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DYNAMIC VISCOSITY OF SOME SILICATE MELTS TO 1688°C UNDER ATMOSPHERIC PRESSURE

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ABSTRACT

The dynamic viscosity of six anhydrous silicate melts, obtained by fusing rocks collected from the Department of Energy's Nevada Test Site, has been measured from 1120 to 1688°C under atmospheric pressure using a rotational cup viscometer of the Couette type. The melts exhibit Newtonian behavior in this temperature interval. Melt viscosities decrease in the order: tuff > alluvium > basalt. Activation energy for viscous flow in the interval 1120—1688°C ranges from 268 to 393 kJ.

INTRODUCTION

Modeling physicochemical processes which occur within the earth's crust can provide valuable insight regarding the development of geothermal resources; the history of potential underground nuclear waste repositories; and the generation, evolution and emplacement of economic ore deposits. A fundamental prerequisite is a knowledge of the transport and thermodynamic properties of silicate melts as a function of temperature, pressure and activity of water. As a first step in evaluating the effect of these parameters, we have investigated the viscosity of silicate melts obtained by fusing several different rocks collected from the Nevada Test Site. We report here the results of our study which was conducted to 1688°C under atmospheric pressure on tuff, rhyolite, basalt and alluvium melts.

SAMPLES AND EXPERIMENTAL METHOD

The names and identification numbers of these rocks are given in Table 1. In order to form homogeneous, bubble-free glasses, some compositions (e.g. tuffs Ue20u-330′ and Ue20u-357′) required repeated grindings and fusions to approximately 1600°C. All fusions were carried out in air. Of course, this procedure oxidizes the iron in our natural samples, thus producing high Fe^{3+}/Fe^{2+} ratios. But according to the results of Shaw [1], "no systematic differences" occur in tholeiitic basalt viscosities measured at liquidus temperatures in air and under low oxygen fugacities.

Chemical analyses of the rock glasses are provided in Table 2. All compo-

TABLE 1

List of rock glasses studied

No.	Name	Collection area in Nevada Test Side
Ue20u(330′)	Tuff	Schooner
Ue20u(357′)	Tuff	Schooner
Ue20L No. 2	Rhyolite	Cabriolet
Ue18q No. 2	Basalt	Danny Boy
Ue30a No. 4	Basalt	Buggy
U10ba	Alluvium	Sedan

sitions are high in silica. Specimens U10ba and Ue20u (330′) contain 78.50 and 75.58% SiO_2, respectively. Water concentrations in all these glasses are very low.

All experiments were carried out in a rotational cup viscometer of the Couette type. Complete descriptions of this apparatus and the calibration procedures employed in the experiments have previously been reported by Weed et al. [2,3]. Temperatures were measured by Pt_6Rh_{94}—$Pt_{30}Rh_{70}$ thermocouples. They were calibrated as a function of black body temperature which was determined by an automatic optical pyrometer. Details of the thermocouple calibration procedures have previously been discussed by Weed et al. [3]. On the basis of their work, we estimate the average error in the temperature scale to be ±2° C. Before determination of the viscosity of the silicate melt compositions, the viscometer was calibrated using a certified glass standard, No. 717, obtained from the National Bureau of Standards. Their reported viscosities and our results on the same glass are presented in Appendix III of Weed et al. [3]. Additional corrections due to the thermal expansion of the alumina rod holding the bob (see Figs. 1 and 2 in Weed et al. [2]) are reported in Appendix IV of Weed et al. [3].

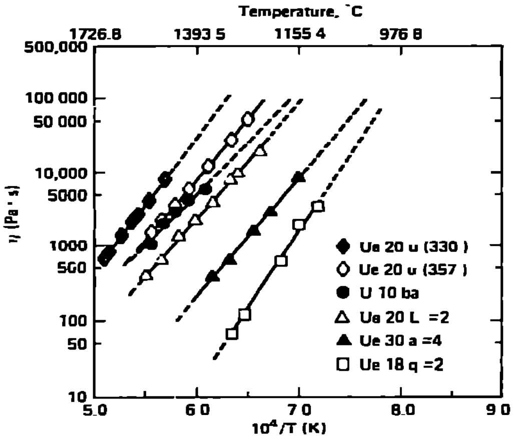

Fig. 1. Viscosity (η) of silicate melts as a function of temperature under atmospheric pressure. η(Pa · sec) = 0.1 η (poise)

TABLE 2

Chemical analyses [a] of rock glasses [b]

	Ue18q No. 2	Ue30a No. 4	Ue20u(330′)	Ue20u(357′)	Ue20L No. 2	U10ba
SiO_2	59.78	59.05	75.58	71.60	65.00	78.60
Al_2O_3	17.82	18.67	14.66	15.19	19.07	8.33
Total Fe (asFe_2O_3)	4.86	4.76	1.15	3.17	3.25	1.95
MgO	4.29	2.61	0.90	0.24	0.58	1.48
CaO	7.64	5.44	0.61	0.40	0.77	5.06
TiO_2	1.63	1.70	0.28	0.53	1.02	0.55
Na_2O	3.45	4.35	2.57	4.70	5.08	0.52
K_2O	1.47	2.66	3.77	3.54	4.03	1.60
H_2O^+	0.06	0.04	0.28	0.17	<0.02	0.40
H_2O^-	<0.21	0.02	0.08	0.08	<0.02	<0.02
Total	101.02	99.00	99.88	99.62	98.84	98.31

[a] Values are reported in wt. %.

[b] Analyst: Robert Lim, Chemistry Department, Lawrence Livermore Laboratory.

TABLE 3

Viscosities and activation energies for rock melts

Rock No.	T (°C)	$10^4/T$ (K)	η (Pa · sec)	E_a (kJ) *
Ue20u(330′)	1688	5.10	660.2	356 ± 13
	1671	5.14	741.5	
	1631	5.25	1386	
	1593	5.36	2246	
	1571	5.42	2484	
	1535	5.53	4044	
	1488	5.68	7888	
U10ba	1535	5.53	980.8	268 ± 21
	1495	5.66	1920	
	1456	5.78	2799	
	1416	5.92	4131	
	1375	6.07	5917	
Ue20u(357′)	1532	5.54	1473	318 ± 8
	1490	5.67	1968	
	1459	5.77	3471	
	1419	5.91	6285	
	1354	6.15	12 690	
	1306	6.33	27 778	
	1270	6.48	53 084	
Ue20L No. 2	1541	5.51	363.6	301 ± 8
	1497	5.65	599.3	
	1444	5.82	1319	
	1401	5.97	2289	
	1359	6.13	3830	
	1312	6.31	7409	
	1294	6.38	9342	
	1239	6.61	18 942	
Ue18q No. 2	1310	6.32	66.9	393 ± 21
	1267	6.49	123.2	
	1193	6.82	609.4	
	1158	6.99	1964	
	1117	7.20	3436	
Ue30a No. 4	1353	6.15	372.8	301 ± 4
	1310	6.32	605.2	
	1251	6.56	1482	
	1212	6.73	2737	
	1149	7.03	8632	

* E(J) = 4.184 E (cal).

EXPERIMENTAL RESULTS

The viscosities are recorded for each composition in Table 3 and Fig. 1. An equation of the type

$$\log \eta = B/T(\mathrm{K}) + A \quad (1)$$

fits the data as shown in Fig. 1. Table 4 contains numerical values of A and

B for each investigated melt composition, obtained by least squares analysis of eqn. (1). It is clear that the rheological properties of the basaltic melts differ from those of the tuffs, rhyolite and alluvium. The former are much less viscous than the highly siliceous melts.

Activation energies (E_a) for viscous flow have been calculated for each composition in the temperature interval 1120—1688°C, and are listed in Table 3. Mean values of the activation energy for the basalts and tuffs are 347 ± 65 and 325 ± 28 kJ, respectively.

DISCUSSION

Shaw [4] recently proposed an empirical method to calculate viscosities of natural, multicomponent silicate melts. Using the data provided in Table 2, silicate melt viscosities were calculated following the method of Shaw. The results are portrayed in Fig. 2. There is fair agreement between the calculated and observed viscosities for the tuff and rhyolite compositions

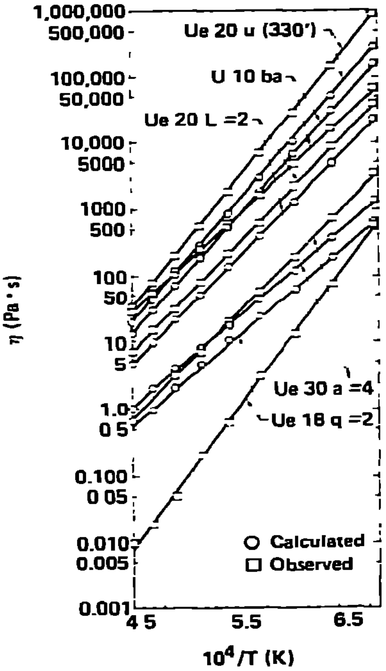

Fig. 2. Viscosity (η) of silicate melts as a function of temperature. Observed viscosities are taken from Fig. 1. Calculated viscosities are after Shaw [1]. See text for discussion.

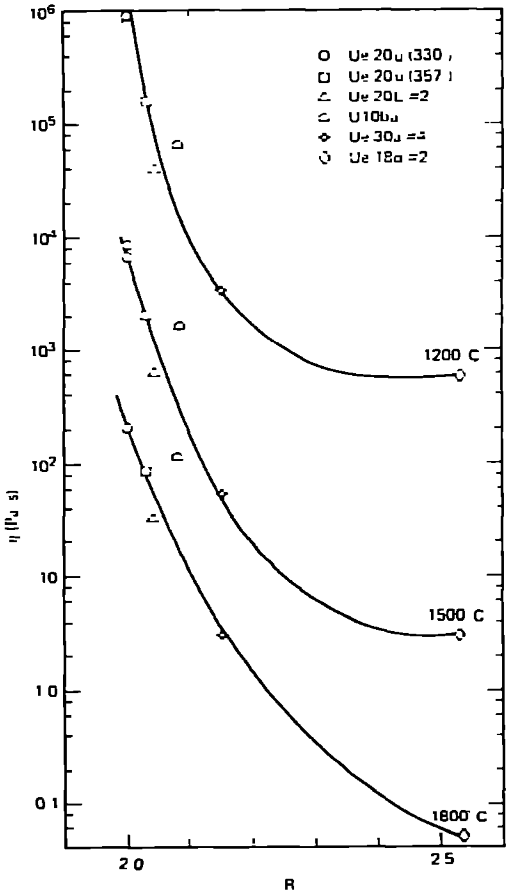

Fig. 3. Viscosity (η) of silicate melts as a function of *R* at 1200, 1500, and 1800°C. See text for discussion of *R*.

TABLE 4

Coefficients, A and B, in the viscosity equation
$\log \eta = B/T(K) + A$

Rock No.	A	B
Ue20u(330′)	−6.672	18 616
U10ba	−4.743	14 098
Ue20u(357′)	−6.081	16 637
Ue20L No. 2	−6.067	15 719
Ue18q No. 2	−11.16	20 511
Ue30a No. 4	−7.072	15 632

throughout the temperature interval investigated. However, agreement between the calculated and observed viscosity of the basaltic melt, Ue18q No. 2, is less than ideal at high temperatures.

Viscosity can be related to melt composition if we assume that these multicomponent silicate melts are composed of oxygen ions, network-forming anions and network-modifying cations. As a result of studies in simple binary and ternary systems, Lacy [5,6] proposed using a network-forming index R, where $R = \text{O}/(\text{Si} + \text{Al})$ and is the ratio of molecular % of non-bridging oxygens to network-forming cations. Transforming the glass analyses in Table 2 into molecular percentages, R was calculated for each of the investigated compositions. Results are illustrated in Fig. 3 which plots viscosity as a function of R at 1200, 1500 and 1800°C. It is clear that the viscosities of the investigated melts decrease with increasing values of R. Similar results were recently reported by Scarfe [7] over a much larger range of R than used here.

The data for U10ba lie off the general trends shown at the three temperatures in Fig. 3. We believe that the chemical analysis of U10ba glass may be at fault because of the low oxide total (see Table 2). If small amounts of P_2O_5, CO_2, MnO, etc., were present, additional oxygens would then be available. This would have the effect of decreasing R to a value more concordant with the trends shown in Fig. 3.

SUMMARY AND CONCLUSIONS

The dynamic viscosity of six anhydrous silicate melts has been measured from 1120 to 1688°C under atmospheric pressure. These were obtained by fusing various rocks collected from the Department of Energy's Test Site in Nevada. Within the investigated temperature interval, these silicate melts exhibit Newtonian behavior, and the viscosities decrease in the order: tuff > alluvium > basalt. Activation energies for viscous flow range from 268 to 393 kJ. Using Shaw's technique, silicate melt viscosities have been calculated and compared to experimentally-determined values. Agreement is poor between the calculated and observed basalt melt viscosity, Ue18q No. 2, at high temperature. However, there is fair agreement between the calculated and observed tuff and rhyolite melt viscosities.

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