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Indentation tests for the investigation of the plasticity of glasses

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Various types of indentation tests are used to investigate the plasticity of glassy materials. It is shown that indentation creep tests performed with both flat ended cylindrical and hemispherical indenters are suitable for viscosity measurements in the viscosity range of 10^8–10^{11} Pa·s. It is also shown that the activation energy of the flow process can be evaluated from the results of indentation measurements.

1. Introduction

It is well known that glass, being a Newtonian liquid, deforms by viscous flow, which can be described by the linear relationship

$$\tau = \eta\dot{\gamma} \tag{1}$$

where τ is the shear stress, $\dot{\gamma}$ is the shear strain rate and η is the viscosity of the glass, which depends on the material and temperature. A knowledge of the viscosity value of a glass is the most important factor in determining the correct shaping processes to be applied in the production of glass articles. The range of viscosity is very wide and it is not possible to use one method to make measurements over the whole range. In the high temperature range, the most widely used techniques used in viscosity measurements are the rotation viscometer and the counterbalanced sphere method. For low temperature measurements the most favoured methods are fibre extension and the parallel plate techniques [1].

The viscosity of glasses strongly depends on the temperature and generally obeys the formula:

$$\eta = \eta_0 e^{\frac{Q}{kT}} \tag{2}$$

where η_0 is a constant and Q is the activation energy of the process controlling the viscous flow. Other symbols have the usual meaning. Substituting Equation 2 into Equation 1 and expressing $\dot{\gamma}$ from it, we obtain:

$$\dot{\gamma} = \frac{1}{\eta_0}\tau e^{-\frac{Q}{kT}}. \tag{3}$$

This formula can be regarded as a particular case of the equation:

$$\dot{\gamma} = A\tau^n e^{-\frac{Q}{kT}} \tag{4}$$

which is generally used for the interpretation of the high temperature steady state creep of metals and ceramics. In Equation 4 A is a material specific constant, Q and n are the activation energy and the stress exponent of the activation process, respectively. In the case of a Newtonian viscous flow the value of the stress exponent is exactly 1 and then Equation 4 reverts back to Equation 3. For metals and ceramics, due to the presence of crystalline phases, the value of the stress exponent varies between 3 and 5 [2] indicating that these materials do not deform by viscous flow. In some ceramic materials, which contain glassy phases, the high temperature deformation process is controlled by the diffusion of molecules in the glassy phases which greatly improves the plasticity of the material. Therefore the value of the stress exponent decreases to about 2 [3].

It is also well known that the value of the activation energy, Q, strongly depends on the bonding in the solid state. During the deformation of Al alloys, for instance, due to the metallic bonds the value of Q is usually commensurate with that of the self-diffusion of the base metal ($\sim 130\ \mathrm{kJ\,mol^{-1}}$). In the cases of glasses, glass-ceramics and ceramics the existence of stable and strong ionic or covalent bonds means that the activation energy can be very high ($> 400\ \mathrm{kJ\,mol^{-1}}$). It has been suggested [4, 5], that the value of the activation energy, Q, together with that of the stress exponent, n, clearly distinguishes between metals, glasses and ceramics from the point of view of high temperature mechanical properties. It again emphasizes the importance of the n and Q parameters in the characterization of high temperature creep.

As was previously mentioned, the most favoured method for the determination of the viscosity of glasses as well as for the investigation of the high temperature plasticity of solids is the uniaxial tensile test. The phenomenological interpretation of this test is relatively simple, since the stress field is macroscopically uniaxial and homogeneous in the sample during the test. However, in the case of some glasses, which are not easily drawn into fibres or in the case of advanced structural ceramics for which the preparation of

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tensile samples is very difficult, other methods should be used.

In this paper a short description of an indentation tester developed by our research team and some results obtained from its application to glassy materials are presented. Comparing the results obtained by different methods it will be shown that indentation measurements can also be applied to the determination of the viscosity of glasses.

1.1. The basic idea and technical aspects of the indentation test

The impression tests such as the Brinell, Vickers and Meyer hardness tests and the simple compression test are often used for the investigation of the high temperature mechanical behaviour of materials [6]. These tests are technically simpler than the tensile test, but the correct physical interpretation of the deformation process taking place during these impression tests is very intricate, because the stress field under the indenter is multiaxial and inhomogeneous. However, there are well-established empirical rules for the comparison of the results of indentation and tensile tests.

The indentation creep test performed with a flat ended cylindrical punch was proposed originally by Yu and Li who, using this method for the investigation of the high temperature plasticity of single crystals, have shown that their results are equivalent to those of a tensile creep test [7, 8]. Using the idea of Yu and Li we have developed the indentation creep equipment shown schematically in Fig. 1. The sample of approximate size of $10 \times 10 \times 5$ mm lies on the sample holder in the central region of an electrically heated furnace tube. The temperature of the sample is continuously measured by a thermocouple and the heating of the furnace is automatically controlled. During indentation measurements a cylindrical or a hemispherical punch is pressed into the surface of the sample. The indentation depth, h, of the punch is recorded as a function of time, t, by a linear variable differential transformer (LVDT) unit, which is connected to a computer.

Equation 1 implies that indentation creep tests can be used for the determination of the viscosity of glasses. Several authors have considered the possibility of the evaluation of impression results to obtain viscosity values [9, 10] and it has been generally declared that absolute viscosity measurements can only be performed by equipment containing hemispherical punches. The application of indenters of other shapes (for example a cylindrical punch) has not been considered [11]. However, this situation, is not generally supported by reasonable arguments.

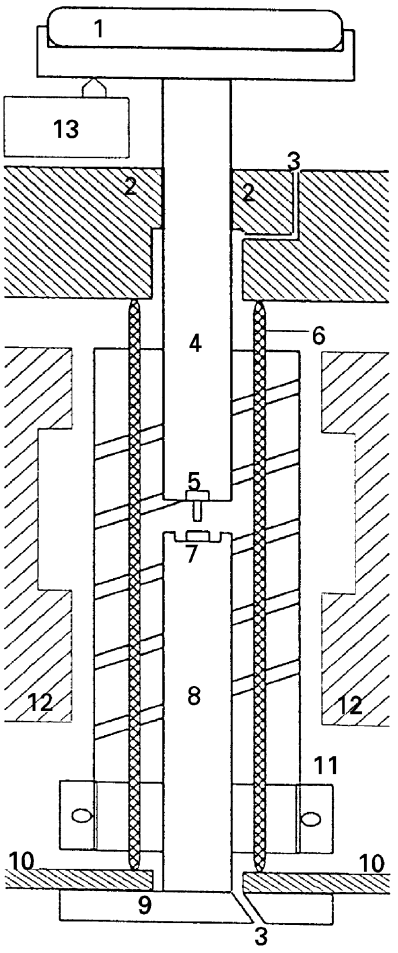

Figure 1 Schematic picture of the indentation tester. key; (1) load (F), (2) bush of load, (3) gas inlet, (4) loader-rod, (5) indenter, (6) worktube, (7) sample holder, (8) sample holder-rod, (9) holder support, (10) support, (11) heating element, (12) insulation and (13) displacement measuring device.

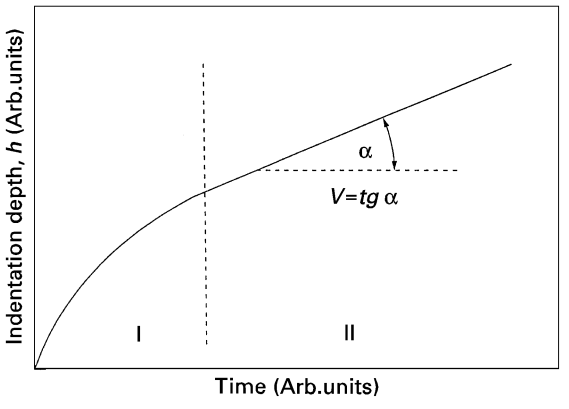

Figure 2 Schematic creep curve taken by using a cylindrical indenter with a constant load at a constant temperature.

1.1.1. The indentation test with a cylindrical punch

Fig. 2 shows a schematic creep curve taken during an indentation test performed by a flat ended cylindrical punch. Applying a constant load, F, at a constant temperature, T, after a short initial transient the punch penetrates at a constant impression velocity, v, into the sample. Several authors have shown that in order to compare the results of impression creep and those of uniaxial tensile tests the impression velocity,

v, and the load, F, should be converted into the equivalent tensile stress, σ, and strain rate, $\dot{\varepsilon}$, using the formulas [7, 12–18]:

$$\sigma = \frac{p}{3} = \frac{4F}{3\pi d^2} \tag{5}$$

$$\dot{\varepsilon} = \frac{v}{d} \tag{6}$$

where p is the pressure just below the punch and d is the diameter of the punch. The applicability of the impression creep test and the validity of Equations 5 and 6 have been shown for several metals, alloys and ionic crystals [12, 16–21]. In some cases the applied arguments are based on continuum mechanics, and we therefore consider that Equations 5 and 6 are applicable to glassy materials. This means that using Equations 5 and 6 an equivalent stress–strain rate (σ–$\dot{\varepsilon}$) function can be constructed from the indentation creep data.This function corresponds to the data of a uniaxial tensile test, which can be described for Newtonian viscous flow by the following equation:

$$\sigma = \mu\dot{\varepsilon} \tag{7}$$

where μ is a constant characterizing the viscous flow.

Supposing that the volume constancy criteria is satisfied the following well known equations can be applied:

$$\tau = \frac{\sigma}{2} \tag{8}$$

$$\dot{\gamma} = \frac{3}{2}\dot{\varepsilon}, \tag{9}$$

and then Equation 7 can be converted into the relationship:

$$\tau = \frac{\mu}{3}\dot{\gamma}\,. \tag{10}$$

Comparing Equation 10 to Equation 1 which defines the viscosity we obtain:

$$\eta = \frac{\mu}{3} \tag{11}$$

which means that the viscosity coefficient can be evaluated from the equivalent stress–equivalent strain rate function. We note that the evaluation of the fibre extension measurement is executed essentially in the same manner. We also emphasize that Equations 10 and 11 are strictly correct only for a uniaxial tensile field and not for the multiaxial inhomogeneous field found under the punch during an indentation test. Therefore, a correction factor might need to be introduced in order to obtain the viscosity from the indentation measurements.

1.1.2. Indentation test with a hemispherical punch

The pressure continuously alters under a hemispherical indenter, due to the geometry of the punch. (The pressure is $p = F/A$, where F is the applied load and A is the area of the projection of the contact surface between the punch and the sample to the plane perpendicular to the direction of the load.) This means that the deformation state, contrary to that developed under a cylindrical punch, is not in a steady state because the penetration velocity of the hemispherical indenter continuously decreases as is shown schematically in Fig. 3. This is why measurements performed with a hemispherical punch cannot realistically provide information on the interpreted creep of metals. However, indentation tests using a hemispherical indenter are valid for the determination of the viscosity of glasses. The theoretical interpretation of these measurements was developed by Douglas *et al.* [10] who derived the formula:

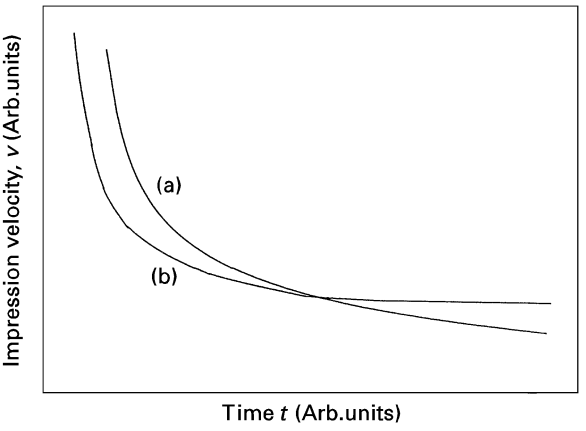

Figure 3 The variation of the velocity of (a) a hemispherical and (b) a cylindrical indenter pressed with constant load.

$$\frac{\mathrm{d}h}{\mathrm{d}t} = \frac{3F}{16\eta}\,\frac{1}{(h(d-h))^{1/2}} \tag{12}$$

from which the viscosity can be assessed from experimental data. In Equation 12 $\mathrm{d}h/\mathrm{d}t$ is the indentation rate, and the meaning of the other parameters is the same as in the case of the cylindrical punch. Direct integration of Equation 12 is difficult and its result would be unsuitable for practical computations. However, assuming that $d \gg h$, Equation 12 can be rewritten in the following form:

$$\frac{\mathrm{d}h}{\mathrm{d}t} = \frac{3F}{16\eta}\,\frac{1}{(dh)^{1/2}} \tag{13}$$

from which

$$h^{3/2} = \frac{9}{32\eta(d)^{1/2}}\,Ft \tag{14}$$

and then

$$\eta = \frac{9}{32(d)^{1/2}}\,\frac{Ft}{h^{3/2}}. \tag{15}$$

Equation 15 can be applied relatively easily for the determination of viscosity from experimental data taken with a hemispherical punch. Experimental data confirm that the deviation between results obtained by the use of Equation 15 and from the full theoretical form of Equation 12 can be neglected. In the case of a punch with a diameter of about 2 mm at large

penetration depths (0.3 mm) the deviation between these two results is only about 5%. If the depth is 0.05 mm this deviation decreases to about 1%.

2. Experimental procedure

The viscosity of three different glass types was determined using an indentation test with various sized cylindrical and hemispherical punches, the diameter, d, was between 1.2–3 and 2–4 mm, respectively. One of the glasses was a common magnesia-silica which will be denoted in the future as material A. The other two are alkali-silica glass (material B) and lead glass (material C), which were made in the Bródy Laboratory of Tungsram-G. E., Budapest. The indentation measurements were carried out at different loads in the temperature range of 450–600 °C.

3. Results and discussion

3.1. Determination of the viscosity of glasses

3.1.1. Indentation measurements with a cylindrical punch

Fig. 4(a and b) shows a series of indentation curves taken using a cylindrical punch at different conditions on samples of material A. For all the investigated materials the indentation curves are similar to these curves and to that shown in Fig. 2. Using Equations 1 and 5–7 the following formula can be obtained for the viscosity at constant temperature:

$$p = c\eta v \tag{16}$$

where $c = 9/d$ is a constant for the same punch. On the basis of Equation 16 the viscosity can be evaluated from the slope of the line fitted to p–v data.

Fig. 5 shows the p–v relationships obtained at different temperatures on the investigated glasses. It can be seen that the relationship between the p–v data is linear in each case. It is worth noting that this linearity proves that the value of the stress exponent, is really 1 for the glasses investigated. (Values of the viscosity determined on the basis of Equation 16 will be compared to those obtained by other methods, in section 3.1.3).

On the basis of knowledge of the viscosity, η, the activation energy, Q, of the viscous flow can be determined using Equation 2. It can be obtained from the slope of the straight line fitted to $\ln\eta$–$1/T$ data points. However, we can also find another procedure for obtaining the activation energy, Q, directly from the indentation data. Taking into account Equations 2 and 16 one can derive the formula:

$$p = c\eta_0 v e^{\frac{Q}{kT}} \tag{17}$$

from which:

$$\ln p = \ln(c\eta_0) + \ln v + \frac{Q}{kT} \tag{18}$$

or

$$\ln v = \ln p - \ln(c\eta_0) - \frac{Q}{kT} \tag{19}$$

This equation shows that the activation energy can be determined from the slope of the straight line fitted to $\ln v$–$1/T$ data points. Fig. 6 shows these lines for the investigated materials. It can be clearly seen that this method does not need any information about the viscosity values. Applying this method, the activation energies were 450, 550 and 410 kJ mol^{-1} for glasses A,

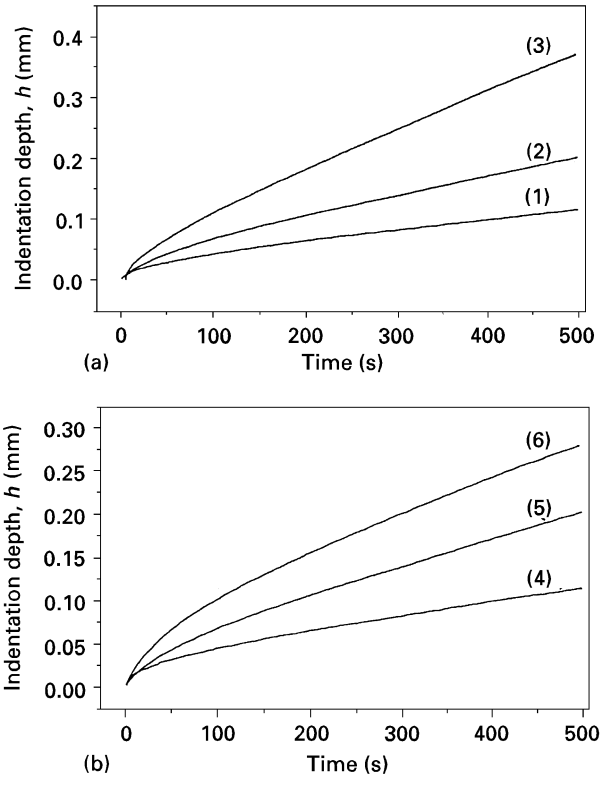

Figure 4 Indentation curves taken with a cylindrical punch for material A (a) at a constant pressure of 11.37 MPa, at temperatures of (1) 588 °C, (2) 598 °C and (3) 604 °C and (b) at a constant temperature of 598 °C and at pressures of (4) 5.66 MPa, (5) 11.32 MPa and (6) 14.15 MPa.

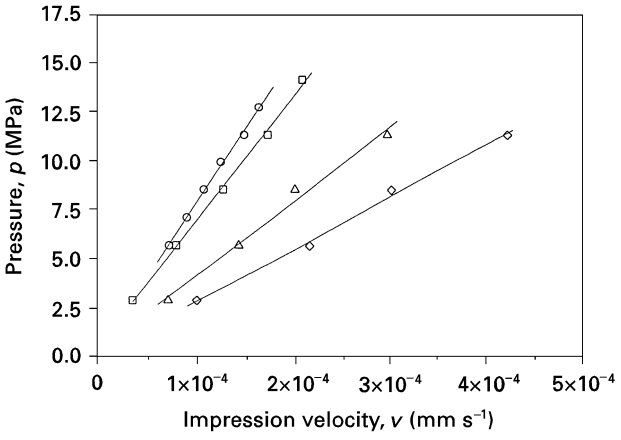

Figure 5 The p–v functions for material A at temperatures of (○) 589 °C, (□) 593 °C, (△) 598 °C and (◇) 604 °C. The viscosity values at these temperatures were; 1.67×10^{10} Pas, 1.41×10^{10} Pas, 8.4×10^{9} Pas and 5.9×10^{9} Pas respectively.

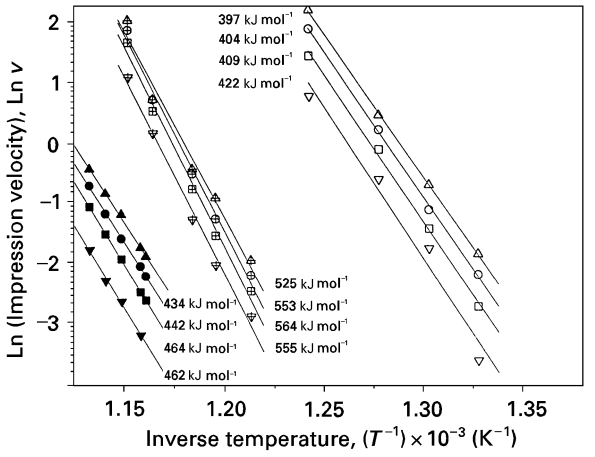

Figure 6 The $\ln v$–$1/T$ functions taken by indentation measurements with a cylindrical punch for the determination of the activation energy. For sample A the loads (F) used were; (▼) 20 N, (■) 40 N, (●) 60 N and 80 N. For sample B the loads (F) used were; (▽) 20 N, (⊞) 40 N, (⊕) 60 N and (◬) 80 N. For sample C the loads used were; (▽) 20 N, (□) 40 N, (○) 60 N and (△) 80 N.

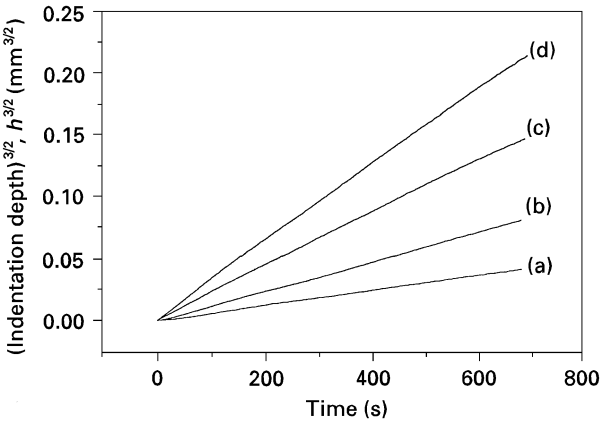

Figure 7 The $h^{3/2}$–t functions for material A at a temperature of 598 °C at a load of (a) 10 N which results in a viscosity of 2.67×10^{10}Pas, (b) 20 N which results in a viscosity of 2.72×10^{10}Pas, (c) 30 N which results in a viscosity of 2.61×10^{10} Pas and (d) 40 N which results in a viscosity of 2.51×10^{10} Pas.

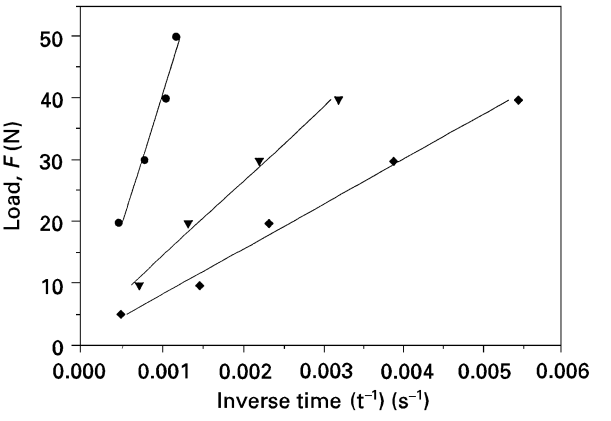

Figure 8 The F–$1/t$ functions for material A at an indentation depth, h, of 215 μm at temperatures of: (●) 588 °C and (▼) 598 °C and (◆) 604 °C.

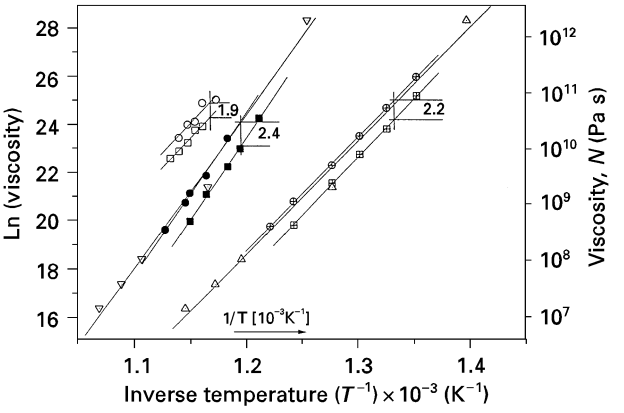

Figure 9 The $\ln \eta$–$1/T$ functions for material A (○) spherical indentor with an associated Q 408 kJ mol^{-1}, (□) cylindrical indentor with an associated Q of 406 kJ mol^{-1} and (▽) elongation with an associated Q of 524 kJ mol^{-1}. For material B, (●) spherical indentor with an associated Q of 557 kJ mol^{-1} and (■) cylindrical indentor with an associated Q of 555 kJ mol^{-1}. For material C: (△) elongation with an associated Q of 386 kJ mol^{-1}. (⊕) spherical indentor with an associated Q of 387 kJ mol^{-1} and (⊞) cylindrical indentor with an associated Q of 397 kJ mol^{-1}.

B and C, respectively. The accuracy of these values is about 10%. The relationship between the activation energies and the structure of the investigated materials will be discussed in a future paper.

3.1.2. Indentation measurements with a hemispherical punch

Our measurements support the validity of Equation 15 derived by Douglas *et al.* [10]. Each measurement shows that applying a constant load and a punch of the same diameter, the value of $h^{3/2}$ varies linearly with time, as can be seen in Fig. 7 for material A. The linear dependence of the load, F, on the reciprocal time, $1/t$, at a constant indentation depth, h, (see Fig. 8 for glass A) also confirms the validity of Equation 15. This linear connection is valid for all the investigated glasses. Similar relationships were found for each investigated glass.

3.1.3. Comparison of the results obtained by the different methods

In this section the results obtained from indentation tests performed with cylindrical and hemispherical indenters are compared with each other and also with the results of control measurements made with the conventional fibre elongation method at the Bródy Laboratory of Tungsram-G. E., Budapest. To execute this comparison and in order to determine the activation energy, $\ln \eta$–$1/T$ diagrams were constructed from the viscosity data determined by inserting experimental data into Equations 15 and 16.

Fig. 9 shows these $\ln \eta$–$1/T$ diagrams for the investigated glasses. It can be clearly seen that the activation energies obtained from the slope of $\ln \eta$–$1/T$ lines closely agree with each other. These values are also in good agreement with those determined from the slope of $\ln v$–$1/T$ lines produced in the way described in section 3.1.1. The good agreement between the

TABLE I Summary of the experimental results for samples A, B and C

Method	Sample	η (550 °C) (Pas)	$\Delta\eta/\eta$ (%)	$Q(\ln\eta - 1/T)$ (kJ mol^{-1})	$\Delta Q/Q$ (%)	$Q(\ln v - 1/T)$ (kJ mol^{-1})	$\Delta Q/Q$ (%)
Fibre elongation	B	8.9×10^{10}	–	524	–	–	–
	C	2.4×10^{8}	–	386	–	–	–
Semispherical indenter	A	5.5×10^{11}	$\pm$ 14	408	$\pm$ 18	–	–
	B	1.0×10^{11}	$\pm$ 15	557	$\pm$ 9	–	–
	C	2.7×10^{8}	$\pm$ 17	387	$\pm$ 7	–	–
Cylindrical indenter	A	2.9×10^{11}	$\pm$ 8	406	$\pm$ 11	450	$\pm$ 13
	B	4.0×10^{10}	$\pm$ 9	555	$\pm$ 8	549	$\pm$ 9
	C	1.2×10^{8}	$\pm$ 10	397	$\pm$ 5	408	$\pm$ 6

activation energies obtained by different methods convincingly proves the applicability of the indentation test for the investigation of high temperature processes occurring in glasses. For the sake of easy comparison the experimental results obtained by different methods are summarized in Table I.

The viscosity values determined by the extrapolation of $\ln\eta - 1/T$ lines at $T = 550\,^{\circ}\mathrm{C}$ can also be seen in Table I. These data and the $\ln\eta - 1/T$ functions of Fig. 9 show that the viscosities evaluated from the fibre elongation tests and the indentation measurements performed with a hemispherical punch agree each other within the error. However, these values are significantly higher than those obtained from indentation tests performed with cylindrical punches. According to our measurements at a given temperature $\eta_e = \eta_h$ for each investigated material, where η_e and η_h are the viscosities evaluated from fibre elongation tests and indentation tests carried out by hemispherical punches, respectively. At the same time for material A:

$$\frac{\eta_e}{\eta_c} = \frac{\eta_s}{\eta_c} = C_A \cong 1.9, \tag{20}$$

for material B

$$\frac{\eta_e}{\eta_c} = \frac{\eta_s}{\eta_c} = C_B \cong 2.4, \tag{21}$$

and for material C

$$\frac{\eta_e}{\eta_c} = \frac{\eta_s}{\eta_c} = C_C \cong 2.2 \tag{22}$$

where η_c denotes the viscosity value assessed from indentation measurements with a flat ended cylindrical punch.

Taking into account the relatively high error in the indentation measurements it seems that the C_A, C_B, C_C values express only the deviation of the geometry of the hemispherical and the cylindrical punch and do not depend on material properties. Consequently we can accept that $C_A = C_B = C_C = 2.2$. This means that the viscosity of glasses can be estimated with good accuracy from measurements performed with a cylindrical punch.

It is worth making a parallel between the equation obtained for an indentation test with a cylindrical punch and Stokes formulas ($F = 6\eta\pi rv$). Taking into account that $p = 4F/\pi d^2$, Equation 16 can be written as:

$$\frac{4F}{\pi d^2} = \frac{18}{2d}\,\eta v \tag{23}$$

and then

$$F = \frac{18}{4}\,\eta\pi r v \tag{24}$$

where $r = d/2$ is the radius of the punch. Equation 24 implies that during an indentation test the empirical Stokes formula can be used in the viscosity determination (with a change of the constant from 6 to 4.5). Using Equation 24 the viscosity of glasses can be simply determined from an indentation test with a cylindrical punch.

4. Conclusions

It has been shown that indentation creep measurements performed with both cylindrical and hemispherical punches can be successfully used for the determination of the viscosity of glasses in the range of about 10^8–10^{11} Pa·s. The viscosity values obtained from indentation tests performed using a hemispherical punch closely agree with those determined by the conventional fibre elongation method. Measurements performed with cylindrical punches can be evaluated by using a theoretically supported formula which is analogous to that of Stokes. However, in order to obtain an exact viscosity value for the investigated materials the proportionality factor in the Stokes formula has to be modified from 6 to 4.5. It has also been shown that the activation energy of the viscous flow can be assessed from the indentation measurements without the evaluation of the viscosity coefficient.

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