EDUARD SERGEEVICH PERSIKOV*

THE TEMPERATURE, PRESSURE AND COMPOSITION DEPENDENCES OF THE VISCOSITY OF MAGMATIC MELTS

(Figs. 11)

A bstract: Systematic viscosity measurements in magmatic melts combined with the theoretical interpretation of the results have made it possible to derive the general patterns of the temperature, pressure and composition dependences for such melts. Experiments were run on a radiation high-pressure viscosimeter, using the falling sphere method, the sphere was filled with ^{60}Co . The error limit was $10-15\,\%$ rel. A new method is proposed for calculation and prediction of the rheological properties (viscosity, η ; activation energy, E) of magmatic melts in the complete series from acidic to ultrabasic rocks. The features of the proposed method are simplicity, high accuracy (Δ E = \pm 1.5 %, Δ η \pm 30 %0) and the possibility to calculate the temperature, composition and pressure dependences in hydrated melts.

Резюме: На основе систематических экспериментальных исследований вязкости магматических расплавов и теоретического анализа полученных результатов установлены обобщенные закономерности их температурной, барической и концентрационной зависимостей. Эксперименты осуществлены с помощью радиационного вискозиметра высокого давления с падающим шариком, начиненным радиоактивным изотопом 60 СО с погрешностью 10-15 отн. $^{9}_{0}$. Предложен новый метод расчета и прогнозирования реологических свойств (вязкость — η , энергия активации — E) магматических расплавов в полном ряду кислые — ультраосновные. Простота, высокая точность расчетов (Δ E = \pm 1,5%, Δ η = \pm 30%), возможность расчета температурной, концентрационной и барической зависимостей флюид-содержащих расплавов являются характерными особенностями предложенного метода.

Viscosity is a most important physico-chemical and structure-sensitive property of magmatic liquids. Much of the work that has been done in the field in the last 50 years or so is due to Volarovich, Kani, Shaw, Khiratov, Lebedev, Carron, Murase, Kushiro and others.

This laboratory (Persikov, Epel'baum and Bukhtiyarov) has also been engaged in systematic viscosity measurements on magmatic and model melts over a wide composition range with respect to petrogenic and volatile components. Experiments were made using the falling sphere method, on a radiation high-pressure viscosimeter, with the sphere filled with 60 Co.

The apparatus, described in detail elsewhere (Persikov, 1976; 1984), is represented schematically in Fig. 1. It features a three-layer vessel (1) which is rated at the maximum pressure 15 kb and can move by 180° around the horizontal axis. The container is fitted with the cooling system, and two blind holes to collimate the γ — quantum flux from the radioactive filling of the sphere; the container houses a three-unit heater (2) with independent power control for each unit; the capsule (5) placed inside the heater is loaded with run sample and a platinum or platinum-rhodium sphere (3) filled with 60 Co.

^{*} Dr. E. S. Persikov, Institute of Experimental Mineralogy of the Academy of Sciences of the U.S.S.R., 142 432 Chernogolovka, Moscow district.

The capsule (5) is fitted with a special separator (6) to ensure a gradient-free argon pressure on the melt which is also exposed to the pressure of fluid of the varied, often complex composition. Also shown are the automatic temperature control and measurement unit (19) and the radiometric devices (15—18) to measure and record the intensity of the γ — flux from the sphere.

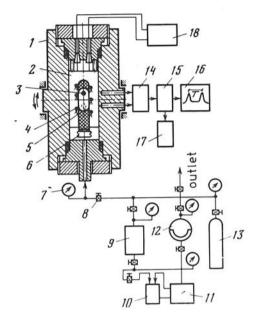


Fig. 1. Diagram of the gas pressure radiation viscosimeter-densitometer. Explanations: 1 - vessel: 2 - heater; 3 - sphere filled with 60Co; 4 - thermocouples; 5 - capsule with experimental melt; 6 - container with the gas-fluid separator; 7 - pressure gauges; 8 values; 9 - booster oil-gas; 10 - oil tank; 11 - high pressure oil pump; 12 - oil-gas compressor-separator; 13 - gas bottle; 14 spectrometric gamma scintillation counter: 15 - radiometric post: 16 electronic recorder; 17 - digital printer; 17 - temperature measurement and automatic control panel.

Without going into much detail of the method and equipment, it will be noted that the overall uncertainty in the viscosity measurements in magmatic liquids did not exceed 10 ± 15 %, which is twice the accuracy of the quenching method used by most authors. The temperature and pressure measurements were accurate to 2.5 ± 5 °C and ± 1 %, respectively.

Fig. 2 presents some of the obatined results. The more important implications from them are as follows: 1. The temperature dependence of the viscosity of melts under study varies exponentially and does not depend on either the petrogenic and volatile component composition or presure (lithostatic and fluid). The data in Fig. 2 can be described fairly well by the simple ${\tt Arrhenius-Frenckel-Eyring}$ equation:

$$\eta = \eta_0 \cdot \exp(E/RT) \tag{1}$$

where η_0 is the pre-exponential constant (P), E is the activation energy of viscous flow (cal/mole), T is the temperature in °K.

It has been shown for the first time for compositionally complex magmatic melts that the pre-exponential η_0 in eq. 1 is constant and approaches the theoretical value. Based on this, comparable values were obtained for the activation

energy of viscous flow, the critical rheologic and structural property, which is found from the slope of the isoconcentrates in Fig. 2, i. e. from simple equation:

$$E = 4.576 \text{ T (lg } \eta_{\text{T}} + 3.5)$$
 (2)

where η_T is the viscosity of the melt of T, determined from the experimental temperature dependence line in Fig. 2.

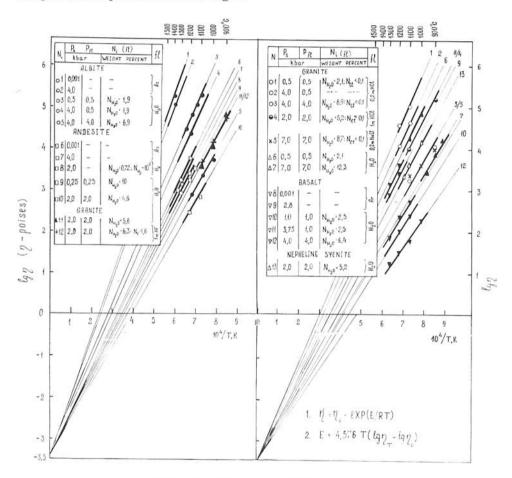


Fig. 2. Diagram of the reaction capsule. Explanations: 1 — platinum cap with holes; 2 — alundum plug; 3 — sphere with γ — radiation source; 4 — melt; 5 — platinum capsule; 6 — powdered glass (guenched melt).

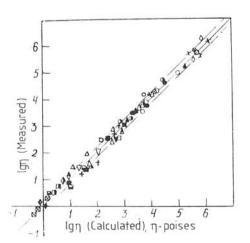
It is also important that the activation energy does not depend on temperature over the experimental temperature range. 2. Viscosities and activation energies of viscous flow of dry and hydrous melts decrease with increasing lithostatic pressure, $\rm H_2O$ content, $\rm P_{H_2O}$ and the melt basicity. Water, of a wide

spectrum of volatiles, has the greatest effect on the viscosity of magmatic melts, and the pseudobinary system alumosilicate melt — H_2O describes fairly well the rheologic behaviour of such melts at different hypsometrical levels in the Earth's crust.

Modern conceptions regard magmatic melts as polyanionic liquids, complex in composition and structure, which combine the properties of electrolytes, polymers and semiconductors. Natural melts show such a diversity in composition that it would be an unrealistic task to try to accomplish viscosity measurements for all of them. Furthermore, no comprehensive theory on viscosity of such complex system has as yet been offered. The existing empirical methods for calculation and prediction, including the well-known Bottinga—Weill model (1972), do not normally allow for the effects produced by volatiles and besides they give by far less accurate results than experiments.

The relationship between the structure of magmatic melts and the evolution in their chemistry as treated in terms of the Frenckel molecular-kinetic theory of liquids, has formed the theoretical basis for the new method of calculating and predicting magmatic melt viscosities. The experimental basis for the new method comprised the bulk of author's and other scientists' results on the viscosity of magmatic melts at atmospheric and high pressures, as well as abundant experimental evidence on viscosity in binary, ternary and more complex silicate and alumosilicate liquids.

The first step was to derive the generalized patterns; the fact that the pre-exponential in the Arrhenius – Frenckel – Eyring equation remains constant over a wide composition range has made it possible to analyse all the experimental evidence available on the viscosity of systems under study. The results are shown in the histogram (Fig. 3) which basically obeys the normal distribution rule. The weighted mean $\eta_o=10^{-3.5}$ poises is seen to be well within the theoretical range. Note that in the Frenckel and Eyring theories, the pre-exponential characterizes the viscosity of a liquid at $T \longrightarrow \infty$ and does not depend on the T, P and liquid composition under the boundary conditions specified by the authors for the near liquidus melts. It is only in the case that



comparison of the activation energies in compositionally varying liquids will have a strict physico-chemical meaning.

Fig. 3. The temperature dependence of viscosity of dry and hydrous melts (experimental results, see explanation in the text, P-MPa).

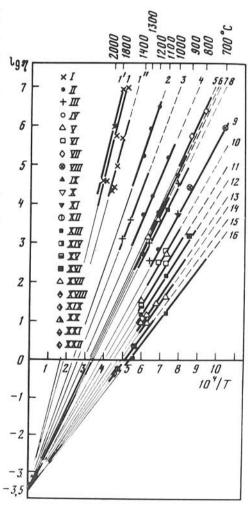
Figs. 4 and 5 present most of the generalized data on the temperature dependence of viscosity in magmatic and simpler silicate and alumosilicate melts obtained with the determined pre-exponential constant, and the uncertainty range \pm 30 %. It will be noted that it is seemingly wide error limit, in terms of the strict molecular theory of viscosity, in compositionally simple liquids, in often the highest possible precision achieved for such systems, especially at high pressures. What in more, results from different authors show sometimes even greater variance for compositionally close systems. This is illustrated in Fig. 4 the silica melt (isolines 1-1'-1'').

Owing to the regular arrangement of the isoconcentrates in Figs. 4 and 5, the experimentally determined patterns can be applied to the whole composition range of magmas, from granitoid to ultramafic, with sufficient reliability.

Fig. 6 presents the summary diagram for the pressure dependence of viscosity of dry and hydrous melts. based on the experimental data of the author and co-workers (1972-1984), Khitarov-Lebedev Kushiro et al. (1976; 1978 and oth.). It appears that none of the known theoretical equations for the pressure dependence which state that the liquid viscosity should increase with pressure, can allow for the reverse dependence of viscosity on pressure found in experiment with alumosilicate and magmatic melts. Below is given the semi-empirical equation which adequately describes the available experimental evidence for the whole range of natural composition within the stated uncertainty limits:

(3)
$$\eta = \eta_{\rm o}$$
 . exp [E/RT + α (Ps-P $_{
m H_2O}$)]

Fig. 4. Statistical distribution of the preexponential values ($\lg\eta_0$). (The histogram based on the bulk of experiments available on the temperature dependence of viscosity of rock melts and also on the more representative data on the viscosity in synthetic silicate and aluminosilicate melts).



Where α is the viscosity piezo-coefficient which, to a first approximation, is independent of temperature, taking on the negative values: $\alpha_1=5.02\cdot 10^{-6}$ MPa⁻¹ for dry melts; $\alpha_2=1.2\cdot 10^{-3}$ MPa⁻¹ for water — undersaturated melts; η_0 is the pre-exponential constatnt of eq. (1); E is the activation energy of viscous flow in dry melts at P $_{\rm H_2O}=$ O, or in hydrous (in the general case-fluid-bearing) melts at P $_{\rm H_2O}$, P $_{\rm S}$ is the total (lithostatic) pressure, MPa. It follows from eq. (3) that at P $_{\rm S}={\rm P}_{\rm H_2O}$, the pressure correction for viscosity is naught; in this case, the P $_{\rm H_2O}$ effect on viscosity shows up as a decrease in the

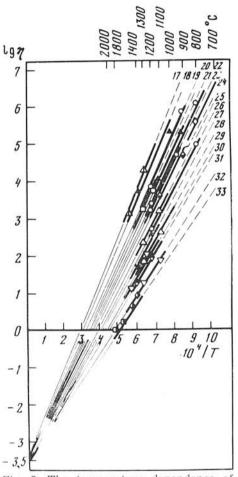


Fig. 5. The temperature dependence of viscosity of magmatic, aluminosilicate and silicate melts $(\lg \eta_{i0} = -3.5)$. Explanations: $I - SiO_2$ (1 - data Bacon, Avray from the handbook*, 1' - data Brucner, from the handbook*,

1" — Volarovich, 1937, Bockris

and oth., 1955, Solomin, 1940); II albite (2 - dry, 4, 6, 9, - hydrous NHO = 1.4; 4.5; 6.9 - Persikov, 1984); III - system $K_2O + SiO_2$ (3, 8, 9 - N_{K_2O} = = 3.9; 22.5; 41.6 - author's data and from $V = \text{system basalt} + H_2O$ (7, 10, 14, 26, 29, 32 - $N_{H_{2}O} = 0.7$; 1.5; 2.3; 6.4; 8.1;); VI - system andesite + H2O (10, 20 - $N_{H_2O} = 0$; 4.6); VII – system obsidian $+ H_2O$ (Shaw, 1963; 5, 24 - N_{H_2O} = = 4.3; 6.2); VIII - system pegmatite + $+ H_2O$ (9 - $N_{H_2O} = 8.8$; Burnham, 1963); IX — system granite + H₂O + HCl (21 - N $_{\mathrm{H}_{2}\mathrm{O}}$ = 5.2, N_{Cl} = 0.1); X — basalt (30 - Murase et al., 1973); XI - ba- $N_{H_2O} = 5.2$); XIII - system $Na_2 + SiO_2$ (8, 12, $16 - N_{\text{Na} \cdot \text{O}} = 15.4$; 40; $51 - B \circ c$ kris et al., 1955 and from the handbook*); XIV - system MgO + SiO₂ (33 -N_{MgO} = 41.7, from the handbook*); XV system $\text{Li}_2\text{O} + \text{SiO}_2$ (11 - $\text{N}_{\text{Li}_2\text{O}} = 17$ from the handbook*); XVI - system CaO $+ SiO_2$ (15 - $N_{CaO} = 54.3$ - from the handbook*); XVII — system $SiO_2 + Al_2O_3$ +CaO (17 - Sheludjakov, 1980); XVIII — system 0.471 SiO₂.0.055 Al₂O₃ 0.414 CaO 0.06 Na₂O (31 - from the handbook*); XIX - lunar basalt (32 - Murase-McBirney, 1970); XX - systems granite + H_2O , basalt + H_2O (18 granite $+ H_2O$, $N_{H_2O} = 5.7$, 13 - basalt $+ H_2O$, $N_{H,O} = 3.5$ – Lebedev-Hitarov, 1979); XXI — system granite + + $H_2O + HF (25 - N_{H_2O} = 6.27, N_F =$ = 1.6); XXII - dunite (16 - I v a n o v-Shtengelmeer, 1982). * Mazurin et al., 1973, 1977.

activation energy E owing to the dissolution of the equilibrium $\mathrm{H}_2\mathrm{O}$ concentrations in the melt.

Although the mechanism of the observed phenomenon is not yet fully understood, it is clearly related to structural changes because, as the experiments show the activation energy of the constant-composition melt decreases with increasing pressure (Fig. 2). The more likely explanation is that Al shifts from four to six-fold oxygen coordination in the melt at higher pressures. However, this possibility suggested by Waff (1975) is now rejected by Mysen and oth. (1980).

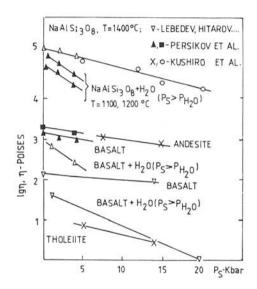


Fig. 6. A generalized diagram of the pressure dependence of viscosity of the magmatic melts.

Fig. 7 is a plot of the amount of aluminium transformed to 6-fold coordination in the albite and jadeite melt with pressure. The diagram is based on the experimental viscosity measurements and estimates obtained by the proposed method. In doing so, we proceeded from the obvious assumption that all Al available in the melt occurs in 6-fold coordination in the vicinity of the monovarium equilibrium jadeite-jadeite melt at, for instance, Ps = 35 kb, T = 1500 °C. In the experimental pressure range to 2400 MPa the octahedral aluminium content in the jadeite and albite melts is only $8 \pm 10\%$, the melt retains its albite (framework) structure and the ratio of the aluminium to silicon oxygen tetraherda changes by as little as 3 %. Clearly, such minor structural changes will not be reliably recorded in Raman spectra of albite and jadeite glasses quenched at high pressures, on which Mysen and oth., had based his conclusions. In addition, the diagram shows that the essentially pyroxene (metasilicate) structure in the jadeite melt (A $^{VI} \ge 50 \pm 100^{\circ}$) will form in an abrupt over a very narrow pressure range (33-35 kb), T = 1500 °C, and thus the possibility for this structure to survive quenching reduces considerably.

Of all the physico-chemical properties of a liquid, the viscosity shows the most complex dependence on melt composition. Although the measurements of

Volarovich (1937), Murase (1962), Bottinga — Weill (1972), Carron (1969), Shaw (1973), Urbain (1979) Shelubyakov (1980) have been valuable contributions toward an understading of the dependence, no reasonably accurate method applicable to the whole composition range of natural magmas has as yet been proposed.

In effect, the problem reduces to the following two questions:

1. It is necessary to devise some reasonably sensitive criterion to describe the composition and structure of the melt. Through the effort of many scientists, this task has been fulfilled. The criterion is defined as a structural-chemical parameter of the melt which reflects to a degree the equilibrium of the quasi-chemical forms of oxygen in the melt.

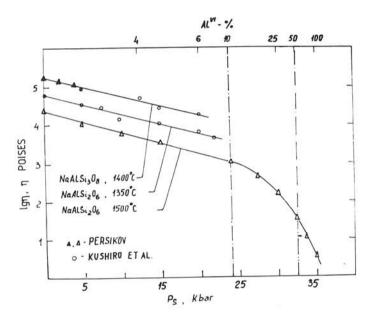


Fig. 7. The interrelationship between viscosity, pressure and coordination transition $Al^{|V|} \rightarrow Al^{|V|}$ in the albite and jadeite melts.

However, no solution has as yet been found to the second problem — in which way to correlate viscosities? Most previous correlation schemes involved isothermal composition which is, however, only valid for very limited composition ranges, — and that was the general practice. Since the liquidus temperatures of magmatic melts show a wide span, isothermal composition may often fall to have a physico-chemical meaning at all as is the case for granite and dunite. Evidently, the more complete information can be obtained from comparing the activation energies of viscous flow as a temperature — independent rheologic property of melts. This, however, could not be achieved in the past. The concentration dependence of the activation energy of viscous flow for the complete range of composition governing acid-ultramafic magmatic melt is given in Fig.

8. The melt composition is expressed through the criterion K which is called the degree of depolymerisation or "basicity coefficient". This parameter shows a good fit to "the percentage of broken bonds" of Saussier—Carron (1969) and to the NBO/Si or NBO/T of Bockris et al. (1970).

The diagram in Fig. 8 shows that the activation energy decreases steadily in the series acid-ultramafic melts as their basicity increases. The inflections in the E=f(K) curve divide it into four composition ranges that are interpreted in terms of the silicate melt structure theory (Bockris, 1965; Esin, 1965 and oth.). At K=17, which corresponds to the andesite average composition (after Le Maitre, 1976), the framework structure breaks down; at K=100 and 200, i.e. at the olivine tholeite and pyroxenite average compositions, diand metasilicate structures, respectively become completely formed in the melt.

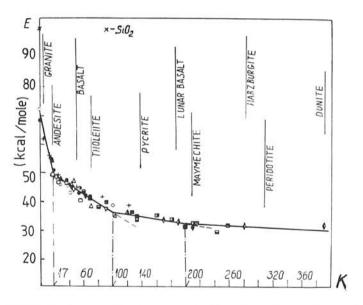


Fig. 8. A generalized diagram for the concentration dependence of the viscous flow activation energy in magmatic (aluminosilicate) and silicate melts (symbols are the same as in Figs. 4, 5).

The results clearly indicate that the difference between aluminosilicate and silicate melts shows up only within the first compositional range (K < 17). It appears that the structure of the aluminosilicate melts in this range can be envisaged as a continuous three-dimensional framework of molten silica in which Al replaces some part of Si in the tetrahedra [Si, O]. Owing to considerable energy difference between the Al-O and Si-O bonds (80 and \sim 109 kcal mole, respectively, Z h a r i k o v, 1969 and oth.), the quantitative ratio of the activation energies of viscous flow transition from silicate to aluminosilicate melts will be defined by the molar ratio $\varepsilon = \text{Al}^{1\text{V}}/(\text{Si}^{1\text{V}} + \text{Al}^{1\text{V}})$.

The breakdown of the framework structure (K > 17), i.e. in the other compositional ranges makes the structures of silicate and aluminosilicate melts indistinguishable in energy, because at K, being equal, their activation energies are also equal.

Linear approximation of the E = f(K) for each of the composition range has led to the simple equations for K:

The correlation coefficients for the obtained relations are: 0.83; 0.92; 0.97; 0.94, respectively. Simultaneous solution of these equations with eq. (3) gave the generalized equation for the compositional, temperature and pressure dependences of viscosities of magmatic melts

$$\lg_{\eta_{1,2,3,4}} = E_{1,2,3,4}/4.576T - 3.5 + \alpha (P_S - P_{H_2O})$$
 (5)

Consequently, the procedure for calculating the rheological properties of melts, using the proposed model, is very simple. The operations are:

1. The basicity coefficient (K) is computed from the chemical analysis (C u rron, 1969; Persikov, 1984). The effect of volatiles is accounted for in the following way: water (the dissociated form) is calculated from the equation:

$$H_2O_{(g)} + O^{\circ}_{(m)} \rightleftharpoons 2(OH)_{(m)}$$
 (O° is the bridging oxygen)

carbon dioxide:

$$CO_{2(g)} + O^{2-}_{(m)} \approx CO^{2-}_{3(m)}$$
 (O²⁻ is the free oxygen in the melt)

fluorine —
$$HF_{(g)} + O^{2-}_{(m)} = OH^{-}_{(m)} + F^{-}_{(m)}$$
 where (g) — gas, (m) — melt

- 2. Depending on the K, the value of E is calculated by one of eq. (4).
- 3. Eq. (5) is used to calculate the viscosity of near-liquidus melts for different T, P_s, P_{H_2O} .

The direct test of the method was made in an experimental viscosity study on dry and hydrous andesite and nepheline-syenite melts, for which there were predicted estimates. The calculated and measured values agreed to within 15— $-30\,^{0}/_{0}$. Fig. 9 compares most of the published viscosity measurements with calculated values for magmatic, silicate and alumosilicate melts. The specified uncertainity range is delineated by dashed lines. A relatively small number of values which fall outside the uncertainity region, are easy to account for and, except where the measurements are not self-consistent, arise from discrepancies for the oxygen potential in runs with iron-bearing melts.

The proposed method can be widely used in geochemistry and petrology of natural magmas. The approach has already proved useful in solving many problems bearing on structural positions of ions in magmatic melts, solubility mechanisms, the regimes of oxygen, water and other volatiles. Using this method, the author has confirmed the water dissolution mechanism he proposed back in 1974 for the acid — basic melts series: predominantly chemical dissolution

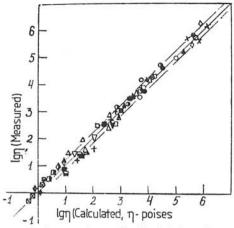


Fig. 9. Comparison of calculated and experimental viscosities of aluminosilicate, silicate and magmatic melts (symbols are the same as in Figs. 4, 5).

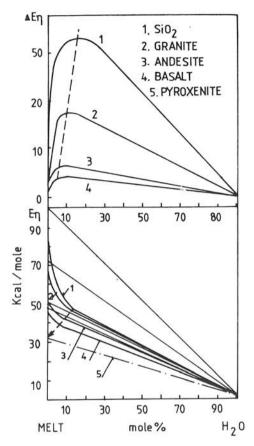


Fig. 10. A generalized diagram for the E - composition of the pseudobinary systems aluminosilicate (silicate) - H $_2$ O. Explanations: $1 - \text{SiO}_2$ - H_2 O; 2 - granite- H_2 O; 3 - andesite- H_2 O; 4 - basalt- H_2 O; 5 - pyroxenite- H_2 O.

(in the form of OH^- -hydroxyl) at the early stages is replaced by essentially physical dissolution (in the form of the H_2O molecule) at later stages.

The inflection points in the given relationships in Fig. 10 which, in the regular solution theory, record changes in the H_2O dissolution pattern, show a systematic shift toward decreasing H_2O contents with increasing basicity of the anhydrous melt. The absolute values of Δ E at these points decrease nearly one order of magnitude in the series silica-basalt. It is also seen that the amount of the chemically bound H_2O in pyroxenite melts tends to zero (shown by the arrow).

In keeping with the fundamental principles of acid-base interaction and chemical bond maximum polarity, these results clearly point that chemically dissolving water is a base relative to the melts within the specified composition range. This conclusion is fully supported by the large set of experimental results. Here are only some of them:

1. The viscosity and electrical conductivity patterns in hydrous melts; 2. Anincrease in $\rm H_2O$ solubility with increasing acidity owing to the dissolved fluorine: author's data (Persikov, 1984) for granite melts, and Kovalenko's

results (Kovalenko, 1979) for ongonite melts; 3. An increase in CO₂ solubility in albite melts in the presence of H₂O (Mysen et al., 1976; Kadik et al., 1976); 4. The results from the IR and Raman studies of hydrous glasses in the series granite-picrite (Orlova, 1969; Persikov, 1974; Mysen et al., 1980; Stolper, 1982; Epel'baum et al., 1984).

Another application to which the method may be put is calculation and prediction of other magmatic system properties related to viscosity: diffusion mobility of petrogenic and volatile components, electrical conductivity, crystallizability are some of the more important properties.

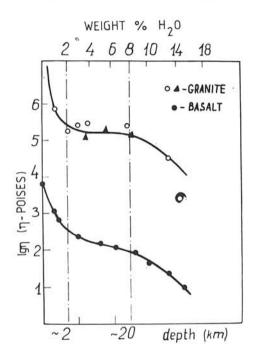


Fig. 11. Viscosity of the dry and hydrous granitoid and basalt melts at the liquidus thermodynamic parameters (T, P_{H_2O} ,

 ${
m ^{N}_{H_{2}O}}$). Explanations: in the text.

The method has worked well for calculating viscosities in dry and fluid-containing melts at the liquidus conditions not amenable to experiment. Some of the results for dry and hydrous granitoid and basalt melts are given in Fig. 11. The thermodynamic rock melting parameters are from Tuttle — Bowen, 1958; Kadik and oth., 1971; Burnham, 1967; Perchuk, 1973.

It has been found for a wide depth range in the crust (2 to 20 km) where according to geological evidence most gramitoid massifs are being formed that the viscosity of near-liquidus granitic and basalt magmas does not actually depend on the H_2O content in the wide range of concentration (2 to 8 mas. $\frac{0}{0}$).

Totally different viscosity patterns determined in near-liquidus granitoid and basalt melts at the $\rm H_2O$ contents < 2 wt. $^0/_0$, are believed to be the cause of the opposing relative occurences of effusive and igneous (plutonic) varieties of acid and basic rocks. In fact, completely degassed near-liquidus granitoid melts with the viscosity 10^7-10^9 poise will be unable to outpour in lava

flows and only occasionally erupt as extrusions or explosive ejections. Therefore, the bulk of melts will not reach the surface and their formation and crystallization will take place in the plutonic (depth) facies.

Unlike granitoid melts, completely degassed, near-liquidus basalt melts, retain their high mobility, as follows from experimental results and field observations in Hawaii (S h a w et al., 1968) and Kamchatka (A n d r e e v, S l e s i n et al.). This seems to account well for abundant basalt relative to gabbro occurrences in the Earth's crust.

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