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DEVELOPMENT OF HIGH TEMPERATURE VISCOSITY MEASUREMENT TECHNIQUE

MASTER

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DEVELOPMENT OF HIGH TEMPERATURE
VISCOSITY MEASUREMENT TECHNIQUES

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ABSTRACT

Viscosity measurements with magmatic rock at high temperatures and occasionally at elevated pressures are nontrivial problems. This report details the conception, development and testing of: 1) an electromagnetic viscometer and 2) an x-ray technique. Both methods employ a falling metallic sphere in the melt and provide real-time data.

Initial testing of the electromagnetic viscometer was performed at one bar and from 700-1300°C. Follow-on experiments with this viscometer were carried out at 1 bar and 0.466 Kbar (6.8 Kpsi) argon up to 1250°C at the Sandia Magma Energy Research Test Facility. The x-ray method was tested entirely at one bar and up to 1325°C. A simultaneous test series with both methods was performed at one bar.

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Various calibration glasses and basalts were used to evaluate the methods. Correlation of the two methods with each other and with reference data was within acceptable limits.

In addition to making real-time measurements of apparent viscosity, several interesting behavioral characteristics of the basalts were observed as the sphere progressed through the non-Newtonian magmatic rock.

These initial experiments definitely demonstrated the feasibility of using these techniques in future geoscience experiments. Bubble formation, crystal migration, and heat transfer studies employing various image-enhancement methods are possible secondary applications for the x-ray technique.

Key words: Viscosity, magmatic fluids, electromagnetic detection.

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Introduction

The Magma Energy Research Program at Sandia has a long range goal of energy extraction from magma chambers. Information on thermal properties, electrical properties, locating techniques, drilling techniques and material characteristics of magma must be developed before this long range goal can be attained.

The Magma Workshop recommendations¹ from the March, 1975 meeting state that viscosity measurements for various basalts over a range of temperatures (1000-1300°C) and pressures (to 4 Kbar) should be a priority item in the material characterization studies. Recommendations pertaining to Sandia's participation in those studies are outlined in SAND78-2227.² Viscosity measurements on reconstituted magmas (basalts in the presence of volatiles) will be needed to provide data for heat transfer studies.

Some measurements of the viscosity of magmatic rock have been made. Field experiments have been carried out in which rotating viscometers were emplaced in ponded lava pools³. Laboratory experiments have been conducted at high temperatures⁴ but at only slightly elevated pressures.² However, the greatest amount of information on the viscosity of molten rock at high pressures has been obtained in the laboratory by the "falling ball" technique.⁵

The "falling ball" technique is a common method of measuring the viscosity of a Newtonian fluid. The time for

a sphere to pass through a known distance can be experimentally determined and the viscosity calculated from the Stokes relationship;

$$\eta_f = \frac{2a^2g(\rho_s - \rho_f)t}{9L} \quad (1)$$

where:

η_f = viscosity of the fluid

a = radius of the falling sphere

g = gravity constant

ρ_s = density of the falling sphere

ρ_f = density of the fluid

L = known distance through which the sphere falls

t = time to fall distance L

This relationship assumes that forces acting on the falling sphere arise only from the fluid and are not perturbed by the wall of the container. Under normal conditions of atmospheric pressure and room temperature this is not a difficult experiment. However, when one wishes to measure the viscosity of magmatic rock at temperatures up to 1300°C and pressures up to 4 Kbar, the experiment becomes more difficult and many interesting and challenging problems arise.

Previous Experiments and Techniques

Previous experiments with the "falling ball" technique may be described as follows. Powdered rock is placed in a noble metal tube of approximately 0.6 cm diameter by 5.0 cm

length. A sphere which will not react with the molten rock during the experiment is placed on top of the powder. The tube is sealed and vertically positioned in the pressurizing/heating system. The system is first pressurized, then heated to the desired test temperature and held, allowing "sufficient time" for the sphere to fall through the molten rock. After the "sufficient time" period has elapsed, the system is cooled rapidly causing the sphere to be frozen into the solid rock. Either sectioning or x-raying the sample will disclose the position of the sphere and the distance through which the sphere had fallen can be calculated. Since it is likely that the sphere moved, if only slightly, during heating and cooling, some corrections to the distance traveled must be made. The magnitude of the correction will increase as the test temperature increases. Further, and perhaps of greatest concern, is the possibility that the velocity with which the sphere first penetrates the surface of the melt is different than the velocity it attains when falling through the bulk material. The viscosity calculated from the apparent velocity of the falling sphere would then not represent a property of the molten rock but rather some average property of the surface and volume effects. Repeated experiments at identical conditions in which the sphere is allowed to fall for increasingly longer times can separate surface and volume effects. These additional experiments are laborious and time consuming.

Clearly there was a need for an experimental method to accurately determine the velocity of a sphere during its travel through the molten rock at high pressures. The method should measure this velocity when the sphere is completely **within the liquid**, away from the surface. The electromagnetic viscometer described in this report meets these criteria.

It should be noted that magmatic basalt is not considered to be a Newtonian fluid, thus the direct applicability of the Stokes equation (1) is questionable,^{6,7} and, in fact, the "falling ball" method of determining viscosity may not be the best technique when using basaltic samples. However, previous investigators⁵ have used this approach to report what may be considered "apparent" viscosities. The purpose of this report is to describe new techniques which may be used to obtain viscosity data at high pressures and temperatures.

Electromagnetic Viscometer Description

The basic concept employed in the electromagnetic viscometer is a tuned circuit (transformer driven at its characteristic resonant frequency). The secondary windings of the transformer are wound on the sample crucible (see Figure 1). When the metallic sphere travels down the vertical centerline of the windings, the resonance of the coil is disturbed and an output signal is produced. Knowing the spacing between detection coils and the travel time, viscosities can then be calculated.

The secondary coil form/crucible is a closed bottom tube of Diamonite high purity alumina machined in the bisque state

ELECTROMAGNETIC VISCOMETER CELL CONFIGURATION

Fig. 1

and then fired to produce a very stable high temperature sample crucible. Two additional concentric sleeves of the same material serve as the coil forms for the primary windings of the transformer. All windings of the transformer are presently 0.5 mm Pt wire with 2 turns on each primary and 12 turns on each secondary. The windings are held in place by machined grooves and a light coating of high-temperature cement.

The metallic spheres in all tests were hardened steel ball bearings with a chrome plating.

For this application, the primary transformer winding is driven at 4-6 MHz and 3.0 volts from a signal generator (see Figure 2). The constant AC output of the secondary windings is rectified, yielding a constant low-level DC voltage. When a sphere passes one of the secondary coils, a change occurs in its DC voltage output and is recorded. The electromagnetic output from each detection coil was recorded on separate strip chart recorders operating at approximately the same speed. To minimize errors in time comparison of detection peaks, the recorders were electrically synchronized and the nearest sync marks were aligned when reducing data.

Electromagnetic Viscometer Development and Testing

Various combinations of primary and secondary coil wire sizes and relative spacing of the coils were tried to maximize the sensitivity of the apparatus. These combinations were tested by suspending a metallic sphere on a string in the electromagnetic field.

ELECTROMAGNETIC VISCOMETER SCHEMATIC

Fig. 2

Considerable time was spent in testing, modifying, and retesting the apparatus with a series of fluids and suspensions at room temperature. More testing was then done with N.M. basalt (La Bajada), lead-borate glass, and NBS 717 borosilicate glass. For the high temperature tests, a helically-wound Marshall tube furnace was used to melt the samples. This furnace has a 6.4 cm bore and is 45 cm in height.

After preliminary bench-testing, the electromagnetic viscometer was interfaced with the high-pressure, high-temperature test vessel. The uniform hot zone in this vessel is 10 cm in diameter and 10 cm in height. The molybdenum furnace is a resistance-heated serpentine design that is located below the uniform temperature zone.

The spheres were initially suspended above the melt by a 0.125 mm diameter Pt wire that was electrically melted when the test temperature/pressure was obtained, causing the sphere to fall into the melt.

NBS 717 glass was used as the sample material for several tests in argon at 0.466 Kbar. Due to interface problems with the electromagnetic viscometer/vessel installation, data was obtained from only one test. This point compared favorably with the previous bench-test data. Two tests were attempted with Hawaiian tholeiitic basalt under the same conditions, but again due to interface problems no usable data was obtained.

At first, the problems encountered in acquiring data could not be immediately attributed to interfacing problems.

Meaningful data could not be extracted from the experiment, if for example, the recorder traces of the secondary coil outputs showed no significant deviation from a constant value. Post mortems of some tests showed that the suspension wire never melted, and the ball never dropped. Other tests showed that the sphere had indeed passed through both coils, but its passage was not detected on the recorders.

In addition, some post mortems showed the sphere to be at the bottom of the crucible with a large void directly above the sphere. The observation that the fluid failed to close after passage of the ball was of particular interest to the researchers concerned with fluid behavior.

X-Ray Technique

In order to more fully understand the passage of a metal sphere through molten rock, a real-time x-ray technique was developed. Visual observation of the sphere falling through the melt was desired to observe void formation. Also, verification was sought for the assumption that the greatest deviation in secondary coil output voltage occurred when the falling sphere was in the center of the coil. Continued development of this technique revealed that it could be used as a viscosity measurement method and in studies of bubble and void formation, crystal migration, and heat transfer by using various image enhancement methods.

X-Ray Technique Description and Testing

A continuous 300 Kv x-ray source in the Sandia Non-destructive Testing Laboratory was used for this technique (see Figure 3). The x-ray source has sufficient power to penetrate the vertical tube furnace, crucible wall, magmatic rock, and metallic sphere. The resulting radiograph is detected on a fluorescent screen which is viewed by a TV camera. The TV image is then displayed on a standard monitor and also recorded on videotape. Audio reference timing from NBS station WWV was simultaneously recorded on the tape.

Data are obtained directly from the TV monitor when the videotape is replayed by timing the passage of a sphere moving down the crucible axis which was divided into 11 increments, each 1 ball diameter in height. This was done by overlaying a grid on the TV monitor screen. Using ball diameter increments eliminates the need for scaling the image back to original size. Increments 6, 7, and 8 were used for data calculations because the temperature and the sphere speed were the most uniform in these areas.

The x-ray method was tested entirely at one bar and up to 1325°C. A simultaneous test series with both the electromagnetic viscometer and x-ray methods was performed at one bar. Sample material was Hawaiian tholeiitic basalt powder, and the test spheres ranged from 1.57 mm to 10.32 mm in diameter. A longer tube furnace (76.2 cm in height) was used to minimize

X-ray Viscometer Schematic

Fig. 3

thermal gradients, and successive drops were made at time intervals sufficient to allow fluid closure behind the falling ball.

Observation of the passage of many spheres revealed that temperature uniformity, bubble formation, and fluid closure patterns in the wake of the falling sphere drastically effect the apparent viscosity data. Additional causes of obviously invalid viscosity determinations were identified. For example, if the sphere passed through the magmatic basalt although not near the centerline of the crucible as intended but instead near the crucible wall, the measured velocity was much lower and the apparent viscosity much higher. Another observation was that sufficient time must be allowed for temperature equilibration after adding powdered sample material to an existing melt.

Test Results for the Viscometers

For both the electromagnetic viscometer technique and the x-ray technique, the time required for a metal sphere to fall through a known distance was experimentally determined and the viscosity calculated from equation (1). Another requirement which must be met in order to use the Stokes relationship is that the crucible holding the fluid be greater in diameter than 10 sphere diameters. The close electromagnetic coupling required for the electromagnetic viscometer to function requires close physical spacing of the primary coils, secondary coils, and the sphere path. This results in the necessity for using a metal ball that is greater than one-tenth the crucible diameter, thus not an ideal situation. A Faxen

correction^{7,8} (K) must be applied to equation (1) to compensate for this situation:

$$n = \frac{n_f}{K} \quad (2)$$

where: n = apparent viscosity

The factor (K) relates drag forces caused by the crucible wall to diameter ratios of sphere/crucible. Typically this correction for a 10.32 mm diameter sphere in the electromagnetic viscometer is 3.8. If the measurement is being made in a short crucible where end effects, as the ball approaches the bottom, are significant, an additional subtractive 10% correction⁸ can be applied to equation (2).

The viscosities determined by the electromagnetic and the x-ray techniques and calculated by equation (1) were corrected by the method suggested by Faxen. These corrected values are shown in Figures 4 thru 6.

Figure 4 shows the data taken on NBS 717 borosilicate glass and a lead-borate glass formulated at Sandia. These tests were performed at one atmosphere. The data at the three test temperatures compares favorably with the smooth plot of NBS data.⁹ The NBS data represents the average values obtained by five laboratories using a total of four different techniques (rotating cylinder, fiber elongation, beam-bending, and parallel plate). The rectangular boxes indicate the spread of data with several samples taken by dropping balls of various sizes. Temperature nonuniformity in the melt

NBS 717 Borosilicate and Lead Borate Glass (1 bar)

Viscosity Data Summary - Various Basalts (1 bar)

Fig. 5

Viscosity Data - NBS 717 Glass

Fig. 6

probably has the greatest effect on these measurements and could be the reason for the offset from the NBS data.

The plot for the lead-borate is a composite obtained with the electromagnetic viscometer and data taken in the plastic region by J. A. Gilbert,¹⁰ using a loaded beam viscometer.¹¹ The electromagnetic viscometer data in the liquid region seems to correlate well with the loaded beam data. Again, the data spread is indicated by the brackets.

Figure 5 is a summary plot of various basalts at 1 bar shown with a reference NBS plot. Two different sphere sizes, 2 different techniques, and 2 different basalts are combined in the figure. This data plot can only be used as a general display of apparent viscosity for the Hawaiian tholeiitic basalt samples.

Figure 6 represents various experiments performed with NBS 717 glass. The electromagnetic viscometer data at 1 bar and 0.466 Kbar are consistent as expected. Again, the data spread with various sphere sizes and different viscometer cells is indicated by brackets.

CONCLUSIONS

Performance of Electromagnetic Viscometer

The development models of the electromagnetic viscometer performed quite well. The data obtained is consistent within itself and correlates reasonably well with reference data. The apparatus functions reliably at elevated temperatures (1300°C) and pressures (0.466 Kbar) and with proper signal conditioning yields reliable data. The method is a practical way to measure real-time viscosities of geologic samples in closed pressure vessels.

Evaluation of the X-ray Technique

This method served the purpose of confirming behavior that was observed, but not understood, in the electromagnetic viscometer. In its own right, the technique is a valid method for 1 bar viscosity studies. It could probably be used at some elevated pressures with a specially designed vessel. The technique yielded information about previously unobserved rheological behavior of magmatic basalts. This technique also showed that the maximum change in secondary coil voltage output occurred when the falling sphere was centered in the coil.

ADDITIONAL STUDIES

Improvement of Techniques

Additional studies with various geologic materials over a wide temperature range with comparison to previous experimenters' data³ is needed. For materials that are completely molten at the test temperatures, the modified Stokes equation is usable to obtain viscosity directly. However, most geologic materials are characterized by a melting or crystallization interval and contain crystalline phases suspended in the liquid. The corrections needed to the apparent viscosity obtained in these measurements are not known at this time, although some experiments with simulant materials have been made¹² to confirm that corrections are required.

The electromagnetic viscometer and possibly the x-ray scheme must be tested at increasingly higher pressures. The future plan² to inject volatiles (SO_2 , H_2S , Co, CO_2 , H_2) into

the magmatic basalt complicates the analysis for those tests. Modeling of these effects needs to be done to make viscometer data meaningful.

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REFERENCES

1. J. L. Colp, R. W. Decker, J. F. Hermance, D. L. Peck, and P. L. Ward, "Magma Workshop Assessments and Recommendations," SAND75-0306.
2. P. J. Modreski, "Experiment Plan for Characterization of the Properties of Molten Rock at Atmospheric and Elevated Pressures: Magma Energy Research Projects," SAND78-2227.
3. H. R. Shaw, T. L. Wright, D. L. Peck, and R. Okamura, "The Viscosity of Basaltic Magma: An Analysis of Field Measurements in Makaopuhi Lava Lake, Hawaii," American Journal of Science, Vol. 226, April 1968, pp. 225-264.
4. T. Murase and A. R. McBirney, "Properties of Some Common Igneous Rocks and Their Melts at High Temperatures," Geo. Soc. of Amer. Bul., Nov. 1973.
5. H. R. Shaw, "Obsidian-H₂O Viscosities at 1000 and 7000 Bars in the Temperature Range 700°C to 900°C," Journal of Geophysical Research, Vol. 68, No. 23, 12/1/63.
6. Private Communication, C. E. Hickox, Org. 5512, to R. P. Wemple, January 1979.
7. J. Happel and H. Brenner, "Low Reynolds Number Hydrodynamics," Noordhoff Int. Publishing, 1973.
8. Private Communication, C. E. Hickox, Org. 5512, to R. P. Wemple, dated July 9, 1979.
9. A. Napolitano and E. G. Hawkins, "Standard Reference Material 717, Borosilicate Glass," NBS Certificate dated 11/18/69.
10. Private Communication, J. A. Gilbert, Org. 1471-4, to R. P. Wemple, June 1979.
11. J. A. Gilbert, "A Beam-bending Method for Determining the Viscosity of Glass," SAND76-0219.
12. Private Communication, A. M. Kraynik, Org. 5813, to R. P. Wemple, December, 1979.

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