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# High Pressure-Temperature Viscosity Study of an Indian Natural Rock

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Viscosity measurement of the natural rocks is an important experimental tool for the knowledge of extrusive and intrusive igneous changes inside the earth. The viscosity is measured by the falling sphere technique. The chemical composition of the rock was analyzed by the EPMA.

**Key Words: Viscosity, High pressure-temperature, EPMA, Carbonatites, Piston-cylinder apparatus.**

## INTRODUCTION

Viscosity of a magma is an important property governing both extrusive and intrusive igneous processes, and has important influence on magmatic differentiation and crystal growth. There is a close relation amongst viscous flows, diffusion and convection in magmatic silicate melts[1]. Different groups like Kushiro[2] and Scarfe *et al.*[3] started the measurement of viscosity of iron rich liquids at high pressures and found the quench probe technique useful for high viscosity liquids. Rutter and co-workers[4] from the University of Western Ontario, Dobson and co-workers[5], Lablance and Secco[6], and others[7, 8] measured the viscosity of Fe-S liquid at different pressure by X-ray radiography technique (this technique is equally good for low and high viscosity liquids) and found that the X-ray measurement technique of terminal velocity for the falling ball is more accurate than the quench probe technique.

The viscous flow of a magma is very much dependent on the crystal structure of the silicate bearing phases present in the melt, *e.g.*, a komatiite is characterized primarily of minerals which are either nesosilicates (olivine with isolated $SiO_4$ tetrahedron) or inosilicate (pyroxine with chain structure). In case of the olivine structure there is no bridging oxygen whereas in case of pyroxine there are two bridging oxygens; thus the komatiitetic melt has the least viscosity amongst all magma types; the granite however is characterized by the presence of K-feldspar and quartz, which are tectosilicates (the $SiO_4$ tetrahedron have three-dimensional network structure). In the present communication, we report the chemical analysis and viscosity study of a carbonatite rock particularly collected from Ambadondar region.

## EXPERIMENTAL

The high pressure-temperature viscosity experiments were carried out on the natural carbonatite rock collected from Ambadongar, Gujarat (India). The place Ambadongar is located at 37 km south of the Chhota Udaipur (latitude 22°N, longitude 74°5′E in Baroda District. in all the collected rock samples we chose arbitrarily a sample (Amba-2). This rock was crushed in different steps (the grain size of the powder < 200 mesh). This fine powder was used for the viscosity measurement experiment.

The chemical analysis of the rock sample was done by electron probe micro-analyser (Jeol, Japan, JXA-8100 model). For this we used a slide (thickness > 30 micron). This slide was ground by corundum powder (of different particle sizes) and mirror polished by diamond and alumina paste (minimum size *ca.* 0.1 micron). A carbon coating of 20 micron thickness was done on the upper layer of the slide for conductivity.

The high pressure-temperature (experimental range: 8–25 kbar and temperature up to 1400°C) viscosity experiments were done on piston cylinder apparatus (Tungalloy, Japan). In this instrument, a core of WC (tungsten carbide) is fitted in a circular steel plate. The cell assembly (pyrophellite-graphite assembly) is put in this core and the pressure is generated on the sample by a piston (diameter = 12.5 mm and length = 45 mm) sliding in this, through a static pressure ram (manually). The pyrophellite-graphite assembly is shown in Fig. 1; the description of different components is as below:

1. a pyrophellite cell (L = 40 mm, OD = 12.45 mm and ID = 10 mm);
2. a pyex glass tube (L = 37.5 mm, OD = 10 mm and ID = 8 mm) placed inside cell-1;
3. a graphite heater (L = 37.5 mm, OD = 8 mm, and ID = 6 mm) placed inside cell-2 with a disc (thickness = 2.5 mm, diameter = 10 mm) at the bottom of cell-1;
4. a pyrex glass tube (L = 35.5 mm, OD = 6 mm and ID = 4 mm) placed inside the heater to avoid the possible oxidation.
5. In (4) a solid pyrex glass rod (OD = 4.0 mm and L = 12 mm) was kept in the lower side.

   Then put the *sample capsule* and a small amount of alumina powder which can fill the empty gap between the capsule and glass and also provided a cushion effect. In the upper side a hollow glass tube (L = 16 mm) was placed to pass the thermocouple up to the sample.

   To prepare the sample capsule, the fine powder of carbonatitic rock (weight *ca.* 10 mg) was placed in a platinum tube (OD = 3.6 mm, ID = 3.4 mm and length = 13 mm). One end of the platinum tube was flatly sealed by an arc welding method and the powder in the form of a cylinder was filled up to 70% of its volume. A small platinum ball was placed centrally in the tube and again the upper end was also sealed. The schematic diagram of the cross section of the sample capsule is shown in Fig. 1.
6. On the top of the cell a steel plug insulated by pyrophellite was used as the above electrode for the conduction of current.

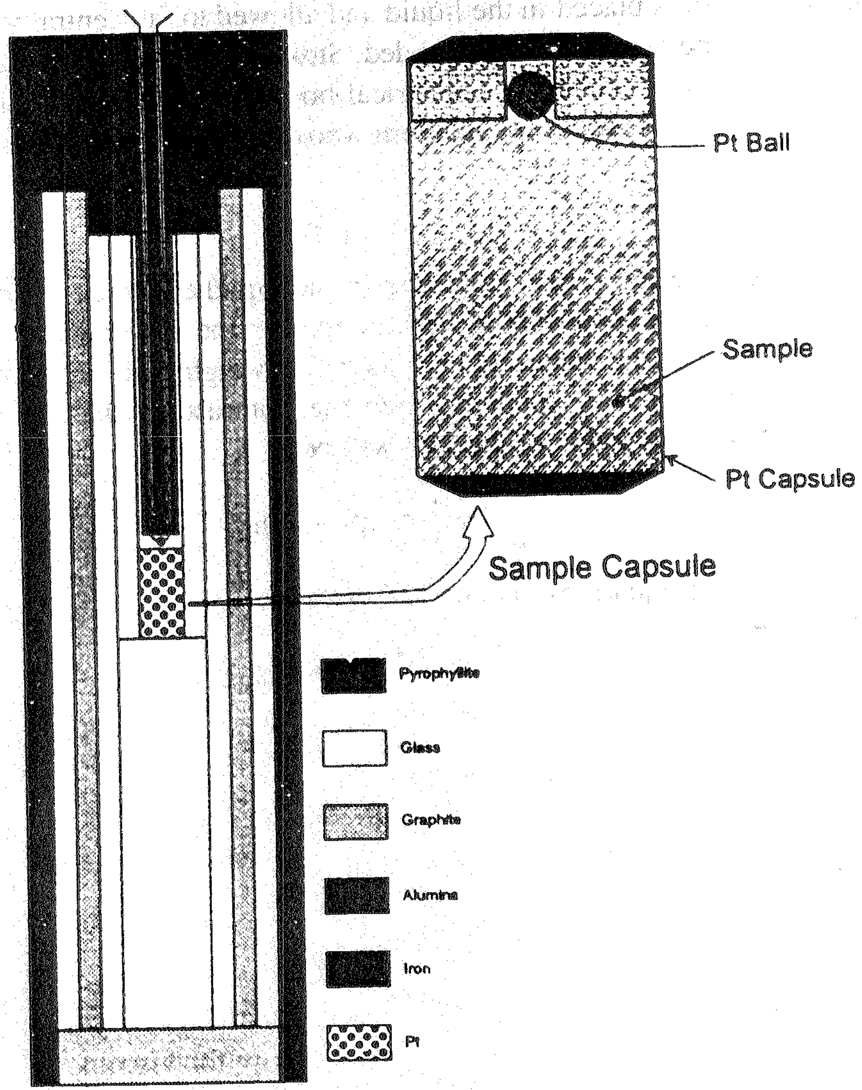

Fig. 1. The pyrophellite graphite assembly for the piston-cylinder experiment

A thermocouple (namely $Pt_{100}$-$Pt_{87}Rh_{13}$), covered by a two-hole alumina tube, was passed through the plug and pyrex tube up to the sample container. A lead foil (0.01 mm thickness) was wrapped around the cell assembly to minimize pressure loss due to friction. This wrapped cell assembly was placed in the core. Pressure was generated to end load the vessel and tungsten carbide piston statically. Temperature was maintained and monitored by an electronic temperature controller atached with the main unit. After attaining the desired pressure, heating was started up to the required temperature for the fixed time (depending on the experiment time) and quenched (cooling rate *ca.* 700°C/min). The quenched sample was fixed in epoxy and ground up to the clear visibility of the ball.

## Theory Used

The property of any liquid by virtue of which it opposes relative motion between its different layers is known as the viscosity of the liquid. For the determination of the viscosity of any liquid, Stoke's method is applicable, in which a tiny ball is placed in the liquid and allowed to fall centrally. The terminal velocity $v_s$ of the falling ball is recorded. Stoke showed that the retardation force $F$ due to the viscous drag for a spherical body (or ball with a radius $r$) moving with a terminal velocity $v_s$ in a medium whose coefficient of viscosity $\eta$ is given by

$$F = 6\pi v_s r\eta \tag{1}$$

When the sphere (or ball) is falling in the liquid the force acting on it is its own weight and the buoyancy of liquid opposing the movement of the sphere.

If the density of the spherical body is $\rho_1$, its weight will be $4/3\ \pi r^3\rho_1 g$ and if $\rho_2$ be the density of the medium, then the upthrust on the ball $= 4/3\ \pi r^3\rho_2 g$. Resultant downward force on the ball will be

$$F = \frac{4}{3}\pi r^3(\rho_1 - \rho_2)g \tag{2}$$

After equating equations (1) and (2), we get

$$6\pi v_s r\eta = \frac{4}{3}\pi r^3(\rho_1 - \rho_2)g$$

or

$$\eta = \frac{2}{9}\frac{r^2 g\,(\rho_1 - \rho_2)}{v_s} \tag{3}$$

Arrhenius proposed an empirical relation between the change in shear viscosity with temperature for many liquids, *i.e.*,

$$\eta = A_\eta e^{-E_\eta/RT} \tag{4}$$

where $A_\eta$ is the pre-exponential factor and $E_\eta$ is the activation energy per mole. In Eyring's theory of liquids, the activation energy for viscous flow is assumed to be proportional to the energy of sublimation and comparable to the energy required for a particle to jump into an adjacent hole. For the calculation of activation energy $E_\eta$, we plot log ($\eta$) value with 1/T (in Kelvin) scale. The slope of this curve at each point gives the value of $E_\eta = 2.303R\dfrac{\Delta \log \eta}{\Delta(1/T)}$. Substitution of this value in eqn. (4) gives the pre-exponential constant $A_\eta$.

For the estimation of effect of pressure on viscosity, a semi-empirical theory (Poirier, 1988) describing transport properties in liquid metals under pressure was proposed. According to this theory the effect of pressure on the viscosity can be quantified through the activation volume $\Delta V_\eta^*$ of the viscosity, defined as

$$\Delta V_\eta^* = RT(\partial \ln \eta/\partial P)_T \tag{5}$$

where R is the universal gas constant, T is the temperature in Kelvin and P is the pressure.

## RESULTS AND DISCUSSION

The viscosity of a natural carbonatitic rock was determined with the help of Stoke's law. The terminal velocity of the sphere ($v_s$), was determined by the displacement of the ball under the high pressure and temperature (*i.e.*, displacement-time curve). The displacement of the platinum ball under different P-T conditions (for different time spans) was measured by fixing the sample capsule in the epoxy and ground them up to clear visibility (shown in Fig. 2). In this figure (A) indicates the position of ball before the pressurization and (B) and (C) give the position after different intervals of time. The density ($\rho_1$) of the sample is determined and found to be 2.278 g/cc and $\rho_2$ for Pt-ball is 21.5 g/cc. The pressure dependence of viscosity from isothermal experiment (T = 1000°C and T = 1200°C) is shown in Fig. 3. Here, it is clear that at constant temperature, viscosity decreases with increasing pressure, presumably because of the participation of $Al_3^+$ preferentially in the six co-ordinated site, instead of four co-ordinated at higher temperature[8]. Similarly, at constant pressure (P = 10 kbar and 20 kbar) the viscosity decreases with increasing temperature shown in Fig. 4. The activation energy[9, 10] can be determined from the Arrhenius equation, which can be expressed as

$$\eta = A_\eta e^{-E_\eta/RT} \tag{6}$$

where $A_\eta$ is constant, $E_\eta$ is activation energy and R is the gas constant.

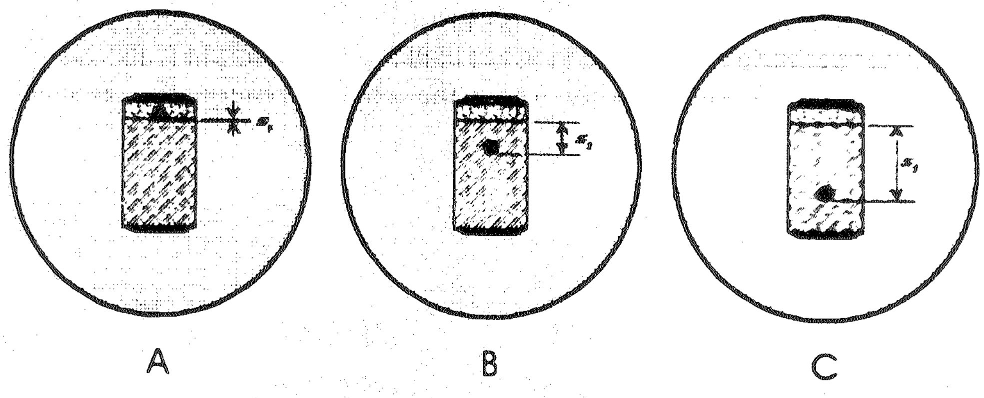

Fig. 2. Different positions of the ball in platinum capsules: (A) before the pressurization, (B) after 3600 sec, (C) after 7200 sec

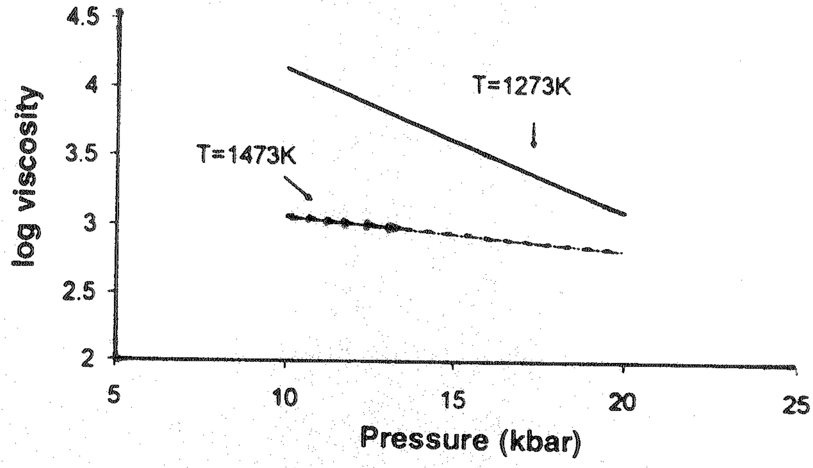

Fig. 3. Variation of viscosity with pressure at T = 1273 K and 1473 K

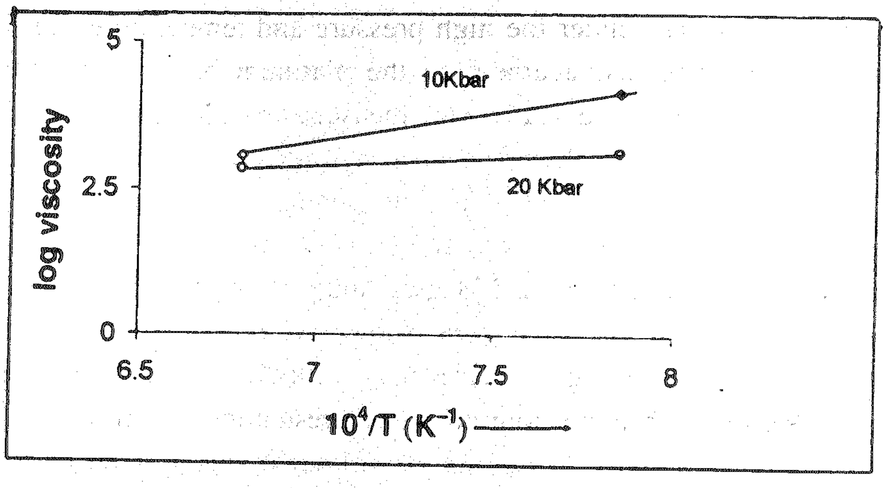

Fig. 4. Variation of viscosity with temperature at 10 and 20 kbar pressure

The slope of the curve $\frac{\Delta \log \eta}{\Delta 1/T}$ at each point in Fig. 4 multiplied by 2.303R will give the value of activation energy. The calculated values of $E_\eta$ at 45.93 kcal/mol and 12.2 kcal per mol respectively for 10 kbar and 20 kbar pressure.

In Fig.5 the variation of viscosity with temperature at different pressures of some samples (namely, rhyolite, obsidians with varied amount of water, carbonatites and tholeiite) indicates that viscosity decreases with increasing pressure. Similarly, at constant temperature an increase in water content in the sample is also an important factor for the decrease in viscosity. And in the activation energy this effect is clearly visible.

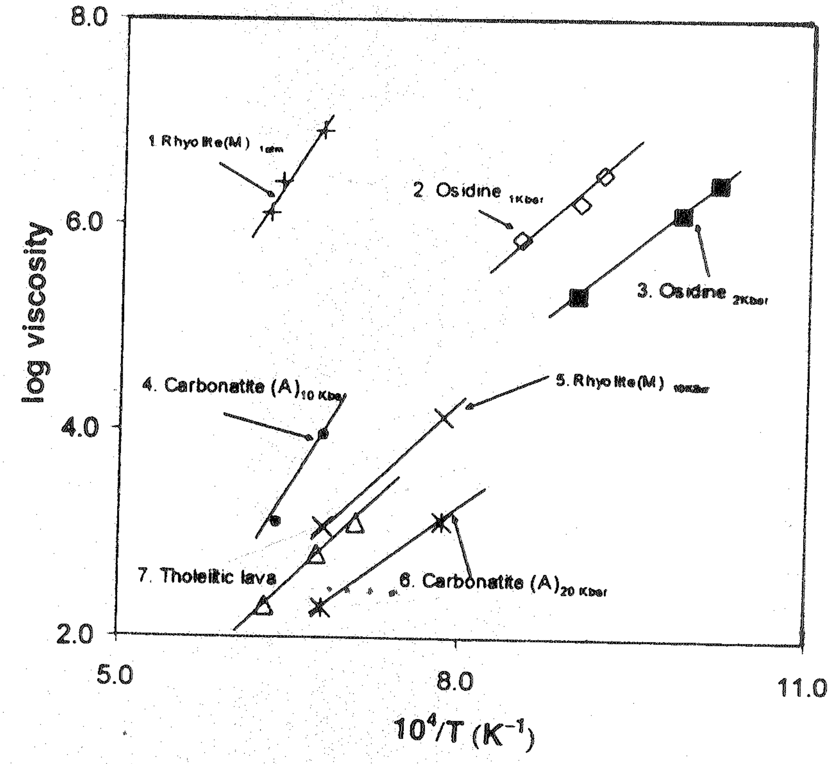

Fig. 5. Variation of viscosity with temperature: (i) rhyolitic lava$_{(Malani)}$ at P = 1 atm, (ii) Obsidian[11] with 4.3% $H_2O$ at P = 1 kbar, (iii) Obsidian[11] with 6.2% $H_2O$ at P = 2 kba,r (iv) Carbonatite$_{(Ambadongar)}$ P = 10 kbar (present study), (v) Rhyolitic lava$_{(Malani)}$ at P = 10 kbar (to be published), (vi) Carbonatite$_{(Ambadongar)}$ P = 20 kbar, (vii) Tholeitic lava[12]

The chemical analysis of the natural rock slide was carried out by EPMA and their chemical compositions are shown in Table-1. The microprobe analysis of the carbonates in the carbonatites is characterized by calcite and iron rich dolomite. In this case average presence of calcite is *ca.* 54%. On the basis of the chemistry of the carbonatites, it is characterized by the high concentration of $P_2O_5$, $Na_2O$, $K_2O$, $CO_2$ etc. The percentate of $CO_2$ is *ca.* 44% and $P_2O_5$ was found to be 0.15%. The petrographic analysis (given in Table-2) of the Ambadongar carbonatitic rock shows that it is a silicocarbonate rock in which the maximum volume in phenocrust is covered by calcite (*ca.* 60%).

TABLE-1

PETROGRAPHY OF AMBADUNGAR ROCK (AMBA-2)

| Sample No. | Locality | Texture | Phenocryst (Vol. %) | Microcryst (Vol. %) | Rock type |
|---|---|---|---|---|---|
| Amba-2 | Eastern fringe of the ring dyke | Prophyritic | Calcite (60)<br>K-Feldspar (5)<br>Magnetite (10)<br>Apatite (5)<br>Quartz (5) | Calcite (5)<br>Magnetite (10) | Silico-carbonatite |

TABLE-2

PRELIMINARY MICRO PROBE ANALYSIS (IN WEIGHT %) OF SAMPLE AMBA-2

| Analysis | Point-1 | Point-2 | Point-3 | Point-4 |
|---|---|---|---|---|
| $SiO_2$ | — | — | — | 0.09 |
| $TiO_2$ | 0.30 | — | — | — |
| $Al_2O_3$ | 0.08 | 0.44 | — | — |
| FeO | 0.49 | — | 0.21 | 0.26 |
| MgO | 0.96 | 0.11 | — | 0.21 |
| CaO | 52.82 | 54.32 | 53.96 | 54.21 |
| MnO | 0.40 | 0.43 | 0.19 | 0.49 |
| NiO | 0.52 | 0.68 | 0.56 | 0.67 |
| $P_2O_5$ | 0.11 | — | 0.16 | 0.16 |
| $CO_2$ | 44.32 | 44.02 | 44.92 | 43.89 |
| Total | 100.00 | 100.00 | 100.00 | 100.00 |

The high pressure-temperature viscosity of Indian natural rock melt (specially from Ambadongar, Gujarat) decreases with the increasing temperature and pressure. This leads us to conclude that the structure of the mineral constituents has a definite control over the viscosity of magma.

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