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FLUID REGIME OF GRANITE FORMATION IN ZONES OF ULTRAMETAMORPHISM

(5 Figs., 1 Tab.)

Abstract: Petrological data on the Ladoga, Muzkol, Belomorian and Aldan complexes provide a great body of evidence for P-T conditions of ultrametamorphism, the bulk fluid composition and the nature of mineral-forming media. It is shown that the onset of ultrametamorphism, when anatexis occurred, is likely to be characterized by the temperature 680–770 °C (Ladoga: P=4.5–5 kbar; Aldan: P=5 kbar), 705–750 °C (Muzkol: P=8–9 kbar), 700–715 °C (Belomorian: P=7.5–8 kbar), 800–850 °C (Aldan: 6 kbar). These temperatures correspond to a decrease in H_2O activity from 0.8–0.9 to 0.3–0.4 in the transition from amphibolite to granulite facies.

On the basis of the fluid and melt inclusions study it is suggested that originally homogeneous Co_2-H_2O-salt fluid is unmixed into two phases at temperatures close to the granite solidus: 1) a gaseous one consisting mainly of CO_2, sometimes with admixture of CH_4 and other hydrocarbons, and with an insignificant amount of water and N_2; 2) a liquid representing a high salinity aqueous solution with NaCl, KCl, $CaCl_2$ as the main species. Taken together all these data permit a model composition of the fluid participating in ultrametamorphism to be calculated. The proposed model "equal hydrogene concentration" seems to provide a consistent explanation of the H_2O mole fraction decreasing under high temperature within single fluid flow.

Key words: P-T conditions, ultrametamorphism, granites, fluid inclusions, Baltic Shield, Aldan complex.

Introduction.

Ultrametamorphism is a combination of intimately interrelated phenomena including both the replacement of gneisses and schists by granitoid rocks and the melting of them (granitization or magmatic replacement by Korzhinsky, 1952).

Notable extents of magmatic replacement, physico-chemical and geochemical homogeneity of the autochthonous granitoids, and pertinent thermal regimes of granitization within particular tectonic zones suggest that ascendent fluid flows (transmagmatic solutions by Korzhinsky, 1952) are important means of mass and heat transfer in ultrametamorphism (Sedova–Glebovitsky, 1984; Sedova in this volume). In these flows the water activity decreases in deep parts. In order to explain the low H_2O various models have been invaded including dilution of pore fluid with CO_2 external buffered (Touret, 1971; Newton, 1985), that is in agreement with Korzhinsky's idea.

Petrological data on the different metamorphic complexes are presented here. These data allow calculation of a model composition of the fluid participating in such ultrametamorphism to be made.

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P-T conditions of metamorphism and ultrametamorphism

Low pressure Ladoga complex, Baltic Shield. Highly precise estimates of P-T conditions in Svecofennian belt (Fig. 1) have been performed based on the garnet-biotite-cordierite thermobarometer (Glebovitsky–Drugova, 1979; Perchuk et al., 1983), using data on the mineral assemblages Grt+Bt+Sil+Mus and Grt+Bt+Cord+KFsp which occur in Ladoga complex (Glebovitsky et al., 1985). Redistribution of Mg and Fe among Grt, Bt and Cord in retrograde metamorphism results in the marked displacement of calculated P-T conditions from garnet-cordierite-orthoclase subfacies of granulite facies toward lower P and T, for example in leucosomes of late migmatites (T=600 °C, P=3 kbars). Maximum temperature estimates, however, are fairly consistent with those of homogenization of melt inclusions from leucosomes in early migmatites of different zones: 775 ° and 770 °C – for granulite facies, 700 ° and 710 °C for a high temperature part of amphibolite facies, and 680 °C for the first migmatites emerging in the stability field of the mineral assemblage Grt+Bt+Sil+Mus. Pressure estimates (4.8–5.5 kbars) based on the most dense CO_2 inclusions in early leucosomes developed earlier to diatectic magma (Glebovitsky et al., 1985) (γδ, Fig. 2) are always higher than those determined from Grt-Bt-Cord barometry (3.8–4.8 kbars). Thus, the onset of ultrametamorphism when anatexis occurred is characterized by temperatures of 680, 700–710 ° and 770–780 °C respectively at pressure 4.5–5 kbars. It is important to note here high temperature of stability of muscovite assemblages (more 680 °C), corresponding to coexistance of melts with an almost pure H_2O fluid (mole fraction of water close to 0.9), that is agreement with the generation of low-temperature (680 °C) granitic melts.

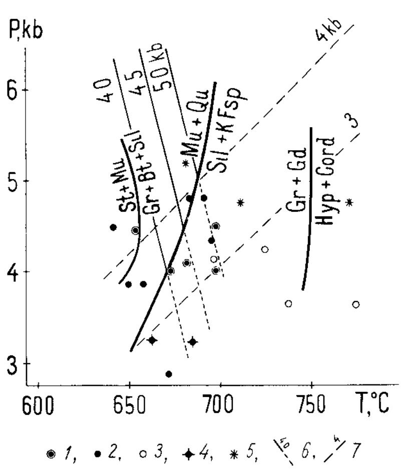

Fig. 1. P-T conditions of metamorphism and ultrametamorphism of Proterozoic assemblages from the Svecofennian belt.

Temperature and pressure estimates are for: 1 – Grt-Sil-Bt-Mus; 2 – Grt-Cord-Bt-Orth gneisses of amphibolite; 3 – and the same rocks of granulite facies; 4 – Grt-Bt granites and granodiorites constituting the leucosome of IV generations using mineralogical thermometers; 5 – for same rocks P-T are determined from the melt inclusions; 6 – the diagram shows isobars of a water (P_{H_2O}) for equilibrium Mus+Sil+KFsp; 7 – and for the fluid in equilibrium with the granite melt.

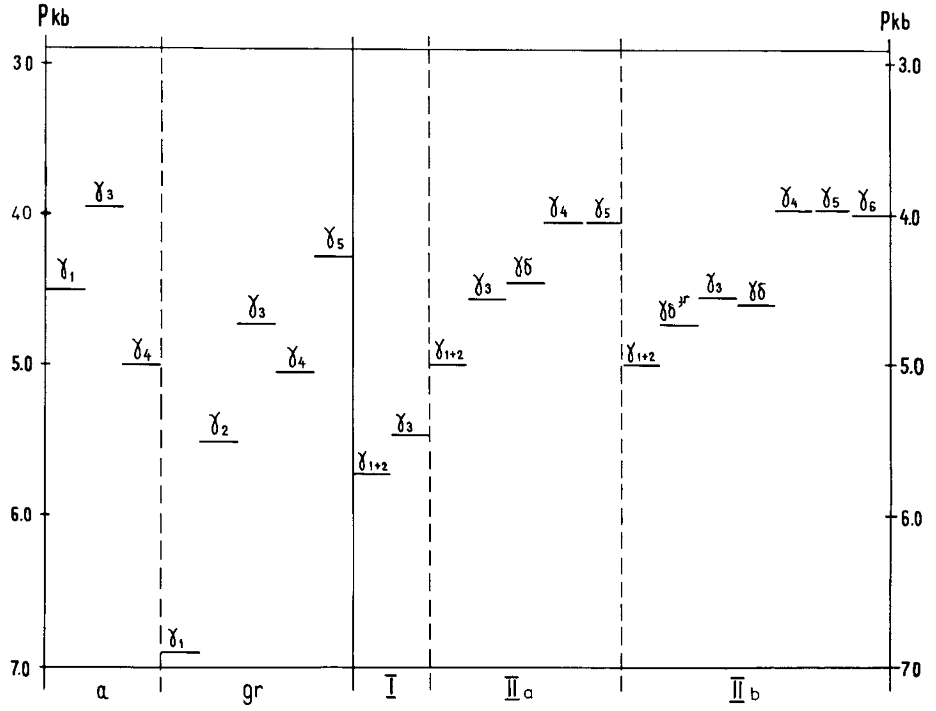

Fig. 2. Pressures of formation of ultrametamorphic granitoids in the zonal metamorphic complexes. Aldan and Ladoga complexes determined from the most dense CO_2– inclusions. The Aldan complex: successive generations of granitoids under conditions of amphibolite (a) and granulite (gr) facies: γ_1, γ_2, γ_3, γ_4, γ_5. γ_3, γ_5 – granitoids of diatectic genesis. The Ladoga complex: successively forming leucosomes of migmatites under conditions of amphibolite (I), granulite (IIb) facies and in the transitional zones (IIa): γ_{1+2}, γ_3, γ_4, γ_5, γ_6. $\gamma\delta$ – granodiorites, $\gamma\delta^{gr}$ – enderbites. γ_3, $\gamma\delta$ – diatectic granitoids.

High pressure complexes. It means metamorphic complexes attributed to kyanite-sillimanite type. A temperature range of 600–690 °C and correlated pressure variation from 6 to 8 kbars (Fig. 3) occurs within the staurolite zone of the Alpine Muzkol complex from the Central Pamirs (Letnikov et al., 1980). Maximum pressures of 9 kbar are recorded at the high temperature boundary of the staurolite zone where the mineral assemblage Grt+Bt+Ky+Mus is preserved at $T=675$ °C. The high temperature both of the staurolite and the above mentioned assemblage suggests that metamorphism occurred at thigh partial pressure of $H_2O \geqq 7$ kbars (H_2O mole fraction in fluid is about 0.8).

The temperature of complete homogenization of melt inclusions in migmatites which occur at the upper stability limit of muscovite and within the highest temperature zone varies from 705 to 780 °C at P ranging from 8.5 to 8.0 kbars. Likewise in the adjacent kyanite-muscovite zone, the calculation of H_2O mole fraction $= 0.8$ in equilibrium fluid corresponding to the lowest-temperature melts suggests that migmatization and metamorphic recrystallization were simultaneous. It is evident that both the H_2O mole fraction in the fluid and its activity will decrease in a zone with newly formed high-temperature melts.

The upper Archean Belomorian complex (Baltic Shield) is characterized by a zoned metamorphic sequence (Glebovitsky et al., 1985). The metamorphic conditions inferred from the observed zoning vary from Grt-Bt-Ky-Mus to Grt-Bt-Ky (or Sil)-Ort subfacies, which correspond to the temperature range 600–660 °C estimated on the basis of Grt-Bt thermometer. Pressures vary in the range from 7.5 to 9.0 kbars (Fig. 3). Pressure determined from the most dense CO_2 inclussions in migmatite leucosomes does not exceed 7.5 kbars. The univariant equilibrium of Grt+Bt+Ky+Mus+KFsp (Yolodichev, 1975, 1977) suggests a temperature range from 600 to 625 °C at an average pressure of 7.5 kbars, while the measured temperature of kyanite-ortoclase-absent muscovite gneisses varies from 615 to 645 °C at average pressures of 8.5 kbars. This suggests that the approximate temperature of muscovite dehydratation at a pressure of 8 kbars is 620 °C. Hence, P_{H_2O} is 3.5 kbar, corresponding to a H_2O mole fraction in the fluid of 0.44. Melt inclusions in the leucosomes of early anatectic migmatites yield consistent temperatures of complete homogenization 705 °C (Glebovitsky et al., 1985). Therefore, the transition of the Belomorian gneisses to anatexis also implies an increase in the partial pressure and mole fraction of H_2O in the fluid up to 6.4 kbars and 0.8, respectively, together with additional heating and decopression from 9 to 7.5 kbars.

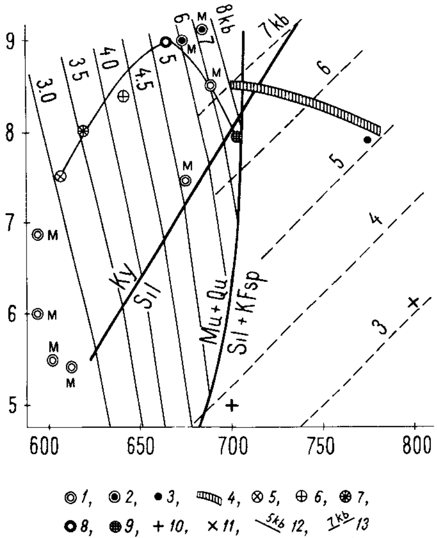

Fig. 3. P-T condititons of metamorphism and ultrametamorphism of Paleozoic and Mesozoic units of the Muzkol complex, the Central Pamirs, of Archean Belomorian and Aldan complexes.
The Muzkol complex (M): conditions of mineral assemblage formation 1 – St+Grt+Ky+Bt+Mus; 2 – Grt+Ky+Bt+Mus; 3 – Grt+Sil+Bt+KFsp based on garnet-biotite thermometer and; 4 – barometer, for anatectites on the basis of the melt and simultaneous fluid inclusions.
The Belomorian complex: 5 – middle parameters of formation of mineral assemblages Grt+Bt+Ky+Mus; 6 – Grt+Bt+Ky+KFsp; 7 – Mus+Ky+KFsp based on Grt+Bt thermobarometry; 8 – Grt+Hb+Cpx+Ky+KFsp+Pl based on Grt+Cpx+Amph thermobarometry; 9 – anatectites from the melt and simultaneous fluid inclusions. *The Aldan complex:* 10 – for mineral assemblage Grt+Bt+Cord+Fsp at amphibolite; 11 – granulite facies. The diagram shows water isobars in equilibrium with the melt – 12 – and for the muscovite-bearing assemblages – 13. Pressure–kbar.

Aldan granulite complex. Like the Ladoga complex, P-T parameters of metamorphism and ultrametamorphism within the Western part of the Aldan granulite-gneiss area have been determined using garnet-biotite-cordierite thermometry and barometry. Granulite-grade rocks (T=800 °C, P=6.1 kbars, Fig. 3) are widespread, while rocks of high temperature subfacies of amphibolite facies (T=700 °C, P=5 kbars, Fig. 3) occur near the Western margin. These P-T conditions suggest the existence of different levels of exposure in the region. Pressure is independently determined from pure CO_2 inclusions, the most dence ones being correlated with maximum temperature in the given zone. This indicates that pressures during early migmatization to be 6.0–6.5 kbars in the granulites and 5 kbars (Fig. 3) in the amphibolite grade rocks (Vapnik–Sedova, 1986) and is in exelent agreement with the estimate based on mineral equilibria.

Fluid inclusions and their compositions

On the basis of investigations over the last 20 years fluid inclusions occurring in rocks metamorphosed at amphibolite and granulite facies can be classed into four types: 1) CO_2-inclusions; 2) low salinity solutions (1–3 mass % NaCl equivalent), high salinity solutions (3–25 mass %) and aqueous salt brines; 3) mixed H_2O-CO_2 inclusions; 4) CH_4 and N_2 inclusions. The first two types are the most abundant in the granitiods formed under conditions both amphibolite and granulite facies.

CO_2 inclusions are of great interest and attract much attention from petrologists. Their discovery in the highest temperature metamorphic rocks (Touret, 1971) has led some investigaters to suggest that a dry fluid of nearly pure carbon dioxide participated in granulite-grade metamorphism. It is now clear (Sedova, 1977; Vapnik–Sedova, 1986; Vapnik, 1988) that such inclusions are characteristic for both granulite and lower-grade rocks.

Purity of the CO_2 inclusions in the metamorphic minerals in this study has been verified on the basis of the decrease of melting temperature below −56.6 °C by using a criometrical technique. Sometimes significant changes in the melting temperature could be attributed to CH_4 which occurs in mole fractions exceeding 0.1. Smole admixture of N_2 is not recognized by this method because nitrogen results only in decreases of homogenization and causes a systematic error in the density determination. Perhaps pressure overestimation using CO_2 inclusions is a consequence of the presence of N_2 in the fluid.

High density CO_2 inclusions are commonly considered to have originated during a metamorphic culmination or soon after (Berdnikov–Karsakov, 1984; Perchuk, 1985; Berdnikov, 1987; Touret, 1971; Rudnic et al., 1984; Touret–Dietvorst, 1983; Santosh, 1986; Newton, 1985; Schreurs, 1984; Hansen et al., 1984a, b). This approach readily account for the qualitative consistency between pressures determined on the study of CO_2 inclusions and mineralogical geobarometry.

Another abundant type of inclusion comprises aqueous salt solutions. These include two-phase (salt solution+gas) and single-phase (salt solution), and rarerely three-phase (salt crystal+salt solution+gas) systems at room temperature. A freezing temperature depression of up to −60 °C is recorded for such inclusions with common salinities of 20–25 mass % NaCl (Fig. 4) (Konnerup–Madsen, 1979; Sisson et al., 1981; Vapnik–Sedova, 1986). This cryometric evidence is indicative of predominant $CaCl_2$ in the aqueous salt solutions (Borisenko, 1982). The majority of investigations report a young relative age of such inclusions on the basis of their elongate angular shape and arrangement along certain crystallographic planes (Santosh, 1986; Rudnic et al., 1984; Touret–Dietvorst, 1983;

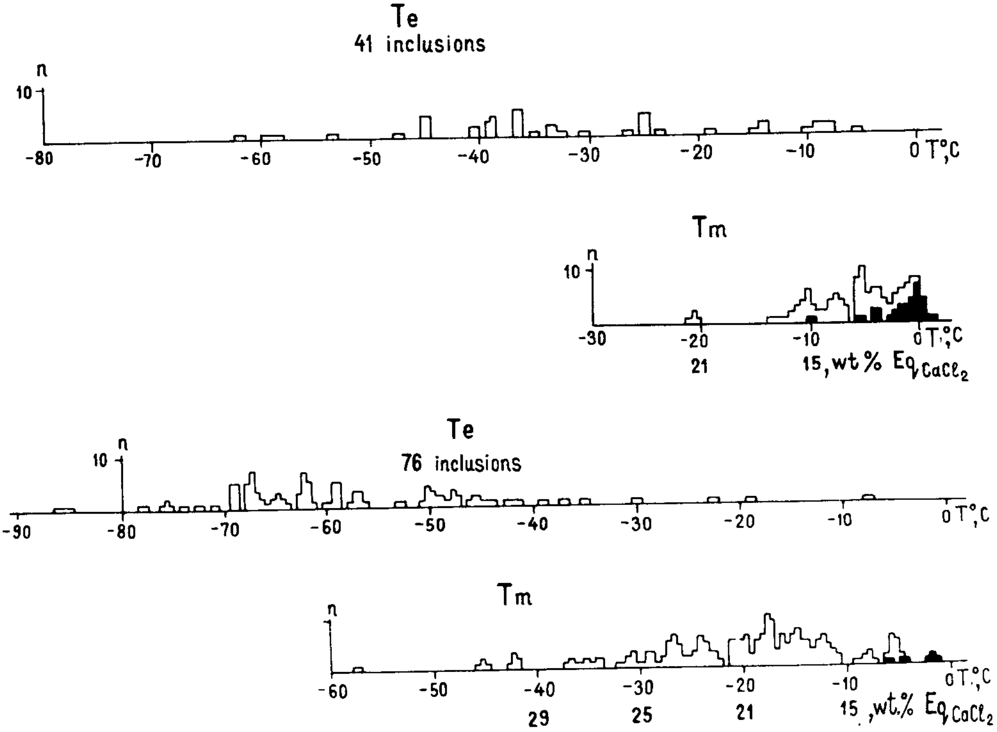

Fig. 4. Initial (Te) and terminal (Tm) temperatures of melting of aqueous-salt inclusions in ultrametamorphic granitoids from the Ladoga (4a) and Aldan (4b) complexes.

Low value of Te (<−50 °C) are indicative of the dominance of $CaCl_2$ in solutions (Borisenko, 1982). Black colour on the diagrams Tm shows the inclusions with Te>−25 °C, i. e. those with dominant NaCl . wt . % Eq_{CaCl_2} – is a concentration of salt in equivalent of $CaCl_2$.

Berdnikov–Karsakov, 1984; Hansen et al., 1984a; Konnerup–Madsen, 1979; Newton, 1986).

However, an alternative approach is to explains the origin of the aqueous-salt inclusions as synmetamorphic followed by the CO_2 inclusions formed after a metamorphic peak. In addition the aqueous-salt inclusions either isolated or restricted to definite directions can be regarded as representing early or even pre-metamorphic products, as was suggested by Touret (1985).

The origin of the isolated aqueous solutions, CO_2 and other gases remains yet uncertain. Nevertheless, we do not think that these different gases were introduced at different times. An alternative explanation seems to be the breakdown of an originally homogeneous fluid into two phases: a gaseous, essentially carbon-dioxide-bearing one, and a concentrated liquid solution. Examination of the phase diagram H_2O-CO_2-NaCl (Shmonov et al., 1986) suggests the presence of a "solvus" marked domain of immiscibility at temperatures near the granite solidus. On the other hand, the temperature of homogenization of the salt inclusions were shown to reach 650 °C (Sedova, 1977). Good agreement between the pressure measured on the basis of the most dense inclusions and those estimeted using mineralogical barometers are therefore consistent with this mechanism.

On the base of study of mechanism of fluid entrapment by a growing crystal it is evident that conservation of volatiles in the defects of crystalline lattices or other limited volumes may occur alongside with the formation of the true primary inclusions with typical observed diameter (5–25 μm). In this case, entrapment of a different proportion of the fluid components is to be expected and a subsequent coagulation (Geguzin et al., 1971) may be responsible for the formation of pure gaseous inclusions. Naturally, the constituent of such inclusions should have a significantly lower density than those corresponding to the peak of metamorphism.

Numerical model of fluid composition

The above mentioned concept of dry fluid, developed after the discovery of pure carbon dioxide inclusions in granulite-grade rocks (Touret, 1971) was not confirmed, because in ultrametamorphic zones of the amphibolite facies the situation is found to be similar. In addition, there is good reason to suppose that water, bacause of its distinctive behaviour (as a polar gas), is preferentially trapped by crystal defects or retained in other microscopic volumes, thereby creating an illusion that CO_2 is dominant in inclusions (Shmonov et al., 1984).

Two alternative hypotheses have been formulated to explain the abrupt decrease in water activity in the fluid: 1 – the effects of dissociated electrolyte on H_2O activity (Aranovich, 1984) and 2 – a lower activity of water in fine pore relative to free space (Shmonov et al., 1984).

To test the first hypothesis the analysis of equilibrium conditions of garnet-cordierite-hypersthene-sillimanite assemblages in deep-seated granulites have been used (Aranovich, 1984). The real proportions of Mg/(Mg+Fe) ratios in garnet (0.5–0.6) of this equilibrium were found to correspond to extremely high values for CO_2 activity. As suggested by Aranovich (1984), these high CO_2/H_2O ratios are due to effects of an electrolyte whose dissociation degree is increasing with increasing pressure.

The main features of interaction of fluid components in homogeneous mixtures should be treated to find a satisfactory solution of the problem.

Theoretical and experimental data (Kerrick–Jacobs, 1981; Jacobs–Kerrick, 1981; Shmulovich et al., 1982) show that the mixture H_2O-nonpolar gas is non-idial. Matual increases in activity coefficients are observed, this effect being less with increasing temperature and $\alpha^*_{H_2O}$ approaches 1. Introduction of electrolyte increasingly lowers this value as temperature decreases or pressure increases, whereas activities of nonpolar gases increasingly become higher (Shmulovich et al., 1982; Bowers–Helgeson, 1983). If the assumption of $a_{H_2O}=0.4-0.1$ in granulite facies (T=800 °C, P=5 kbars) is valid then this activity should be 0.35–0.05 in amphibolite facies of the same depth (T=700 °C, P=5 kbars), thus farbiding granite melting. This contradicts observations from zoned metamorphic complexes. Nevertheless, the above considerations may explain a transition to anatexis with increasing temperature and concurrent decompression in the Belomorian complex.

The second hypothesis regarding the drop of H_2O activity under granulite facies conditions as a result of the effect of pores also fails to solve the problem. Calculations on the basis molecular dynamics suggest that the effect of water-crystal interfaces with an average pore size down to 10^2 Å results in an abrupt drop of H_2O fugacity.

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*a – activity, α – activity coefficient.

A corrected fugacity coefficient is 0.75 at pore width 81.42 Å and $\varrho = 1.2$ g/cm^3, and 0.43 at pore width 30 Å and $\varrho = 0.8$ g/cm^3 (Belonozhko–Shmulovich, 1986). The applications of the calculations to explain the lower water activity in fluid requires a very fine dispersion of the fluid (pore 10^2 Å) during granulite metamorphism, which, however, precludes the development of primary inclusions of the sizes observed ($> 10^5$ Å). This value is the limit for the minimum pore size during a peak of metamorphism, consequently, this effect does not explain the specifics of the fluid regime during granulite facies metamorphism, but it can be used to interpret a trapping mechanism of water from a composite fluid by growing crystal. Due to preferential capture of water at a submolecular level in microscopic volumes an impression arises that the inclusions, especially primary ones, are dominated by CO_2. The theoretical calculations and experimental data mentioned above enable us to estimate activity coefficients of major fluid constituents (Tab. 1).

Table 1

Activity coefficients of fluid components in the system H_2O-nonpolar gas-electrolite under conditions of the amphibolite and granulite facies

Facies	T °C	P kbars	H_2O	CO_2	CO	CH_4	H_2
amphibolite	700	5	0.80	1.32	1.38	1.38	1.76
granulite	800	5	0.85	1.10	1.10	1.10	1.32
granulite	800	7	0.80	1.20	1.20	1.20	1.45

The reported values indicate that the fluid dewatering in granulite facies rocks cannot be explained by the interaction of the fluid components, and must be relate to real decreases in H_2O mole fraction within a single flow. A model of such fluid flow showing vertical differentiation is treated below.

The most important parameter in terms of fluid equilibrium is oxygen fugacity expressed as log f_{O_2}. Calculations of mineral and fluid equilibria for deep-seated petrogenesis suggest that this value is on average similar to that of the QFM-buffer, deviating from it towards the CCO-buffer, and rarely acquiring much lower values (Ryabchikov, 1975; Perchuk, 1973; Hansen et al., 1984; Mattioli–Wood, 1986).

The calculation of a model fluid for different thermodynamic conditions is based on the idea of extremely high mobility of H_2 which is accumulated and oxidized at the upper portion of the flow, enriching it in H_2O. Traces of this hydrogen maximum at the front of magmatic replacement have been detected through analysis of bulk fluid composition (Letnikov et al., 1980). Hydrogene concentration is given as a function of log f_{O_2} such that $X_{H_2O}=0.9$ at the fixed log f_{O_2} (T=1000 K, P=5 kbars) under conditions of the amphibolite facies. X_{H_2} is identical for both facies at the same log f_{O_2}. Fig. 5 shows the model of gas equilibrium calculated taking into account activity coefficients of the fluid compositions in the mixture H_2O-nonpolar gas-electrolyte (Tab. 1). The calculations indicate that at a difference in temperature of 100 °C within the field of graphite instability (to the right of the CCO-buffer) $X_{H_2O} \gg X_{CO_2}$ for the amphibolite facies and $X_{CO_2} < X_{H_2O}$ for the granulite facies. These are in very good agreement with the inferences from the determinations of temperatures of anatectic granitic magmas and H_2O mole fraction in the equilibrium fluid. Changing activities of the fluid components in the mixture H_2O-nonpolar gas-electrolyte enhance the temperature effect of the fluid dewatering, but they are not determining factors since this effect is fairly strong even for assumed ideal behaviour of gaseous mixtures. In addition, H_2O/CO_2 ratio increases somewhat with increasing pressure, but not sufficiently to significantly change the picture.

At f_{O_2} values below those defined by the CCO-buffer, fluid component composition changes mainly by increasing contents of H_2 and especially CH_4 and other hydrocarbons and lowering contents of CO_2 and CO. This appears to be the approximate original composition before anatexis and oxidation in reaction with the rocks during metamorphism. It is important to note that in the amphibolite facies X_{H_2O} is equally high both under reducing and oxidizing conditions, whereas in the granulite facies it shows much higher values at lower f_{O_2} than at higher f_{O_2}. Hence, if amphibolite-grade rocks together with their anatectites underwent granulite facies metamorphism a crystallization of melt would occur followed by a new burst of anatexis accompanied by high temperature recrystallization as temperature rose to 800 °C. This approach to the thermodynamic and fluid regimes offers the possibility to interpret charnockitization superimposed on granite-gneisses of amphibolite facies know in S. India, Sri Lanka, the Aldan Shield and elsewhere in the world.

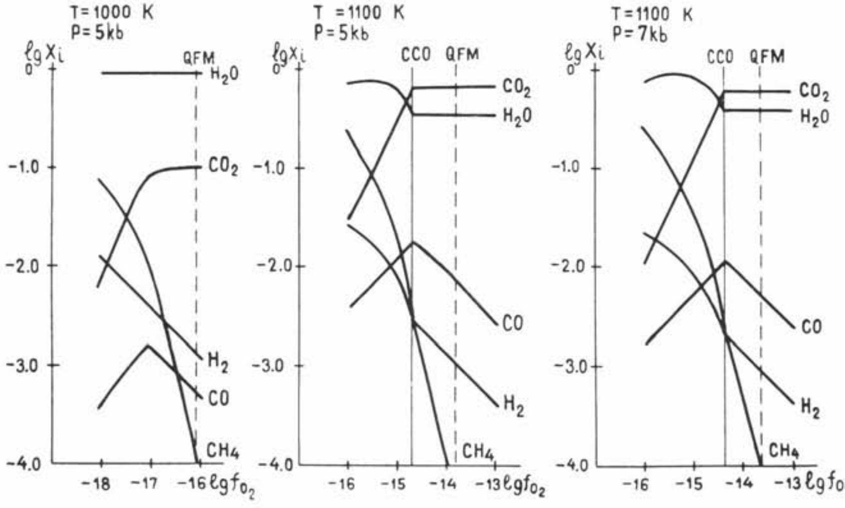

Fig. 5. Isoconcentration model of hydrogene in a single fluid column under conditions of amphibolite (T = 1000 K, P = 5 kbar) and granulite (T = 1100 K, P = 5 and 6 kbar) facies of metamorphism.

The evolutionary path of fluid systems during crystallization of anatectic melts with stratification of the originally homogeneous fluid whose patterns of compositional change as functions of temperature and time have been treated above is of importance. The above evidence of heterogenization, though being not comprehensive, appears to be rather convincing to infer the separation of the gaseous, essentially carbon dioxide bearing, phase from the liquid aqueous solution (salting-out effect) at near solidus temperatures. Two important consequences are inferred from this conclusion. First, a significant tendency for water fugacity to rise in this process, would, coupled with decompression, stimulate a new wave of anatexis and granitization due to the stepwise increase in transfer ability of pure water fluid. Both effects of the fluid segregation following the crystallization of diatectic magmas have been recorded in the Ladoga and Karategin complexes. Second, the separation of a gaseous phase showing high mobility and acidity is one of the causes of the appearence of an advance wave of acidic components (Korzhinsky, 1959). Further cooling of the water-rich solutions enhances the acidic tendencies. A whole pattern of the allochemical transformations at the post-migmatitic stage of the processes leading to the formation of associated basic, acidic and alkaline metasomatites within regionally metamorphosed sequence appears to be determined by these mechanisms (Glebovitsky–Bushmin, 1983).

Conclusions

A combination of data both on total composition of fluid in metamorphic rocks and autochthonous deep-seated granites and various fluid inclusions suggests the existence of steady-state ascending fluid flow and enable its composition and evolutionary patterns to be revealed. At the onset of evolution the fluid appears to be dominanted by reduced compouds with prevailing methane and other hydrocarbons as compared to hydrogen. Oxidation of fluid takes place due to reactions with the rocks undergoing metamorphism. Simultaneous enrichment in water in the upper, lower- temperature, portion of the fluid flow, and carbon dioxide in the high temperature part, occur its leveling-out along the flow. The absence of this effect would preclude a passage to granulite facies, and thermostatic control of system would develop under amphibolite facies conditions. It appears that the proposed "hydrogen model" of a steady-state fluid flow could match revealed empirical patterns of fluid regimes in metamorphism and ultrametamophism.

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