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## TO THE GRANITISATION AS MAGMATIC REPLACEMENT – EXPERIMENTAL EVIDENCE

(8 Figs., 2 Tabs.)



A bstract: Experimental data on the granitisation problem are summaried. On the basis of data on surface tension, viscosity, and diffusion coefficients, the sizes of the fluid bubbles intruding into magma is defined. Perfectly mobile state of water and alkalies during magmatism is caused by infiltration fluid flows which maintain chemical potentials of these components at a constant level. The effect of the fluid composition on the melt composition in the systems Q-Or, Ab-Or, Q-Or-Bi, Q-Or-Ab is shown. These investigations indicate that: melts from Q-Fsp substrates are compositionally close to granitoids; dark minerals form only a small proportion of the model; the fluid composition has an effect on the composition of the primary melt and its temperature. Peculiarities of melting in the fluid-silicate system are considered and data on modelling of the "Granitisation" process are given. Magmatic replacement is considered as incomplete melting in the open system.

Key words: granitisation, fluid composition, fluid-silicate system, magmatic replacement.

Up to present, granitisation problem has remained as an acute one. Korzhinsky (1952) pointed out a wide-spread replacement of sedimentary sequences by granites, he claimed that the polymineralic character of granites and the quantitative uniformity of their mineral composition strongly indicate the magmatic and not metasomatic nature of their formation. Based on petrologic and physico-chemical considerations, Korzhinsky regarded granitisation as "infiltration magmatic replacement". It is produced by the flow of ascending transmagmatic solutions of abyssal origin, bringing in heat and causing the infiltration magmatic replacement of the rocks. Korzhinsky attributed the diversity of granitic types in different geologic complexes to the dependence of eutectic and cotectic compositions on the K and Na concentrations in these solutions. It has been supposed that the components of transmagmatic solutions (H2O, CO2, R2O) originate in the abyssal zones of the Earth. In the years that followed, this viewpoint was shared by many scientists. In Zharikov review (1987) it was pointed out that Korzhinsky's ideas stood the test of time and became one of the best-founded theories. And yet, we must remember the words Korzhinsky himself wrote back in 1952: "... no conclusive solution to the granitisation problem can be achieved with the help of general petrologic and geologic studies alone... They should be substantiated by accurate physico-chemical theories which would adequately describe the processes postulated, and it is also necessary to carry out reliable geochemical experimentation which would either dispel or support these theories". Pursuing these objectives, Korzhinsky both in his works (1952, 1959) and in lively discussions with colleagues at the Institute, defined a specific set of experimental problems to be dealt with. They can be divided into the following groups.

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- 1. Transport of fluids and their components through the melt, and metamagmatic debasification.
- 2. Study of systems modelling granitoids: the dependence of melting diagrams on the fluid composition.
- 3. Experimental modelling of granitisation (partial melting and magmatic replacement) in multicomponent systems, in particular experiments involving a model fluid which originated in the abyssal zones of the Earth.

Some of this results obtained from experiments along these lines are discussed below.

## Transport of fluids through magmas, perfectly mobile components (alkalies and water) and metamagmatic debasification

The materials pertaining to this studies have been published elsewhere; therefore we shall give here only the important results. The "transport properties" (surface tension, viscosity and diffusion coefficients) of the fluid-silicate system determined in various studies, have made it possible to determine the behavior of fluids interacting with melts. We have determined experimentally the diffusion coefficient for water and all petrogenetic components in the albite melt; a technique has been proposed for calculating  $D_i$  in an arbitrary melt at different P and T (Chekhmir-Epelbaum, 1981). A method has been proposed for viscosity calculations for melts and magmas at any thermodynamic conditions (Persikov, 1984). The surface tension on the melt-fluid interface has been calculated (Epelbaum – Salova, 1977).

All these materials have made it possible to make calculations of the size of the bubbles of the fluid phase under different conditions of its intrusion into magma, as well as the character and velocity of the mass-exchange of bubbles with magma (Epelbaum, 1980). The calculations show that in order to overcome a surface tension while forming a bubble a redundant pressure of less then 0.1 bar is required. One can judge about the sizes of the forming bubbles from different considerations. However, since the sizes of nozzles are unknown, through which fluid is intruded (capillary pores or cracks), then it was expedient to estimate a size of bubbles from the conditions of a mechanical equilibrium of moving bubbles in a viscous medium, i. e. from the equality of a force of a viscous resistance and ascending force. The estimate from these considerations yielded a size of the bubble of not more than the first millimetres.

Thus, if there exists a certain rock permeability in the basis of the magmatic chamber then a formation of bubbles and their further rapid ascending does not involve any difficulties. (In the course of ascending bubbles can dissolve or just exchange by the petrogeneous components what depends on the equilibrium degree of fluid and magma). Such a process, creating an infiltration fluid flow seems quite probable to us.

It will be noted that the rising bubble exchanges components with the magma more actively than the motionless one. Having penetrated into the magma, such a bubble dissolves or continues to exchange components until equilibrium is attained, over a relatively short time. The exchange of components between transmagmatic fluid and melt and a change in the acidic reactivity of the fluid with decreasing temperature produce further debasification of the melt (metamagmatism). The experimental modelling of this effect was described by Salova et al. (1980); Korzhinsky et al. (1983); Epelbaum — Salova (1980).

The investigation of water and alkali diffusion in the melt indicates that, though water and  $R_2O$  are the most mobile (and volatile) components, but on the whole, their mobility is comparable with that of other components and is defined by the Cartly potential  $-\mathbb{Z}/\mathbb{R}$ . They

do not possess any anomalous mobility. So, the "perfectly mobile behavior" is a thermodynamic term connoting a property of natural systems to maintain  $\mu_j$  at a certain level under the action of the fluid flow. Its convenient to refer such systems as open ones.

The influence of such fluids is discussed in the next section.

The modelling of granitoid systems. Melting relations. The role of acid-base interaction and fluid transport

The classical study of Tuttle-Bowen (1958) has clearly demonstrated the eutectic (cotectic) nature of granitoids. Since then extensive studies of various model systems, including investigations of fluid composition, have gained a very important place in the literature. In addition to the temperature effect on eutectic compositions, there are also data on the effects of pressure (Zharikov, 1976) and chemical potentials of perfectly mobile components (Epelbaum-Kuznetsov, 1980). In Fig. 1-a schematic melting diagram  $T-\mu_j-X$  for a three-components system in which A and B are inert components and j is the perfectly mobile one is presented. In this representation, it is possible to determine the eutectic not only with respect to temperature but to the chemical potential of the perfectly mobile component as well.

Very important results were obtained on the effect of minor components in the fluid on the geometry of melting diagrams. The most important works are those of von Platen (1965),

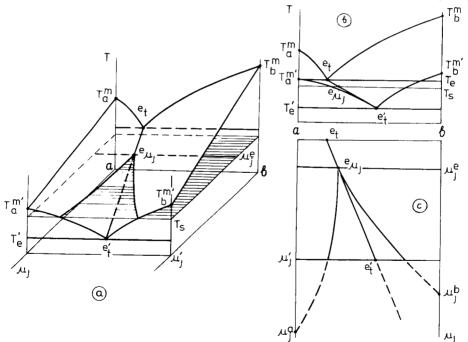


Fig. 1. Schematic  $T-\mu_j-X$  melting diagram for the system containing two inert (A+B) and one perfectly mobile (J) components.

a — the general diagram T- $\mu_h$ -X; b — eutectics with respect to temperature for different; c — eutectic with respect to  $\mu_j$  at  $T = T_s$ .

Manning (1981), Pichavant (1981), Tchelishchev (1967), Kuznetsov – Epelbaum (1985) and others. It has been found that the fluid composition has a very pronounced effect on the composition of melt being generated. It is known that the acidity of a component added to the system influences the activity coefficients of melt components. Hence, the melt eutectic composition is also changed. The mechanism of this effect is simple and relates to the formation of new compounds in the melt.

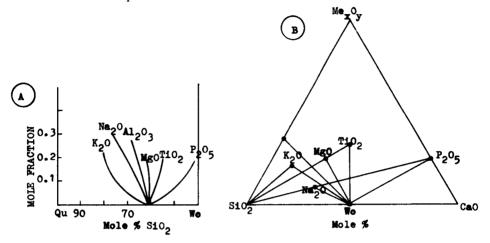


Fig. 2. The effect of additional components on the cotectic composition in closed systems SiO<sub>2</sub>+CaSiO<sub>3</sub>+Me<sub>x</sub>O<sub>y</sub>.

A - after Kushiro (1975); B - explanation for the arrangement of the elementary triangles.

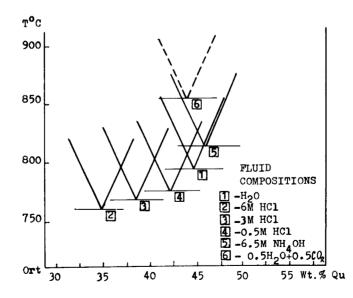


Fig. 3. The effect of fluid acidity on the eutectic composition Qu-Ort. At  $P_{total} = P_{fl} = 1$  kbar.

The changes which take place in the eutectic composition when different components are introduced into the system quartz-wollastonite we illustrated in Fig. 2-a (after Kushiro, 1975). At a triangular diagram 2-b eutectics in the ternary private systems Q-Woll-X are shown, where X – is a mineral, formed as a result of interaction of the system with the added component. A relative location of points, corresponding to X, explains a character of the addition effect on eutectic position.

A very important inference drawn by Korzhinsky (1959) from such diagrams was that the fluid composition influences the composition of the melt (eutectic). We have given much time and effort to study this effect. As an example. Fig. 3 shows the effect of the fluid acidity on the position of the quartz-orthoclase eutectic, the effects of fluid acidity on the ternary eutectics Q-Or-Bi and on the melt-minimum in the system Ab-Or we shown in Figs. 4 and 5 respectively (from Kuznetsov — Epelbaum; 1985; Bogolepov — Epelbaum, 1987). All these experiments indicate that although the melts from quartz-feldspar substrates are compositionally close to granitoids, the fluid composition has a marked effect on the exact composition of the primary melt and its temperature. For example, this effect is comparable with that of  $H_2O$ -pressure.

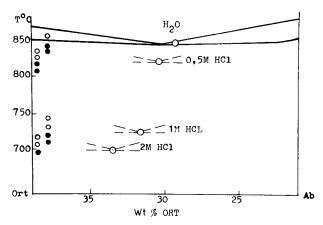


Fig. 4. The effect of fluid acidity on the composition of the ternary eutectic Q-Ort-Bi. At  $P_{total} = P_{fl}$  = 1 kbar. The results of T determination are shown in the left part of diagram.

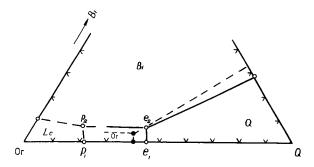


Fig. 5. The effect of fluid acidity on the composition of the ternary eutectic Q-Or-Bi e – eutectic; p – peritectic;  $P_{total} = P_{fl} = 1 \text{ kbar}$ ;  $-H_2O$ ; - – 3n HCl.

Melting in a fluid-silicate system; the modelling of "granitisation" as melting in multicomponent systems, particularly in the presence of a "basaltic" fluid

In fluid-silicate systems, heating to the melting temperature exhibites some peculiar features that have been, however, neglected. It is very essential that the melt develops not only on mineral contacts but also over the surface of each individual grain in contact with the solution.

The experiments which support this conclusion are shown schematically in Fig. 6. A mineral charge in which we determine the first melt composition place into the downer part of the platinu-m tube, then water or solution in which presence melting will take place, and in the upper part of the tube minerals are suspended. It is convinient to take feldspars which are present in the charge and enter the composition of the first melts (Fig. 6a) as melt indicators. Naturally, in any other systems minerals-indicators would be different.

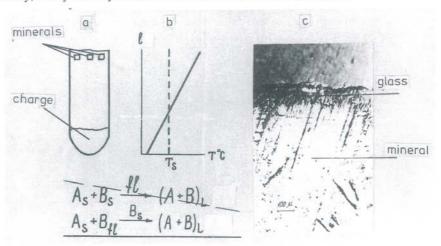


Fig. 6. Experimental scheme: a-Pt-tube with charge and minerals; b-g radient of temperature in the tube; c-mi rophotograph of thin section of the mineral-indicator showing melt formed on the surface.

The tube is hermetized and then in the high pressure set it is introdused into the necessary regime  $P_{fl}$  and T. Here along the tube such a gradient of temperatures is created when minerals-indicators are at the temperature higher than T of the minimum melt, but lower than T sol. of separate minerals-indicators; the charge is somewhat lower than T of the minimum melt (Fig. 6b). Thus, chemical potentials of the charge minerals are supported in the fluid. Usually we write down an eutectic melting (for instance, for a two-component charge A + B with the fluid) as follows:

 $A_S + B_S \xrightarrow{fl} (A + B)_L$ 

However, due to the equality of chemical potentials of the components in all co-existing phases one can write melting in such a way:

$$A_S + B_{fl} \stackrel{B_S}{\to} (A + B)_L$$

It is such melting that is realised on the surface of minerals-indicators. The thin section fragment of this mineral after quenching is shown in Fig. 6c. The melting according to this equation is impossible without fluid transfer of the components to the surface of grains. Such

Table 1

Compositions of "melt-min" in the system Q-Ab-Or as determined using the present technique and conventional methods

minerals	normative mineral contents (wt. %) in the melt at different P										
	1 kbar				5 kbar						
	on Ab	on Or	mean	Tuttle 1958	on Ab	on Or	mean	Luth 1964			
albite	36.7	37.4	37.1	38 0	47 0	48.9	48.0	50.0			
orthoclase	24.0	24.6	24 3	25.0	24.1	23.0	23.5	24.0			
quartz	39.3	38.0	38 6	37.0	28.9	28.1	28.5	26 0			

\$T\$ a b l e  $\,$  2 The chemical composition of the primary melt (wt. %) which formed from partial melting of different mineral substrates  $T=730\,^{\circ}C$ 

substrate	gneiss	granite	50% diop- side + 50% (Q-Ab-Or)	granite	granite	70% bas- alt +30% quartz	basalt
fluid	$H_2O$	$H_2O$	$H_2O$	lm KCl	1m NaCl	$H_2O$	$H_2O$
pressure	5 kbar	5 kbar	5 kbar	5 kbar	5 kbar	5 kbar	decompression from 5 to 2 kbar
Na <sub>2</sub> O	4.9	5 0	5.0	2 7	7.0	6.0	4 1
MgO	0.15	0.18	0 2			-	0.4
$Al_2O_3$	17.15	18.5	17 9	16.8	19.3	18.5	17.5
SiO <sub>2</sub>	70.8	70.3	70.9	71 2	68.9	68 5	74.6
$K_2O$	5.55	5.6	5.35	7 9	4 7	4.7	2.4
CaO	1.7	0.22	0.45	1.4	0.1	0.4	0.6
FeO	0.2	0.17	0.2	-	-	-	0.4

melting is a feature of the fluid-silicate system only. As a result of the careful X-ray microprobe analysis it has been established that a composition of the forming glass at the boundary with a fluid on different minerals-indicators is identical to an eutectical one (Bogolepov-Epelbaum 1988).

It is impossible to confuse a forming glass (dense, transparent) with a quenching phase (friable, mat), which can be also observed under a careful examination on the tube walls or on the charge surface. Moreover, melt film is neither formed on noneutectical charge minerals suspended above, nor on any minerals at temperatures lower than T sol., whereas a quenching phase must take place in all cases.

On the one hand, the described observation of the melt forming at the mineral-fluid contact opens new possibilities of studying the initial first melts, on the other hand it explains a simultaneous replacement of grains of different eutectical minerals by the melt.

New experimental uses to which this technique can be put include studies of multicomponent melts and natural substrates.

Note that the composition of the primary melt (eutectic, cotectic minimum) is reliably determined in the single run for a given composition of the substrate, any fluid and pressure.

Tab. 1 compares the results from these and conventional runs. The agreement is fairly good, but Tuttle's results come from a series of descrete runs whereas ours are obtained from a single experiment. According to Tab. 2, the primary melt varies little with the substrate composition, which is no surprising because the melt is an eutectic one. The Fig. 7 shows normative compositions for the first melts from different substrates. It is seen that anchieutectic granitoid melt develops at whatever substrate bulk composition. At the same time, the melt composition depends strongly on the fluid alkalinity (Tab. 2 and Fig. 7).

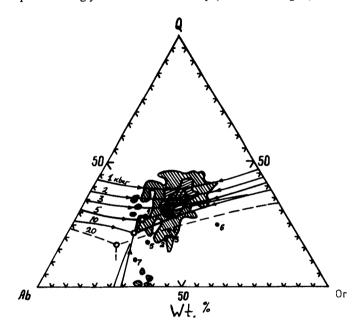


Fig. 7. Partitioning of normative Ab, Or, Q from 1269 rock analyses after Tuttle (1958). Cotectic lines and open circles are as in Tuttle (1958) and Luth et al. (1964). Dark circles are for compositions (recalculated to 100% Ab, Or, Q) of experimental melts at 5 kbar in the systems: 1 - Ab-Or-Q; 2 - Bi-Sill gneiss; 3 - granite; 4 - 50% diopside + 50% Ab-Or-Q (mixture); 5 - basalt + quartz (all in the presence of  $H_2O$ ); 6 - granite + in KCl; 7 - granite + in NaCl.

Now, a critical question is whether granitisation can take place if the wall rock is attacked by a fluid of abyssal origin, which was in equilibrium with highly basic rocks. Tab. 2 presents the results of modelling experiments on this problem. It has been found that forming of granite melt follows the same pathway in the presence of the mixture quartz + basalt. It will be stressed that the decompression of abyssal fluid is accompained by large quantities of SiO<sub>2</sub> being introduced into the wall rock. Fig. 8 shows variations in the silica content of the fluid which is in equilibrium with pegmatite Spruce Pine (Handbook of physical constants, 1966), basalt (Gorbachev-Kashirtseva, 1985) and phlogopite-forsterite assemblage (Ryabchikov-Boettcher, 1980), as a function of pressure. Thus, the intake of alkalies and SiO<sub>2</sub> is a result of decreasing fluid pressure. And this supply of quartz and alkalies will be sufficient for a granite melt to form even during granitisation of basic rocks.

Finally, the mode of melting in fluid-silicate systems accounts for the replacement mechanism as such. The melt appears simultaneously on the surface of different minerals which can even be out of contact with one another. The necessary components are brought up by the fluid from other minerals with the result that during melting the mineral grain is replaced by the melt. With respect to granitisation, we cannot pass over the mechanism of supply and loss of components during this process. Considerable experimental evidence from different laboratories suggests that SiO<sub>2</sub> and alkalies can be brought in by the abyssal fluid. For example, Gorbachev-Kashirtseva (1985) found about 87 wt. % SiO<sub>2</sub>, 4 wt. % Na<sub>2</sub>O+K<sub>2</sub>O, 1 wt. % CaO, 5 wt. % Al<sub>2</sub>O<sub>3</sub> in the dry residue of the fluid in equilibrium with basalt. The contents of FeO and MgO are low. This implies that the fluid cannot dissolve Fe, Mg, Ca from the substrate undergoing granitisation on a level required by the direct dissolution scheme.

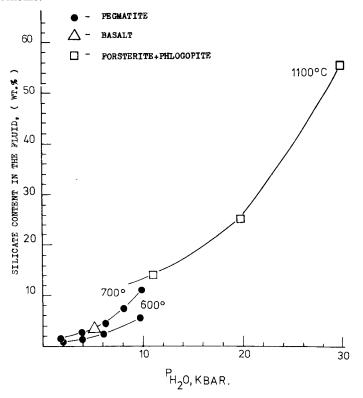


Fig. 8. Silicate content in the hydrous fluid in equilibrium with pegmatite, basalt and phlogopite-forsterite assemblage as a function of pressure.

Zharikov (1987) wrote that debasification is a one-directional process. We share this view point and emphasize that the process follows different pathways at different stages of granitisation:

I. The feldspatisation (and silicification) stage involves the intake of alkalies and SiO<sub>2</sub> as a result of decreasing fluid pressure (Fig. 7).

II.—III. The beginning and development of melting. Migmatisation — it appears that forming a leucocratic magma is the main mechanism during partial melting. At this stage, there is also supply of material due to decompression.

IV. At this stage granitic bodies are formed as a result of melt localisation. The debasification of the melt formed can persist with decreasing temperature and increasing fluid acidity, i. e. under the action of metamagmatic processes.

The following conclusions can be drawn from the experimental modelling of granitisation:

- 1. The present experiments have shown that the primary melts from multicomponent substrates of a diverse composition are granitoid in composition with low biotite (3-7 wt. %) on the eutectic.
- 2. The mineral ratios of granite are defined by the fluid regime (acidity, alkali metal content of the fluid) at the melt generating stage, and by the metamagmatic process patterns at the final stage.
- 3. Granitisation involves the supply of components by the fluid (granitic material in fact) and mobilisation in the parent rock substance. Incomplete melting in the open system results in a granitic melt.
- 4. It has been shown, that granitisation can be produced by a "basic" fluid as a result of the leaching of the dissolved substance (during decompression) and incomplete melting in the open system.

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