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USE OF A PRESSLESS MULTIANVIL HIGH-PRESSURE SPLIT-SPHERE APPARATUS TO MEASURE THE SILICATE MELT VISCOSITY

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The possibility of using a pressless multianvil high-pressure split-sphere apparatus to measure the viscosity of silicate melts was studied experimentally using as an example diopside melt ($CaMgSi_2O_6$) *at a pressure of* 4.0 GPa *and a temperature of* 1800°C. *The viscosity was calculated by the Stokes formula. The measurement error of the melt viscosity was estimated by the falling ball method. Prospects for using devices of this type to estimate the viscosity of silicate melts at high pressures and temperatures are shown.*

Key words: *high pressure and temperature, viscosity, silicate melts.*

The study of the viscosity of basic and ultrabasic silicate melts at high pressures and temperatures, especially the viscosity of magmatic melts at high pressures and temperatures of the Earth's mantle is an important problem [1] related to the development of geodynamic models for the motion of matter in the Earth's interior.

The model object for studying the viscosity of ultrabasic melts at high pressures and temperatures is diopside melt ($CaMgSi_2O_6$), whose viscosity at high pressures was examined in [1–3]. Falling-ball experiments in the cell of a multianvil apparatus using synchrotron radiation are the most informative [3], but such equipment is unique and is not available in Russia.

In the Siberian Division of the Russian Academy of Sciences, work on developing multianvil high-pressure equipment began in the 1970s. The first devices (MEGA-1, MEGA-2, and MEGA-2m) had a number of advantages over high-pressure apparatuses based on presses but they were structurally imperfect. Searches for design and technological solutions have led to the development of improved pressless split-sphere apparatus (BARS) with a sphere diameter of 300 mm. In the 1990s, upgraded BARS apparatuses were developed and begun to be used in Russia and abroad, mainly for growth of large single crystals of diamond [4].

In the present work, we studied the possibility of using a pressless multianvil high-pressure split-sphere apparatus (BARS) to measure the viscosity of silicate melts (using diopside melts as an example).

Experimental Technique. The experiments were performed on a BARS apparatus using the falling platinum ball method [2].

The BARS pressless multianvil split-sphere apparatus consists of a spherical multianvil block placed in a fast-opening case. The high-pressure body has a spherical inner cavity and consists of a lower and an upper semi-bodies fastened with a flange type lock. Inside the semi-bodies there are self-contained hydrostatic-pressure chambers which are formed between the inner surface of the semi-bodies and the hemispheres of the pressure-sealing jacket. The semi-bodies have channels for oil injection into the high-pressure chambers. In each semi-body there are three measuring and one power current leads. The measuring current leads are connected to electric contacts which are located on the pressure-sealing jacket and are pressed to the surface of the anvils by the action of the pressure created in the high-pressure chamber. In the lower and upper semi-bodies there is a system of channels and nipples for cooling the multianvil block.

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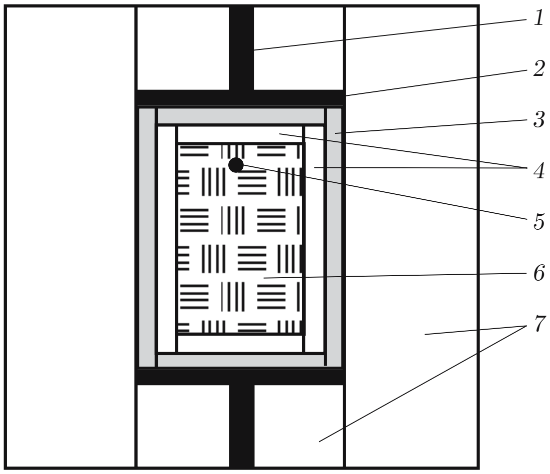

Fig. 1. Diagram of the high-pressure cell: 1) molybdenum rod; 2) molybdenum disk; 3) graphite heater; 4) sleeve and disks of magnesium oxide; 5) platinum ball; 6) diopside melt; 7) container made of a mixture of zirconium and calcium oxides.

The multianvil block of the apparatus of the 8-6 type consists of an inner and outer stages. The outer stage includes eight anvils with a triangular working face, and the inner stage includes six anvils of octahedral shape. A solid-phase reaction cell with a sample is placed in the inner working cavity having the shape of a cube.

Pressure is produced in the solid-phase reaction cell of the BARS apparatus by convergence of the anvils to the geometrical center of the apparatus. An increase in the external load is accompanied by growth in the quasihydrostatic pressure in the working cell. (The apparatus design and the methods for measuring pressure and temperature are described in detail in [4].)

The solid-state reaction cell of the multianvil apparatus has the shape of a 20 × 20 × 23 mm rectangular parallelepiped with truncated edges and vertices (Fig. 1). The cell was made by pressing high-melting oxides ZrO_2, CaO, and MgO. A heater consisting of a graphite cylinder with covers was placed in a 11 mm diameter cavity at the center of the cell. The walls of the graphite heater were 0.5 mm thick, and the molybdenum rods and disks served as current leads. The sample was insulated from the heater by a sleeve with a 1 mm thick wall, from top and bottom by 2 mm high disks of pressed magnesium oxide.

The sample was a diopside powder ($CaMgSi_2O_6$) pressed in the shape of a cylinder. A platinum ball was pressed into the upper face of the cylinder so that it was separated from the upper MgO disk by a diopside layer 0.10–0.15 mm thick. This was necessary to prevent the sticking of the ball, i.e., its introduction into the material of the upper disk.

For temperature measurements, the working cell was provided with a thermocouple which was connected to the sample through the lower current-lead cover. Temperatures up to $T = 1800$°C were measured with a Pr 30/6 platinum–rhodium thermocouple, and higher temperatures with a tungsten-rhenium thermocouple.

The initial material having diopside stoichiometry was prepared by the gravimetric method using specially pure grade oxides annealed at a temperature $T = (1200 \pm 5)$°C. After annealing for 9 h, the mixture was powdered in a mortar of VK-6 solid alloy. The material was melted in a platinum capsule at a temperature of (1550 ± 5)°C and cooled in water at room temperature. As a result, the sample had the shape of a cylinder of transparent glass similar in structure to diopside. The transparent silicate glass was crushed, ground, and pressed in a special cylindrical split-type die mould. As a result, the sample placed in the high-pressure cell was 8 mm in diameter and 7 mm high.

The melting point of diopside is 1395°C [5] at normal pressure, 1725°C at a pressure of 3.5 GPa, and 1927°C at 7.0 GPa [3]. The experiments were performed at a pressure $P = 4$ GPa and a temperature $T = 1800$°C and diopside was molten as was indicated by the motion of the platinum ball.

Platinum balls were made by melting platinum pieces in molten cesium chloride at a high pressure. Balls of 0.1–1.0 mm diameter were selected; their sizes were checked under an MBI-15 microscope with an accuracy of ±0.01 mm.

Results of Experiments with Fall of Platinum Balls
in Diopside Melt at $P = 4.0$ GPa and $T = 1800$°C

Experiment number	t, sec	r, mm	Position of ball in sample
5-36-07	0	0.65	At the upper end
5-37-07	60	0.65	At the lower end
5-38-07	30	0.70	At the lower end
5-39-07	55	0.10	At the lower end
5-40-07	45	0.10	At the lower end
5-41-07	50	0.10	At the lower end
5-42-07	40	0.10	At the lower end
5-43-07	30	0.10	In the upper end
5-44-07	25	0.10	In the upper end
5-45-07	35	0.10	Inside the sample ($h = 4.5$ mm)
5-01-08	60	1.00	At the lower end
5-20-08	90	1.00	At the lower end

The viscosity of diopside melts was measured at high pressure and temperature in the BARS multianvil apparatus using the following experimental procedure.

Prior to the experiment, the assembled cell was dried in a drying box at $T = 120$°C for 10 h. The cell was placed in the apparatus, the apparatus was tightly closed, and the water cooling of the internal power blocks was turned on. After that, oil was injected into the hydrosystem of the BARS apparatus with a NGR-2000 pump to create a pressure, which was recorded by a manometer using calibrated dependences of the working-cell pressure on the oil pressure in the system. The references for the cell pressure calibration were PbSe and Bi, which have pressure-dependents electrical resistances in phase transitions. Based on the known position of the graphite–diamond equilibrium line in the (P–T) coordinates [6], a correction for pressure change (increase) was made for the subsequent heating.

The increase in the working cell pressure was set in the interval of 0.1–0.2 GPa/min. The measured pressure gradient in the working cell at a temperature $T > 1000$°C did not exceed 0.1 GPa/mm and decreased with increasing pressure [4]. The pressure measurement error in the experiments was ±0.25 GPa.

Previously, using two thermocouples, it was established that, at $T = 1500$°C, the temperature gradient in the working zone inside the heater did not exceed 15°C/mm. The temperature measurement accuracy in the experiments was ±25°C.

Next, the sample was heated by passing an electric current from a power transformer through a graphite heater. The heating conditions were set by a RIF-101 temperature controller. The temperature was increased at a rate of 100°C/min to $T = 1400$°C and then at a rate of 300°C/min to the required temperature. After the attainment of the required experimental pressure and temperature, an exposure was made (the time between the moment when the prescribed temperature was reached and the moment of temperature release by the operator was recorded with a digital stop watch). In a series of repeated experiments, isothermal exposures were made with an interval of 5 sec.

After the necessary exposure, the sample was quenched directly at the high pressure by switching-off the electric current. The quenching time was 2–3 sec, which is due to the effective water cooling of the inner stage of the anvils. The pressure was released at a rate of 0.3–0.5 GPa/min.

After the experiments, the cylindrical silicate glass sample was sawn along the axis and polished. The position of the platinum ball relative to the upper end of the sample was determined using a MBS-10 microscope. Three main positions of the platinum ball in the diopside sample were marked: 1) the ball remained in the upper end; 2) the ball was inside the sample; 3) the ball moved to the lower end (see Table 1 and Fig. 2).

An important characteristic measured in the falling-ball experiments is the time of fall of a platinum ball in diopside melt from the moment of reaching prescribed pressure and temperature values to the moment of termination of the heating. For silicate melts with viscosity lower than 100 g/(cm · sec), the recorded time of fall of the ball is not sufficiently precise [2]. This is due primarily to the temperature gradient in the cell because melting of the diopside powder begins at the center of the sample and extends to its ends, leading to a delay in the beginning of fall of the ball.

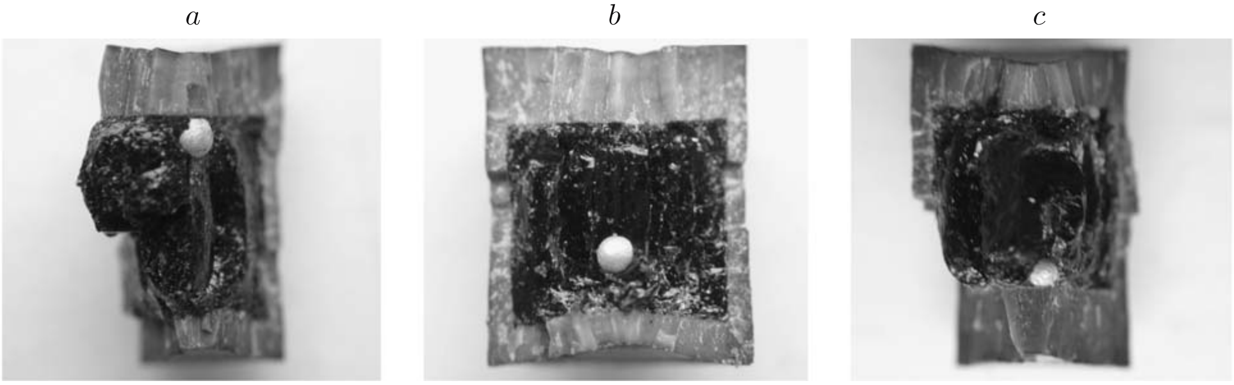

Fig. 2. Position of the platinum ball in the sample after experiments: ball at the upper end (a), in the volume of the sample (b), and at the lower end (c).

Since the moment when the platinum ball begins to fall is not determined in the experiments, a series of experiments was performed to determine the time of separation of the ball from the upper end of the sample. The average time of separation (beginning of movement) from the moment of reaching the prescribed temperature was (30 ± 2) sec. In the calculation of the time of movement of the ball, the indicated value was subtracted from the total time interval from the moment of reaching the prescribed pressure and temperature to the quenching time. The quenching time was set equal to (2.5 ± 0.5) sec.

Difficulties in determining the velocity of a platinum ball falling in molten diopside are due to the dynamics of the process, i.e., nonuniform movement of the ball, which was neglected in the experiments.

Discussion of Experimental Results. Knowledge of the viscosity of silicate melts at high pressure and temperature is of great importance for studying and modeling various geodynamic processes, such as the generation, rise, and differentiation of magmas, plume formation, etc.

The viscosity was calculation by the well-known Stokes formula

$$\eta = \frac{2r^2 g(\rho_2 - \rho_1)}{9V(1 + 3.3r/h)} K,$$

where η is the viscosity of the melt, $g = 980$ cm/sec^2 is the gravitational constant, r is the radius of the ball, h is the height of the diopside cylinder (the distance traveled by the ball during the experiment), $\rho_2 - \rho_1$ is the difference between the densities of the platinum ball and melt, V is the velocity of the ball falling in the melt, and K is the correction for the wall effect in the ampoule which is calculated by the formula

$$K = [1 - 2.104(r/r_a) + 2.09(r/r_a)^3 - 0.95(r/r_a)^5]$$

(r_a is the radius of the diopside cylinder).

In the method considered, the accuracy of determining melt viscosity at high pressures and temperatures depends on several factors: tool errors in measuring the radius of the platinum ball, the sizes of the diopside sample, the density of the melt, the errors of determining the velocity of the falling ball, etc.

In this present work, the accuracy in determining the viscosity of diopside melts on the BARS apparatus was affected by the following factors.

1. The radius of the platinum ball was determined under a microscope with an accuracy of ±0.01 mm; for balls of 0.2 mm radius, the relative error was ±5%.

2. The radius of the diopside cylinder was determined under a microscope with an accuracy of ±0.025 mm; for a sample of 4 mm radius, the relative error was ±0.6%.

3. The height of the diopside sample was determined under a microscope with an accuracy of ±0.025 mm; for a sample 7 mm high, the relative error was ±0.4%.

4. The density of platinum was set equal to 21.45 g/cm^3 [7].

5. The density of diopside melt was set equal to 2.8 g/cm^3 ignoring compressibility according to the data of [3]. The relative error was ±30%.

6. The velocity of the falling platinum ball made the greatest contribution to the error of the viscosity calculation (see Table 1).

Thus, the melt viscosity measured in the experiments can differ from the true viscosity by a factor of approximately 2.0–2.5, although the reproducibility of results in a series of repeated experiments is high enough (see Table 1). In other words, the experiments gave an approximate estimate of the melt viscosity coefficient. However, because the viscosity of magmatic melts can vary by several orders of magnitude, depending on their structure, temperature and pressure, even such approximate estimates are of great importance for studying the processes occurring in the Earth's mantle and crust.

At a pressure $P = 4.0$ GPa and temperature $T = 1800°\mathrm{C}$ ($r = 0.125$ mm and $V = 9.0 \cdot 10^{-1}$ mm/sec), the measured viscosity of diopside melt was 6.3 g/(cm · sec), which is in good agreement with the data obtained using synchrotron radiation in [3] [$\eta = 3.1$ g/(cm · sec), $T = 1730°\mathrm{C}$, $P = 3.5$ GPa, $r = 0.073$ mm, and $V = 8.5 \cdot 10^{-1}$ mm/sec].

Considering the technological features of the BARS high-pressure apparatus and the large reaction volume of the working cell, apparatuses of this type can be considered promising for use in the studies considered, especially considering the possibility of designing an experimental station based on the BARS apparatus and a synchrotron radiation source based on VEPP-4 [8].

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REFERENCES

1. É. S. Persikov and P. G. Bukhtiyarov, "Experimental studies of the effect of lithostatic and water pressure on the viscosity of silicate and magmatic melts. New structural chemical model for calculating and predicting their viscosity," in: *Experimental Mineralogy: Some Results on the Verge of Centuries* (collection scientific papers) [in Russian], Vol. 1, Nauka, Moscow (2004), pp. 103–122.
2. É. S. Persikov, "Viscosity modeling and magmatic melts at the PT parameters of the Earth's crust and upper mantle," *Geolog. Geofiz.*, **39**, No. 12, 1793–1804 (1998).
3. J. E. Reid, A. Suzuki, K. I. Funakoshi, et al., "The viscosity of $CaMgSi_2O_6$ liquid at pressures up to 13 GPa," *Phys. Earth Planetary Interiors*, **139**, Nos. 1/2, 45–54 (2003).
4. A. I. Chepurov, I. I. Fedorov, and V. M. Sonin, *Experimental Modeling of Diamond Formation* [in Russian], Izd. Sib. Otd. Ross. Akad. Nauk, Novosibirsk (1997).
5. F. V. Chukhrov (ed.), *Minerals: Handbook* [in Russian], Vol. 3, Issue 2, Nauka, Moscow (1981).
6. C. S. Kennedy and G. C. Kennedy, "The equilibrium boundary between graphite and diamond," *J. Geophys. Res.*, **8**, No. 14, 2467–2470 (1976).
7. E. L. Shvedkov, É. T. Denisenko, and I. I. Kovenskii, *Handbook Dictionary on Powder Metallurgy* [in Russian], Naukova Dumka, Kiev (1982).
8. A. I. Chepurov, V. M. Sonin, N. V. Surkov, et al., "Design of an experimental synchrotron-radiation station based on VÉPP-4 for studies at high pressures and temperatures on a BARS multianvil apparatus," in: *Digest Reports of the 17th Int. Synchrotron Radiat. Conf.* (Novosibirsk, June 15–20, 2008), Budker Inst. of Nuclear Phys, SB RAS, Novosibirsk (2008), p. 2–25.