); curve 1: green bottle glass [2; figure 2, table 2, experiment no. 1]; curve 2: green bottle glass [2; figure 2, table 2, experiment no. 3]; curve 3: light green glass [1; table 1, figure 3, glass no. 4].

$$Q(x) = Q_{\max} \left( 1 - \frac{x}{L} \right)$$

and at the value of  $Q_{\max} = -200 \text{ kW m}^{-3}$ , when the heat losses are the highest at the upper surface of the glass melt and the lowest at the bottom of the studied glass.

Absolutely the same situation is characteristic for the analysis of temperature profile of the same glass obtained in the experiment no. 3 [2], which is demonstrated in figure 5. Satisfactory coincidence of the experimental and calculated curves can be achieved in this case also by assuming linear change of heat losses along the depth of the glass layer. The only difference is that for this experiment  $Q_{\max} = \sim 100 \text{ kW m}^{-3}$ . It is to be noted that for experiment no. 3 the highest temperature of the glass was  $1300 \text{ }^\circ\text{C}$ , i.e. experimental data of  $\lambda_r$  could be used in the whole temperature range of calculations (figure 2). Thus, results presented in figure 5 can be considered as a certain confirmation of correct extrapolation of the  $\lambda_r$  dependence for the green bottle glass used in simulation of experiment no. 1 (figure 4).

Results of the same type of simulation of the experimental data given in [1] for the light green glass are presented in figure 6. In this case the heat transfer between the glass surface and the surrounding gas was taken into account. For the coefficient of heat transfer the value specific for the heat transfer between a solid and a gas in conditions of forced convection was used ( $\alpha = 40 \text{ W m}^{-2} \text{ K}^{-1}$ ). It was supposed that the temperature of the crown of the furnace ( $T_{\text{up}}$ ) was  $1240 \text{ }^\circ\text{C}$  and at the furnace bottom ( $T_{\text{down}}$ )  $1160 \text{ }^\circ\text{C}$ . The gas temperature was selected equal to  $1500 \text{ }^\circ\text{C}$  for calculation of curve 2, and to  $1600 \text{ }^\circ\text{C}$  for calculation of curve 3 of figure 6. It can be explained by the heat losses in the upper part of the glass layer and by the increase in the amount of heat in the lower part of the glass layer. Good fit of the experimental and calculated curves was achieved at the following changes in the amount of heat along the  $x$  axis:

$$Q(x) = -250 \left( 1 - \frac{2x}{L} \right)$$

for  $x < \frac{L}{2}$  and

$$Q(x) = 75 \left( \frac{2x}{L} - 1 \right)$$

for  $x > \frac{L}{2}$ .

The mentioned effects lead to the non-uniformity of heat fluxes along the  $x$  axis. Figure 7 presents the calculated distributions of heat fluxes corresponding to calculations of temperature profiles coinciding with the experimental ones (curves 3 in figures 4 to 6).

It follows from figure 7 that in experiments with the green bottle glass described in [2], the heat flux decreased monotonously from the glass surface to its bottom, where it reached the minimal values, namely  $14 \text{ kW m}^{-2}$  in the case of experiment no. 1, and  $12 \text{ kW m}^{-2}$  in the case of experiment no. 3. These values correspond well with the values of the heat fluxes measured by the authors of [2] at the bottom of the glass tank:  $14$  and  $12 \text{ kW m}^{-2}$ , respectively. In [1] the results of experimental determination of heat flux are not presented, and thus it is impossible to compare experimental and calculated values of this quantity.

Changes of heat flux along the  $x$  axis are definitely connected with the temperature gradient along the  $y$  and  $z$  axes of the glass layer. Thus, it seems reasonable to assume that in all experiments with the green bottle glass described in [2], the glass temperature near the walls of the tank is considerably lower than that in the center. Calculations based on the thermal balance show that the mean value of the difference between temperatures at the center of the glass layer and the periphery of this layer,  $\Delta T_h$ , is approximately equal to

$$\Delta T_h = \frac{\langle Q \rangle A}{8 \lambda_s}$$

where  $A$  is the width of the tank,  $\langle Q \rangle$  the approximated (along the layer thickness) value of intensity of decrease or increase in the amount of heat in the center of the layer.

Using these values one can estimate that for conditions of experiments no. 1 and 3 for the green bottle glass in [2],  $\Delta T_h$  was equal to about 50 and 25 K, respectively. In the experiment with the light green glass the average value of  $\Delta T_h$  was equal to about 17 K.

Thus, one has to suppose that in the already discussed experiments reported in [1 and 2] the temperature distribution was far from uniform. It should lead to considerable changes in heat flux along the directions of temperature profile measurements and to quite substantial errors in determination of the values of heat flux through the glass, which the authors of these papers assumed to be equal to the heat flux through the refractories at the bottom of the tank. Probably, these errors were the main reason why there appeared to be such a great discrepancy between reported data on  $\lambda_{\text{eff}}$  and  $\lambda_r$ . However, some additional errors in the discussed measurements could also occur. Some of them will be discussed in sections 5. and 6.

One can give several possible reasons for horizontal temperature gradients in glass melts. However, without a detailed knowledge of the construction of the furnace it makes no sense to discuss them here. At the same time, whatever was the reason (or were the reasons) for temperature gradients, the two conclusions from the authors' analyses seem to be clear enough.

a) It is difficult to ensure a temperature distribution in experimental units for performing the discussed kind of

experiments which are necessary to achieve a reasonable accuracy of measurements.

b) One certainly has to check the absence of temperature gradients in horizontal directions in the course of any new measurement, and if such a gradient were found, one would have to take measures for decreasing it down to the values which would permit to perform correct measurements.

For the most of the glasses studied in [1 and 2] the authors of the present paper were unable (for the already explained reasons) to perform the same kind of calculations which were described in this section. However, it is clear that generally the problems of non-uniform temperature distribution should be expected in all experiments performed by the scientists using the described kind of equipment. It is to be noted that the present authors were not the first to pay attention to the mentioned discrepancies. In respect of data published in [11] serious doubts about the validity of the experimental methods used in that publication was raised by Viskanta et al. [12]. The method of effective conductivity measurements presented in [11] was somewhat different from that described in [1 and 2]. However, the reported results were the same: Effective conductivity was considerably smaller than the radiative one. Unfortunately, the information about the method used in [11] is not sufficient to be employed for a computer simulation program.

## 5. Validity of measurements of temperature profile in glass melts by a thermocouple

As it was mentioned earlier, the temperature profile in molten glass was studied in the investigations discussed here by the use of a thermocouple. It is not only the feature of nearly all studies of effective conductivity coefficients. It is also the usual method of investigation of temperature distribution in any glass melting furnaces. However, this method of glass temperature measurement contains possibilities of considerable errors. It does not mean that the authors are against the use of thermocouples for measurements in question: In most cases it is not possible to propose any reasonable alternative. However, everybody using this method should have an idea about the factors affecting the precision of the obtained results. Then it will be possible to take these factors into account and to avoid too high errors. In this section the authors will try to describe the thermal processes affecting the correctness of glass temperature measurements by thermocouples and to illustrate the influence of these processes using as an example the experimental results presented in [1 and 2].

The value of a thermocouple temperature is determined on the base of balance of heat flux densities coming to and out of its hot junction. This balance could be expressed by the following equation:

$$2 \frac{\lambda_c}{D} (T_{\text{th}} - T) = q_{\text{thr}}^+ - q_{\text{thr}}^- = \lambda_j \frac{d^2}{D^2} |\nabla T| \quad (11)$$

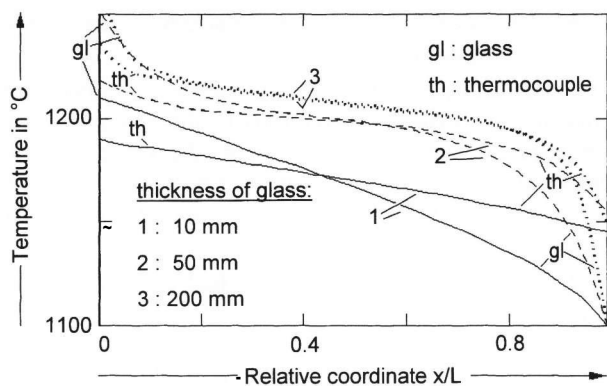


Figure 8. Calculated distribution of temperature for TV-screen glass after [2] (theory).

where  $D$  is the diameter of the thermojunction,  $d$  the diameter of a thermocouple wire,  $T_{th}$  the temperature of the junction,  $q_{thr}^+$  the flux density of radiation energy acquired by the junction,  $q_{thr}^-$  the flux density of energy radiated by the junction,  $\lambda_j$  the coefficient of thermal conductivity of the wire,  $|\nabla T|$  the modulus of temperature gradient along the wire.

The left side of equation (11) is the thermal conductivity flux density from the thermojunction to the surrounding glass. The right side of the equation describes the total radiative heat flux density to the junction and the heat which is going from the junction along the wire. As it follows from equation (11), the temperature of a thermocouple can be equal to that of a surrounding glass only when the right side of equation (11) is equal to zero. This condition could be met if first and foremost the heat flux removed from the thermocouple by the wire were negligibly small.

This heat flux is determined by the temperature gradient along the wire. By comparison of the heat flux value with the thermal conductivity flux from the thermojunction to the melt, one can derive the following condition for the temperature gradient along the thermocouple wire, which permits to disregard the influence of the heat flux along the wire:

$$|\nabla T| \ll 2 \lambda_c / \lambda_j (T_{th} - T) D / d^2. \quad (12)$$

Inserting into equation (12) the following characteristics:

$$\lambda_c = 1 \text{ W/(m K)}; \quad \lambda_j = 100 \text{ W/(m K)},$$

$$D = 2d = 2 \text{ mm}; \quad T_{th} - T = 10 \text{ K}$$

the following condition is obtained:

$$|\nabla T| \ll 400 \text{ K/m}.$$

One more condition of reaching practically equal temperature values of a thermojunction and a melt is the equality of the heat supplied to the junction and the heat removed from it. This condition is satisfied in an

optically thick medium. If the medium is not optically thick, the difference between thermocouple reading and the temperature of the surrounding glass may be appreciable. This problem will be considered in some detail in the following.

Figure 8 demonstrates calculated distributions of temperatures of the glass and a thermocouple for the case of TV-screen glass in the direction of the  $x$  axis. All information needed for such a calculation was taken from [2]. The calculations of temperature distribution in the glass were performed on the base of the one-dimensional mathematical model described in section 4. Temperatures of the thermocouple were calculated by the solution of equation (11). The influence of the heat exchange between the glass surface and the gas was neglected. The diameter of the thermocouple joint was supposed to be equal to 2 mm (the value of diameter does not appreciably affect the results of calculations).

As follows from figure 8, the greatest difference between temperatures of the glass and thermocouple occurs near the borders of the glass layer: Near the surface of the glass the temperature of the glass is higher than that of the thermocouple, near the bottom of the glass layer the situation is opposite.

For an explanation of the described effects some simplifying assumptions are introduced. The absorption spectrum of a certain medium is assumed to consist of two regions, in one of which the medium is absolutely transparent and in the other is absolutely opaque. In the transparent region the medium does not radiate; in the region where the medium can radiate, it is opaque. The part of energy which is radiated by a perfect radiator at a certain temperature within the region of absolute transparency of the medium will be denoted by  $y$ . Then the ratio of the corresponding energy for the opaque region of the medium will be  $1 - y$ . The temperature dependence of  $y$  is neglected for the sake of simplicity. Besides, the conductive heat fluxes in the described medium is thought to be negligible.

At the beginning the thermal balance of the thin surface layer of the medium is considered. The medium will be heated by the radiative heat flux density  $q_r$  from the crown of the furnace in the opaque region of the spectrum. Therefore

$$q_r^+ = (1 - y) \sigma T_{up}^4.$$

The heat radiated by the medium in the direction of the crown is the result of radiation in the same region of the spectrum:

$$q_r^- = (1 - y) \sigma T^4.$$

From the condition of the balance of heat flux densities follows that the temperature on the surface of the medium should be equal to the temperature of the crown:

$$T = T_{up}.$$



Now the temperature of a thermocouple inside the medium is determined. Supply of heat to the thermocouple inside the medium is determined by radiative heat fluxes from the crown and bottom of the furnace in the transparency region of the medium:

$$q_{\text{thr}}^+ \approx \gamma \sigma (T_{\text{up}}^4 + T_{\text{down}}^4).$$

The thermocouple radiates heat also only in the transparency region:

$$q_{\text{thr}}^- = 2 \gamma \sigma T_{\text{th}}^4.$$

The multiplier 2 in the last formula appears due to the fact that the thermocouple radiates the heat in both directions – upward and downward. After equilibration of the heat flux densities the following relation between the temperature of the thermocouple and the temperatures of the crown and bottom of the furnace:

$$T_{\text{th}}^4 = (T_{\text{up}}^4 + T_{\text{down}}^4)/2 \quad (13)$$

is obtained. At the enumerated suppositions the temperature of the thermocouple does not depend on any other factors.

After equation (13) the temperature of the thermocouple should be lower than  $T$ , and accordingly also the temperature of the surface of the medium but it should be higher than the temperature of the medium at the bottom of the tank.

It seems obvious that the stated factors influence the temperatures of the thermocouple inserted into the real glass if the distances between the thermocouple and the upper, or bottom, or both surfaces of the glass layer do not exceed considerably the free path of radiation for the studied glass. In these cases the temperature of the thermocouple should be determined by both the external factors (the influence of which is described by equation (11)) and by the temperature of glass at the distance approximately equal to the free path of radiation.

The dependencies presented in figure 8 will now be considered again. In the case of small thickness of the glass layer the distributions of glass temperatures and thermocouple temperatures are different for the whole depth of the glass layer: Changes in the temperature of the thermocouple are considerably less pronounced than those in glass temperature. With an increase in thickness of the glass layer the two dependencies tend to approach each other. The greatest difference between temperatures of the thermocouple and the glass exists near the borders of the glass layer. It is also seen from figure 8 that the said difference is considerably greater at the bottom of the glass layer than on the surface of the glass.

Experiments with TV-screen glass reported in [2] were performed for a thickness of the glass layer equal to 200 mm. For this case, as it follows from figure 8, the differences between temperatures of the thermocouple and the glass are still quite high. A tendency for the two

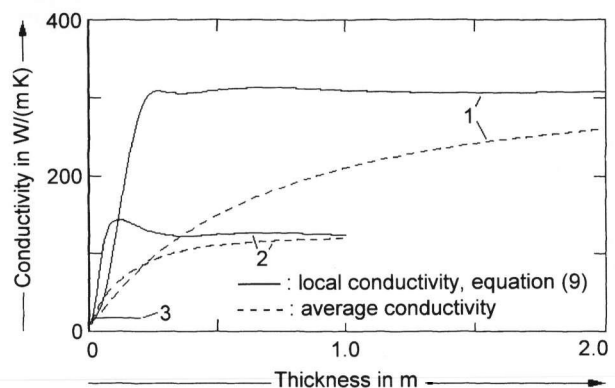


Figure 9. Effective conductivity as a function of glass thickness at a temperature of 1200°C for three glasses; curve 1: TV-screen glass; curve 2: float glass; curve 3: green bottle glass.

dependencies to coincide is observed only at the center of the glass layer. However, such a coincidence alone can not ensure the dependable calculations of thermal conductivity because the performance of these calculations requires the knowledge of the precise enough values of temperature gradients and not the values of temperatures.

Thus, temperature distributions for the TV-screen glass, reported in [2] are correct for the distribution of the temperature of the hot junction of the thermocouple, but differs considerably from the distribution of temperatures in the glass itself. It is evident that temperature curves reported in [1] for the colorless glasses and in [2] for the float and the white bottle glass are also essentially incorrect in regions adjacent to the glass surface and to the bottom of the tank approximately within the free path lengths, which are equal to 30 to 60 mm (figure 1). At the same time the temperatures of the thermocouple, and of the green bottle glass [2] and light green glass should practically coincide. This particular fact permitted to analyze results for these glasses in sections 4. and 5. of the present paper.

## 6. Conclusion

As it follows from this work, it is possible in principle to use measurements of temperature gradient and thermal flux for a precise determination of the total conductivity of glass melts. However, one has to satisfy two main conditions: first, to ensure practical absence of temperature gradients in a glass melt in directions parallel to the surface of the glass, and second, to ensure that the depth of glass layer will be considerably greater than the free path of radiation in the studied glass. Thus, the more transparent the glass, the bigger should be the tank for correct measurements of the total conductivity of this glass.

Figure 9 shows calculated dependencies of effective conductivity on the thickness of glass layers for several glasses with different values of  $l$ . It was found that calculations basing on the theory of heat transfer for infi-

nite layers (equations (2 to 8)) lead to incorrect results for transparent glasses due to the fact that in real measurement units distances between the center of a tank and its side walls are of the same order as those between the center and upper and bottom surfaces of a glass layer. Accordingly, for estimation of dependencies demonstrated by figure 9 the approximated version of calculations was used, where each microvolume of glass was supposed to radiate only in three directions =  $x$ ,  $y$  and  $z$ . For cases when  $L \gg l$ , the results of such calculations fit in well with those obtained by using equations (2 to 8).

Figure 9 illustrates two rather obvious points. For glasses with greatly different values of  $\lambda_{\text{eff}}$  tanks of quite different sizes have to be built. And for determination of minimal dimensions of tanks used for total conductivity measurements, one has to know the value of  $l$  for each particular glass in advance. To check the condition  $l \ll I$ , one has to calculate the value of  $l$  in the whole temperature range of the planned experiment and hence to know in advance the absorption spectra of the glass in question in this temperature range. On the other hand, if these spectra are known it is possible to determine the coefficient of radiative conductivity by using equation (9) independently of any additional conditions. Because at melting temperatures or transparent enough glasses, the radiative heat transfer plays the predominant role and thus, experiments for measuring the effective conductivity could appear unnecessary.

It is also to be noted that the knowledge of absorption spectra permits to use various mathematical models for calculating the thermal fields in glass media of any form and dimensions. At the same time coefficients of effective conductivity could be used only for simulation of thermal fields in large glass tanks. Even in these cases the precision of calculation of heat fluxes in areas of high temperature gradients (i.e., near the borders of the glass body, the source of heat, etc.) could be absolutely unsatisfactory.

This discussion inevitably leads to the conclusion that the precise measurements of the absorption spectrum of a glass in the whole temperature range of interest and in the broad enough range of wavelengths (at high temperatures it should be extended down to about  $0.5 \mu\text{m}$ ) is a much more reasonable way for obtaining precise characteristics for heat flux simulation than the method of temperature gradient determination. Certainly, for correct measurements of absorption spectra at high temperatures one has to overcome a number of difficulties. Analyses of some of them will be done in the second part of the present paper.

It is quite obvious that the progress in computer simulation procedures will lead to demands for more and more precise heat transfer characteristics of glass melts. Thus, ways of precise enough determination of thermal conductivity coefficients in high-temperature glass melts have to be considered. Probably, the only possible way to do this is to perform the measurements

of effective conductivity when the influence of radiative heat transfer on the effective conductivity coefficient is comparatively small. Figure 9 shows that in order to do this one should perform measurements in conditions when the glass layer between two metallic plates is thin enough. To obtain correct results in this case one has to satisfy several special conditions, including the high reflection coefficient of the surfaces of the metallic plates. In principle, however, such experiments are quite possible as was demonstrated recently by Tyutyunnik [8].

## 7. Nomenclature

### 7.1. Symbols

$A$	width of tank in m
$c$	specific heat of melt in $\text{J kg}^{-1} \text{K}^{-1}$
$D$	diameter of thermocouple junction in m
$d$	diameter of thermocouple wire in m
$I$	spectral intensity of radiation in $\text{J m}^{-2}$
$I_{\text{ve}}$	spectral intensity of radiation of the perfect radiator in $\text{J m}^{-2}$
$L$	height of glass layer in m
$l$	spectral free path of radiation determined by Rosseland formula in m
$l_v$	spectral free path of radiation in m
$n$	refractive index of medium
$\langle Q \rangle$	approximated (along the layer thickness) value of intensity of decrease or increase in the amount of heat in the center of the layer in $\text{kW m}^{-3}$
$Q_{\text{max}}$	heat losses in $\text{kW m}^{-3}$
$q_{\text{eff}}$	total heat flux density in the direction of $x$ axis in $\text{kW m}^{-2}$
$q_r$	density of radiative flux in the direction of $x$ axis in $\text{kW m}^{-2}$
$q_{\text{thr}}^+$	density of radiative flux directed to the thermocouple in $\text{kW m}^{-2}$
$q_{\text{thr}}^-$	density of the flux radiated by thermocouple in $\text{kW m}^{-2}$
$\vec{q}_c$	vector of conductive heat flux density in $\text{kW m}^{-2}$
$\vec{q}_r$	vector of radiative flux density in $\text{kW m}^{-2}$
$\vec{q}_s$	vector of total flux density in $\text{kW m}^{-2}$
$r$	coefficient of reflection
$T$	absolute temperature in K
$T_{\text{down}}$	temperature at the furnace bottom in K
$T_{\text{gas}}$	temperature of gas at the glass surface in K
$\Delta T_{\text{h}}$	difference between temperature at the center of the glass layer and the periphery of this layer
$T_{\text{th}}$	temperature of thermocouple in K
$T_{\text{up}}$	temperature of the furnace crown in K
$t$	time in s
$x$	coordinate counted from the upper surface of the glass ( $0 < x < L$ ) in m
$y$	part of energy which is radiated by a perfect radiator at a certain temperature within the region of absolute transparency of the medium
$\alpha$	coefficient of heat transfer describing the heat exchange between glass and gas in $\text{W m}^{-2} \text{K}^{-1}$
$\kappa_v$	spectral absorption coefficient of studied medium in $\text{m}^{-1}$
$\lambda_c$	thermal conductivity coefficient in $\text{W m}^{-1} \text{K}^{-1}$
$\lambda_{\text{eff}}$	effective conductivity coefficient in $\text{W m}^{-1} \text{K}^{-1}$
$\lambda_r$	radiative conductivity coefficient in $\text{W m}^{-1} \text{K}^{-1}$
$\lambda_s$	total conductivity coefficient in $\text{W m}^{-1} \text{K}^{-1}$
$\mu$	independent variable
$\nu$	frequency of radiation in $\text{s}^{-1}$
$\sigma$	Stefan-Boltzmann constant ( $5.67 \cdot 10^{-8} \text{W m}^{-2} \text{K}^{-4}$ )
$\nabla$	gradient operator in $\text{m}^{-1}$

### 7.2. Superscripts

ex	exterior
in	interior
+	plus
=	minus

### 7.3. Subscripts

c	conductive
down	down
eff	effective
gas	gas
r	radiative
s	total
th	thermocouple
up	up
v	frequency

\*

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