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VISCOSITY OF SOME BASALTIC GLASSES AT ONE ATMOSPHERE

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ABSTRACT

The viscosity of glass fibres drawn from liquid basalt were measured in air using a fibre-extension method. At each temperature, constant rates of extension were determined using an optical lever system of magnification. The rate of fibre extension was proportional to the applied load at constant temperature indicating Newtonian flow behavior. Viscosities of the four compositions were between $\sim 10^{14}$-10^{10} poises at temperatures between 600-750°C. For example, a glass of alkaline olivine basalt composition at $T = 637, 689, 741$°C had $\log_{10}\eta$ (poise) $= 13.20, 11.60, 10.45$ and $E\eta = 115$ kcal/mole. In agreement with observations on other glass systems, the temperature where each glass had a viscosity of 10^{13} poise was approximately two-thirds the liquidus temperature. These results were combined with high-temperature (1100-1400°C) melt viscosities by computing the curve of best fit for each composition using the Fulcher equation $\log_{10}\eta = A + B/(T - T_o)$, where T is the temperature in °C and A, B and T_o are constants. These equations allow the calculation of basaltic melt and glass viscosities over the entire range from the melt region to the glass transition point.

SOMMAIRE

La viscosité de fibres de verres étirées à partir d'un liquide basaltique a été mesurée à l'air par la méthode à étirement de fibre. A chaque températude des taux d'extension constants furent mesurés par un système magnifiant à levier optique. Le taux d'extension des fibres était proportionnel à la charge appliquée à température constante, indiquant un comportement de fluide newtonien. Les viscosités pour les quatre compositions étaient entre $\sim 10^{14}$ et 10^{10} poises à des températures de 600-750°C. Par exemple un verre avec une composition de basalte alcalin à olivine à $T = 637, 689, 741$°C avait $\log_{10}\eta$ (poise) $= 13.20, 11.60, 10.45$ et $E\eta = 115$ kcal/mole. La température à laquelle chaque verre avait une viscosité de 10^{13} poises était approximativement les deux tiers de la température du liquide, en accord avec les observations fait sur d'autres systèmes fondus. Ces résultats furent combinés avec les viscosités à haute température (1100-1400°C) et la courbe optimale pour chaque composition calculée au moyen de l'équation de Fulcher: $\log_{10}\eta = A + B/(T - T_o)$ ou T est la température en °C et A, B et T_o sont des constantes. Ces équations permettent le calcul des viscosités de liquides et verres basaltiques depuis le domaine liquide jusqu'au point de transition des verres.

INTRODUCTION

A knowledge of the viscosity of melts is important to many aspects of geochemistry and geophysics. Silicate melts may be studied at high temperatures as liquids, or at lower temperatures as supercooled liquids or glass. Although there are abundant data on the rheological properties of simple glass systems (e.g. Morey 1954; Eitel 1965; Rawson 1967), little is known about the viscous behavior of supercooled basaltic melts below the liquidus and above the glass transition point.

This study was performed as a sequel to viscosity measurements at high temperatures (Scarfe 1973). The purpose was to determine whether meaningful viscosities could be obtained for basaltic glasses at low temperatures (600-750°C); to find the glass transition temperature (T_g) where the glasses have a viscosity of 10^{13} poise; to compare activation energies for viscous flow and glass transition temperatures to values published for glasses of simpler chemistry; and to relate these low-temperature glass viscosities to high-temperature (1150-1400°C) melt viscosities (Scarfe 1973).

EXPERIMENTAL METHOD

The rocks listed in Table 1 were powdered and melted to liquids free from bubbles and crystals at 1400-1500°C for two hours. No significant changes in the composition of the charges occurred during the fusions which were carried out in alumina crucibles in air. Glass fibres drawn from the liquids were ~10 cm in length and 0.5 mm in diameter. The dimensions were not considered critical as long as the diameter varied by not more than 0.05 mm over the length (Dingwall & Moore 1954). Small knobs were melted at the ends of each fibre to permit the attachment of metal support rods (Fig. 1). A fibre was suspended in the hot zone of the furnace and a weight attached to the free end. Constant rates of extension were measured between 600-750°C using an optical lever system of magnification (Dingwall & Moore 1954). Temperatures had an uncertainty of ±5°C and rates of extension could be measured to better than ±1% for all except the fastest rates. All experiments were conducted in air.

Results and Discussion

Viscosities were calculated from the equation for the extension of a fibre under a load

$$\eta = \frac{Mgl}{3\pi r^2 v} \qquad (1)$$

where M is the load, g is the acceleration due to gravity, l and r are the fibre length and radius, and v is the rate of extension (Lillie 1931; Napolitano & Hawkins 1964).

At constant temperature there is a linear relationship between the rate of fibre extension and the applied load (Fig. 2). This is equivalent to a graph of shear rate against shear stress, which for Newtonian viscous flow is a straight line through the origin. These linear relationships are considered strong evidence that incipient crystallization did not occur during the experiments.

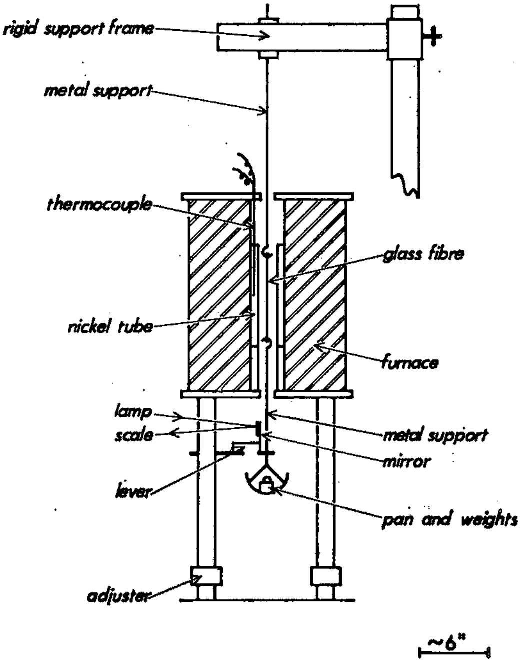

Fig. 1. Apparatus for viscosity measurements using a fibre-extension method.

TABLE 1

Chemical analyses of rocks used to make glass

	1*	2*	5**	7*
SiO_2	56.48	52.51	47.09	40.81
TiO_2	0.68	1.04	2.35	2.68
Al_2O_3	16.93	14.71	14.44	12.49
Fe_2O_3	1.57	2.64	3.83	7.31
FeO	5.77	7.47	9.53*	6.21
MnO	0.12	0.17	0.19	0.24
MgO	5.78	6.29	7.60	7.23
CaO	8.00	10.53	8.26	12.51
Na_2O	2.33	2.36	2.71	3.08
K_2O	0.89	0.72	1.04	1.86
H_2O^+	0.74	0.65	1.66*	2.64
H_2O^-	0.50	0.75	0.70*	1.88
P_2O_5	0.11	0.14	0.47	0.87
TOTAL	99.90	99.98	99.87	99.81

* Wet chemical analyses: A. Stelmach
** XRF analysis: J.G. Holland
1. Basaltic andesite (Mt. Pihanga, New Zealand) collected by P.G. Harris
2. Tholeiite (Antrim, N. Ireland) collected by T.F. Johnston
5. Alkaline olivine basalt (Moroto, Uganda) collected by R. Varne (1968)
7. Olivine melanephelinite (Moroto, Uganda) collected by R. Varne (1968)

The temperature dependence of the viscosity can be described by an Arrhenius relationship of the form $\eta = \eta_o \exp(E\eta/RT)$, where η_o is a constant and $E\eta$ is the activation energy for viscous flow. A plot of the logarithm of the viscosity against reciprocal temperature for the four glasses is shown in Figure 3. With increasing temperature, viscosities decrease from 10^{14} to 10^{10} poise. At all temperatures the basaltic andesite has a higher viscosity than the other compositions. Since all the fibres were prepared under identical conditions, it is unlikely that the differences are due to memory effects induced by the drawing process. The higher viscosity of basaltic andesite is attributed to compositional differences, specifically to its higher content of the network-forming oxides SiO_2 and Al_2O_3 (Scarfe 1973). Similarly, differences in the gradient of the lines, and therefore activation energies, in Figure 3 can be explained by compositional effects. Activation energies for viscous flow range from 155 to 80 kcal mole^{-1} (Table 2). The lowest value is for olivine melanephelinite which also has the lowest content of SiO_2 and Al_2O_3. These values are in broad agreement with activation energies in glass systems with fewer components (e.g. Dingwall & Moore 1954). The ratio of the activation energies at low and high temperature is approximately 2:1 (Table 2). The high activation energies at low temperatures suggest a mechanism of flow by bond rupture (Doremus 1973); however, the reconstructive changes necessary for flow are complex (e.g. Lacy 1968; Goldstein 1969). The lower activation energies at high temperatures have been attributed to the energy required to depolymerize melt structures (reviewed by Doremus 1973).

Most glass-forming substances have two reference temperatures: one is the melting point of the crystalline form and the other is the transition temperature from supercooled liquid to glass. The transition temperature corresponds approximately to the temperature where the glass has a viscosity of 10^{13} poise (Rawson 1967). The relationship between the glass transition temperature (T_g) and the liquidus temperature (T^l) for the four compositions is shown in Table 2. The values for T_g/T^l adhere to the 'two-thirds rule' ($T_g/T^l \simeq 2/3$) which, according to Sakka & Mackenzie (1971), seems to hold for many inorganic substances such as elements, sulfides and silicates. The theoretical basis for the two-thirds relationship has been discussed by Kauzmann (1948) and by Sakka & Mackenzie (1971) and will not be repeated here. The predictive qualities of the relationship are obvious.

Finally, low-temperature glass viscosities may be related to high-temperature melt viscosities (Scarfe 1973) by calculating the equation of best

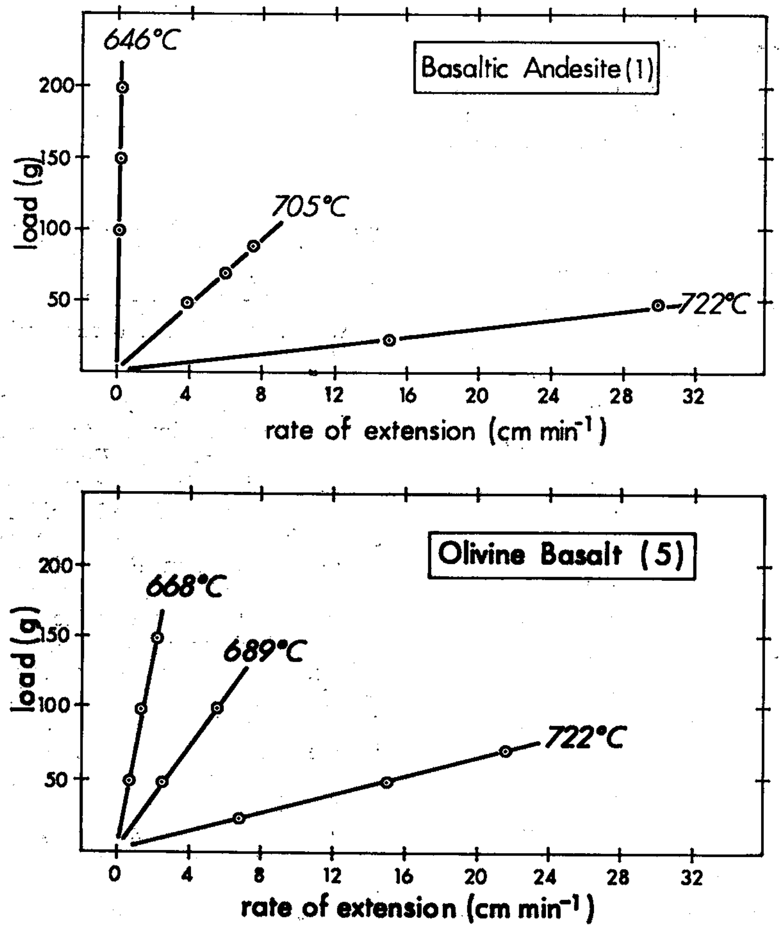

FIG. 2. The rate of fibre extension versus load at constant temperature for basaltic andesite (a) and alkaline olivine basalt (b).

TABLE 2

Some properties of glasses and melts

	T_ℓ (°K)	Tg(°K)	Tg/T_ℓ	E_L(kcal mole^{-1})	E_H(kcal mole^{-1})
1. Basaltic andesite	1433	955	0.67	141	68
2. Tholeiite	1433	917	0.64	155	56
5. Alkaline olivine basalt	1463	913	0.62	115	53
7. Olivine melanephelinite	1473	901	0.61	80	47

E_L and E_H are the activation energies for viscous flow at low (this paper) and high temperatures (Scarfe 1973). Estimated uncertainties are ± 5 kcal mole^{-1} for E_H and ± 15 kcal mole^{-1} for E_L.
Liquidus temperatures (T_ℓ) obtained by hot stage microscope determinations on powdered rock using an argon atmosphere. The liquidus temperatures have an uncertainty of ± 20°C.
Glass transition temperatures (Tg) where the viscosity is 10^{13} poise obtained from Fig. 3.

fit for the two data sets. In this way a complete viscosity–temperature description for basaltic liquids may be obtained for temperatures between the liquidus and the glass transition point.

The empirical equation proposed by Fulcher (1925) was used:

$$\log_{10} \eta = A + B/(T - T_o) \qquad (2)$$

where A, B and T_o are constants and T is the temperature in °C. The theoretical basis for this equation and the success with which it has been used in fitting viscosity–temperature data for a wide range of glass systems has been reviewed elsewhere (e.g. Macedo 1969; Doremus 1973). In order to fit this equation to the data it is rewritten

$$T \log_{10} \eta = T_o \log_{10} \eta + AT + (B - AT_o) \qquad (3)$$

The constants T_o, A and B are then found by a least-squares method (Macedo 1969). The constants for the four basaltic compositions are given in Table 3. These equations, which allow the calculation of basaltic melt viscosities over the entire range from the melt region to the glass transition point, place a useful limit on viscosities of supercooled melts of geological interest.

TABLE 3

Fulcher equation constants

Composition	A	B	T_o
1. Basaltic andesite	-3.670	6815	272.6
2. Tholeiite	-3.023	6086	264.1
5. Alkaline olivine basalt	-5.848	8675	192.6
7. Olivine melanephelinite	-2.180	3178	455.1

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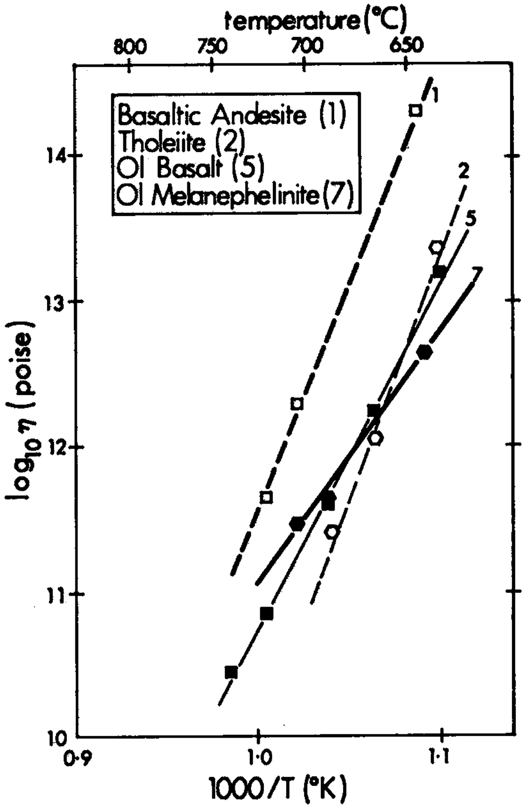

FIG. 3. Viscosity versus reciprocal temperature for basaltic glasses.

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