

Accessory mineral alteration in peraluminous granites at the hydrothermal stage: a review

MICHEL CATHELINÉAU

CREGU and G.S. CNRS-CREGU, B.P. 23, 54501, Vandoeuvre-les-Nancy, France

ABSTRACT. — U, Th and REE distribution in peraluminous granites is essentially controlled by accessory minerals such as monazite, uraninite, apatite and, to a lesser degree, zircon. Although the nature and distribution of such minerals are controlled by magma features, they may be significantly affected by subsolidus alteration. Thus, different examples of hydrothermal alteration have shown that trace elements may exhibit contrasted behaviour, due to differential stability of primary accessory minerals (especially uraninite and monazite) and their replacement by authigenic phases (complex solid solutions, carbonates and phosphates). Crystallization of authigenic minerals having higher solubilities than the magmatic accessory minerals is extremely important for further leaching of trace elements by hydrothermal fluids. Such alteration of magmatic accessory minerals in granites and dissolution-recrystallization processes depend on: 1) factors controlling fluid percolation through the rock (water/rock ratio, degree of microfracturing, etc), 2) nature of the accessory minerals and especially the solubility of the minerals of magmatic origin under hydrothermal conditions (dissolution-alteration stage), 3) fluid composition and changes occurring in the physical-chemical conditions during the later deposition stage.

Key words: Accessory minerals, U, Th, REE, Hydrothermal alteration, Peraluminous granite.

RESUME. — La distribution et les teneurs de U, Th et des terres rares dans les granites peralumineux est essentiellement contrôlée par les minéraux accessoires, tels que la monazite, l'uraninite, l'apatite et, à un moindre degré, le zircon. Bien que la nature et la distribution de tels minéraux accessoires soit d'abord contrôlée par les caractéristiques du magma, elles peuvent être affectées de manière significative par les altérations subsolidus. Ainsi, différents exemples d'altération hydrothermale ont montré que les éléments en traces peuvent avoir un comportement très contrasté, en fonction de la stabilité différentielle de leurs porteurs

(notamment l'uraninite et la monazite) et de leur remplacement par des minéraux accessoires neoformés (solutions solides complexes, carbonates, phosphates). La cristallisation de minéraux néoformés, présentant des solubilités plus importantes que les minéraux accessoires d'origine magmatique, est très importante pour le lessivage ultérieur des éléments traces par des fluides hydrothermaux. De telles altérations ou néoformations de minéraux accessoires dépendent: 1) des facteurs contrôlant la percolation des fluides à travers les roches (rapport eau/roche, degré de microfracturation,...); 2) la nature des minéraux accessoires, et en particulier la solubilité des minéraux accessoires d'origine magmatique en condition hydrothermale (stade de dissolution-altération); 3) la composition des fluides et les changements intervenant dans les conditions physico-chimiques lors de stades ultérieurs de dépôt.

Mots clés: Minéraux accessoires, U Th, Terres rares, altération hydrothermale, leucogranite peralumineux.

Introduction

The distribution of U, Th and REE in peraluminous granites is firstly controlled by the magmatic stage. These trace elements, as well as Zr, P, etc., are not incorporated in the main rock-forming minerals because of the low partition coefficients between the magma and minerals such as quartz, feldspars or phyllosilicates, but in accessory minerals such as uraninite, monazite, zircon or apatite. Consequently, their content in the whole rock is firstly dependant on the factors which control the distribution and nature of the accessory minerals. Thus, knowledge of the mineralogy and thermodynamical properties of accessory minerals, which may bear more

than 80% of the above mentioned trace elements, is the key to understand the geochemical features of granites.

For many years, most of the accessory minerals, except uraninite, have been considered as fairly insoluble and metastable under hydrothermal and supergene conditions. This assumption led to consider first the whole rock contents in U-Th-REE as relatively good tracers of the magmatic processes, and second Th and REE as relatively immobile in granites, except in severe hydrothermal alterations. Thus, uraninite abundance has been mainly investigated as a tool for the recognition of potential source rocks for uranium. Consequently, peraluminous granites which may be rich in uraninite, were considered as favourable environments for hydrothermal leaching and deposition of uranium (RANCHIN, 1971; MOREAU, 1977; FRIEDRICH et al., 1987, among others).

In recent years, new data have been obtained on the stability of monazite and zircon during metasomatic, hydrothermal (CATHELINÉAU, 1987a, b) and ductile structuring processes (GUINEBERTEAU, 1986). Such works have demonstrated that under specific conditions, accessory minerals may be altered and U, Th, REE leached. Thus, the purpose of this paper is to investigate the role of hydrothermal fluids as factors controlling the alteration of accessory minerals, and the consequent changes on the trace element geochemistry of granites.

I. Distribution of U, Th, REE among minerals

At the stage of granite crystallization, accessory minerals have two origins: crystallization from the melt, and mechanical transport by the magma of accessory minerals of external origin, as those inherited in relatively large amount from the partial melting of metamorphic rocks. Thus, the inheritance of zircon (POLDERVAART and ECKERMANN, 1958; HOPPE, 1965) and monazite (MILLER and MITTELFELD, 1982; SAWKA et al., 1986) is relatively common since the solubility of such minerals in magmas is relatively low. The nature, abundance and

distribution of accessory minerals crystallizing from the melt depend on four groups of parameters (PAGEL, 1981, 1982a, b; CUNNEY et al., 1979; CUNNEY and FRIEDRICH, 1987; CATHELINÉAU, 1987c): the trace element contents (1) and chemical features (2) of the magma, (3) the degree of magma evolution, (4) the physical-chemical conditions of magma crystallization. Figure 1 represents the factors controlling parageneses at the magma stage, and a possible sequence of crystallization.

The distribution of U, Th, and REE does not depend only on the nature and abundance of each accessory mineral acquired at this stage, but also on the trace element distribution among each mineral. The U-content in monazite or thorite and the Th-content in uraninite, for instance, may be highly variable according to magma chemistry or magma evolution. Thus, as a given trace element is beared by several minerals, detailed mineralogical studies are required to establish the complete distribution of its content in the rock. Figure 2 gives a schematic distribution of La and U among the different rock forming minerals in U-rich peraluminous granite. It shows that uraninite and monazite are major constraints for U and La contents respectively in the rock, but that the U content in monazite and zircon, or the La content in phyllosilicates and apatite cannot be ignored in the mass balance.

II. Changes in accessory mineralogy at the hydrothermal stage

The alteration of accessory minerals in granites during subsolidus fluid-rock interactions depends on (CATHELINÉAU, 1987c): «1) the nature of the mineral, and especially the solubility of the mineral (S_{ds}) under the dissolution conditions, 2) the solubility (S_f) of this mineral in the fluid which is a function of the initial concentration of the elements such as U, Th, REE and ligands in the percolating fluid, 3) the water/rock ratio and the flow regime in the granite, 4) the exchange and interaction surface between the fluid and the minerals, which depends on the degree of microfracturing and amount of microcracks

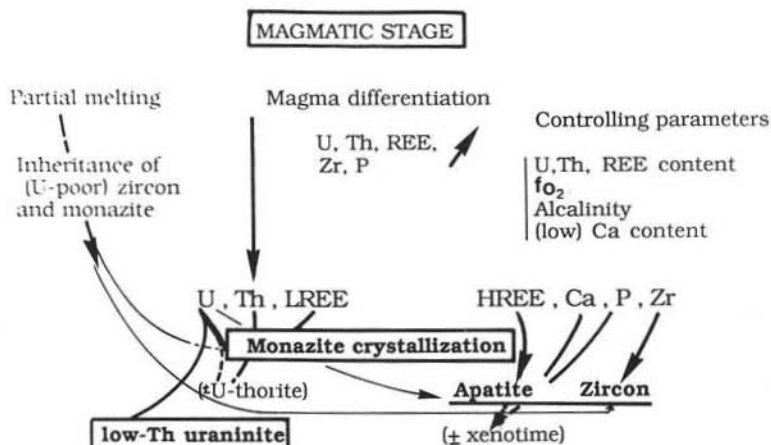


Fig. 1. — Main accessory minerals crystallizing at the magmatic stage in peraluminous granites (following data from literature and especially PAGEL (1981, 1982a) and CUNNEY and FRIEDRICH (1987)).

in minerals. The higher the solubility difference ($S_f - S_{ds}$), the larger the amount of mineral which can be dissolved. Therefore, the most favourable conditions for the dissolution of a given accessory phase may be predicted if the parameters constraining the highest ($S_f - S_{ds}$) values are known.

The two last factors (4 and 5) have an extreme influence on the rate of alteration of minerals. As the accessory minerals are small-sized, scattered in granite minerals (feldspar, biotite, quartz, etc.), and represent a very small volume of the rock, a high degree of microfaulting is required to allow fluids to interact with them. This condition is frequently obtained in peraluminous granites from the Variscan range, which are spatially associated with major shear zones. Thus, demonstrative examples of monazite or zircon dissolution have been described in granites affected strongly by ductile to brittle deformation (northern border of the Mortagne batholith, Vendée, France, CATHELIN, 1982; GUINEBERTEAU, 1986), or in granites affected by subsolidus alteration such as the quartz dissolution process (CATHELIN, 1986) which generally occurs in granites affected by strong microfracturing (PECHER et al., 1985).

Two different cases may be distinguished among the disturbances occurring in accessory minerals of magmatic origin: 1) partial and

differential in-situ alteration of minerals, 2) crystallization of authigenic minerals in other sites (microcracks, vugs, etc.) after moderate to important transport of elements in solution.

1) In-situ alteration of accessory minerals

Accessory minerals of magmatic origin may be partially to entirely altered and replaced by authigenic minerals. Each accessory mineral has a specific behaviour under given physical and chemical conditions, and will transform into specific alteration products, as shown by Figures 3 and 4. In this case, changes in the trace element content of the rock is described by the mass balance of the chemical reactions of solubilization or alteration of the early minerals. The most representative reactions for carbonates and phosphates are given in Table 1.

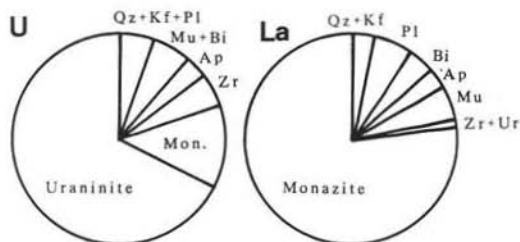


Fig. 2. — Repartition of U and La among main rock forming minerals of peraluminous granites.

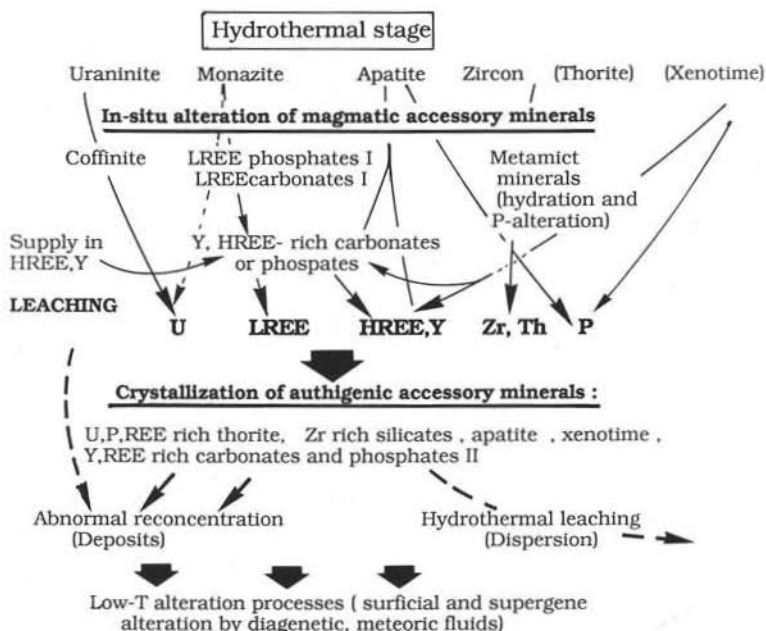


Fig. 3. — Schematic representation of main stages of alteration and crystallization of accessory minerals, and stages of U, Th, REE, P, Zr mobility at the hydrothermal stage.

Uraninite is frequently lacking in surface samples due to supergene alteration, thus yielding significant U losses (BARBIER and RANCHIN, 1969; FRIEDRICH et al., 1987). Uraninite is observed either as a fresh mineral or as a boxwork, giving evidence of complete alteration of the mineral and replacement by phyllosilicates and iron (hydr-) oxides. It is difficult to interpret whether such an alteration results from a hydrothermal or a supergene process on the basis of microscopic observations. This explains why uraninite behaviour under hydrothermal conditions is relatively badly documented and subject to discussion. In relatively infrequent cases, some alteration products have been identified, such as coffinite or hydrous oxides which could be interpreted as hydrothermal phases. However most of them are U(VI) secondary minerals such as phosphates or sulfates (LE and STUSSI,

1973; RIMSAITE, 1982a, b...) typical of the oxidizing zone.

Monazite may be partially (corrosions) to entirely dissolved, or affected by dissolution - recrystallization processes. In the case of the Pierres Plantées alteration pipe (CATHELINEAU, 1986, 1987a; RESPAUT, 1984; RESPAUT et al., 1988), monazite recrystallization caused complete resetting of the U-Pb system, at around 30 Ma after granite crystallization, and a partial LREE loss in the whole rock. Monazite alteration and replacement by REE-carbonates is also a rather common process in granites (RIMSAITE, 1982a, b; MARUEJOL and CUNEY, 1985) submitted to strong alteration processes such as quartz dissolution and albitization, or intensive biotite chloritization. REE-carbonates are parsite or HREE-rich parsite and occur either as microcrystalline or needle

Fig. 4. — Microphotographs of accessory minerals from peraluminous granites (+, or // refer to a microphotograph taken with crossed or uncrossed polars respectively): 1 - uraninite with aureole filled with phyllosilicates (+). 2 - uraninite boxwork filled with phyllosilicates and iron oxides (+). 3 - monazite crystal (/). 4 - parsite crystals replacing a monazite (+). 5 - apatite crystals within a biotite (+). 6 - hydrothermal apatite crystals in a cavity resulting from quartz dissolution (/). 7 - zirconium rich silicates (zrsil) surrounding magmatic zircon cores (+). 8 - hydrothermal (P, U, REE, Y) rich thorite (+).

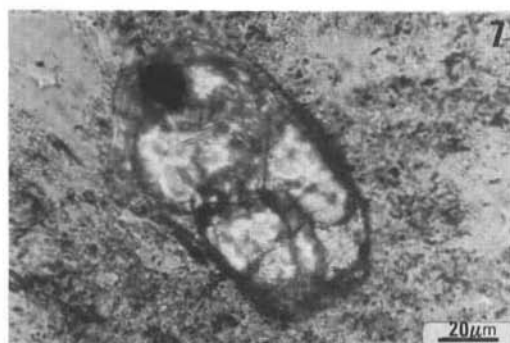
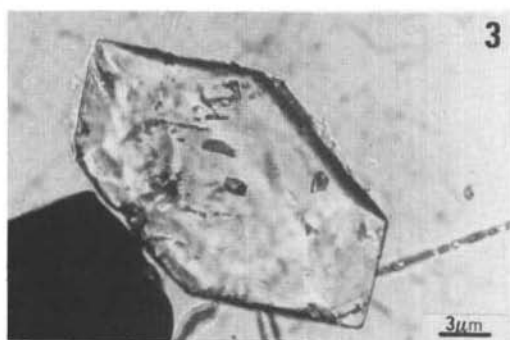


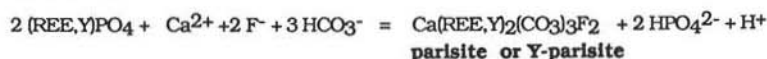
TABLE 1

Possible dissolution and alteration reactions of apatite, monazite and xenotime (choice of species in solution was made from consideration of most probable conditions during studied processes)

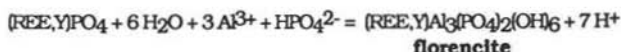
MONAZITE, XENOTIME



monazite
or xenotime

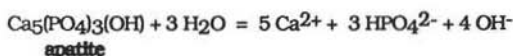


parisite or Y-parisite

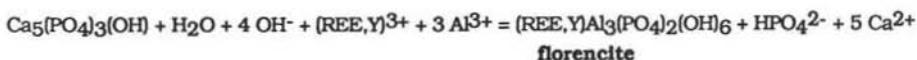


florencite

APATITE



apatite



florencite

masses or small prismatic crystals within microcracks or around monazite crystals.

Zircon, such as monazite, may be corroded up to complete dissolution (CATHELINEAU, 1987b), or affected by metamict disintegration (RANKAMA and SAHAMA, 1950; ANDERSON, 1941; CARUBA et al., 1975) consisting in progressive hydration and destruction of the crystalline network, probably due to the effects of self-radioactive damage. In some cases, a fringe of zirconium rich silicates, considered as resulting of hydrothermal overgrowth, surrounds a previous zircon core. Zirconium rich silicates were observed in albitized and quartz-depleted granites and are especially enriched in Th, P, REE, U, Al and Ca (CATHELINEAU, 1987b).

Apatite may be completely dissolved and then redeposited in albitization processes accompanied by quartz dissolution. Authigenic apatites may be very abundant in such altered rocks as disseminated prisms or sphaerulites made of apatite needles, this producing P enrichments up to 1 or 2%, which are abnormal values for peraluminous granites. In other cases, apatite may be replaced by florencite (SAWKA et al., 1986) or rhabdophane (VLASOV, 1966) during alteration process the origin of which

(hydrothermal, or supergene, BANFIELD, 1985), is questionable.

Anatase is the most frequent Ti-bearing phase even in apparently fresh granites, probably due to the instability of ilmenite and titanomagnetite, and to the lack of titanite crystallization in Ca-poor peraluminous granites. Ti is also expelled from biotite during chloritization, and is mobilized in some cases such as albitization- quartz dissolution processes, during which new anatase crystallizes in abundance in the vugs of quartz-depleted granites.

2) Authigenic minerals (discrete to significant mineralizations)

Numerous descriptions in the literature mention the presence of accessory minerals in sites which undoubtedly attest to the subsolidus crystallization of such minerals. Most of these authigenic phases are the same minerals than those replacing accessory minerals of magmatic origin, but most cannot be identified positively as products of the in-situ alteration of a previous phase. Such data mean that significant transfer of U, Th, REE or P may occur through the partial to complete dissolution of one to several accessory

minerals, transport of elements by a solution and precipitation of authigenic minerals in a site which may be relatively far from the dissolution zone. In this case, the approach to the problem is similar to that of any study of hydrothermal or metallogenic processes. Therefore, the amount of authigenic accessory mineral, which can be precipitated from a fluid as a result of a change in the physical-chemical conditions, depends on the concentration of trace elements under the transport condition, and the solubility difference ($S_t - S_{dp}$): $S_t - S_{dp}$ being the difference between the solubilities of the authigenic mineral under transport and deposition conditions respectively.

Authigenic minerals are mostly complex solid solutions, which may be either silicates (zirconium rich silicates, U-REE-Ca-Y rich thorite,...), phosphate (U-Th rich xenotime, crandallite group, etc.) or carbonates (LREE to HREE-Y rich parisite). The microphotographs of Figure 4 give some aspects of these minerals in thin section.

Major differences characterize the in-situ alteration and new crystallization of accessory minerals: 1) the water/rock ratio and 2) the amount of authigenic mineral which may precipitate, which are much higher in the second case, 3) the time necessary for the achievement of the process which is higher in the case of pervasive in-situ alteration than in typical channelized hydrothermal circulation, 4) the degree of disturbance of magmatic paragenesis, which remains relatively low in the second case, due to low reactivity of accessory phases in the host rocks during the deposition of hydrothermal minerals in veins or microcracks.

Abnormal enrichment in U, Th and REE have been observed for instance in albitized and quartz-depleted, granites from the Mt Lozere area (Bouges peraluminous granite, southern part of French Central Massif, CATHELINÉAU, 1987b). In such altered rocks, the abundance of apatite, phosphothorite, and xenotime is such that element transfer must have occurred along important distances, and that large volumes of the source rock have been altered. This suggests that significant amounts of U, Th of REE may be transported

and redistributed within a granitic pluton under rates which are essentially dependent on the physical-chemical conditions prevailing in the rocks.

Discussion and conclusions

Peraluminous granites, as well as most of granites, are affected by long-lived fluid circulation, which are for the most part later than the magmatic stage and independent of granite cooling (CATHELINÉAU, 1987a; CATHELINÉAU et al., 1988). It is clear that, throughout the hydrothermal history of a granitic body, a significant contribution of hydrothermal process to the present-day geochemical features must be considered not only as a possibility but as a reality. Considering the U, Th, REE content of granites, the rate of changes in the geochemical features acquired at the magmatic stage is a function of:

- 1) the degree of transformation of magmatic accessory minerals in the case of pervasive alteration (such as albitization, or chloritization), and the nature of the alteration products. In this case, the system may be considered as semi-closed, and changes in the mineralogy do not necessarily imply significant U, Th or REE transfer, the mass transfer being controlled by the mass balance relative to each alteration reaction. However, the crystallization of carbonates or phosphates such as parisite or crandallites in place of monazite or apatite is extremely important for further late processes, since such minerals have significantly higher solubilities than the accessory minerals from which they derived. As a consequence, REE may be largely mobilized during stages of low- to medium-range temperature, during which authigenic minerals may be easily leached whilst early accessory minerals remain stable;

- 2) the nature and amount of authigenic minerals which may crystallize in the granite from the fluid phase. In this case, the supply in trace elements from the fluid disturbs the early geochemical features by a superimposition of the different element contents relative to the crystallization of each newly-formed mineral.

Alteration of a specific accessory mineral whatever its origin, magmatic or hydrothermal, probably controls the trace element of the fluid percolating in the granites, independently of the effects of fractionation process which may occur during mineral dissolution. Therefore, the REE pattern of the common hydrothermal mineral (fluorite, carbonates) which reflects the fluid pattern may also be strongly influenced by the nature of the accessory minerals which were leached in the source rocks. Thus, the instability of monazite and LREE-rich carbonates or phosphates may explain the LREE enrichment of pitchblende, calcite or fluorite observed in deposits located in peraluminous granites (CATHELINÉAU, 1987c). Similar conclusions have been proposed by PAGEL et al. (1987) from the study of uranium oxides of different geologic environments.

It is worth noting that fluorine, phosphate and to a lesser degree carbonate activities exert considerable control over all the alteration reactions involving REE (Table 1), and that REE speciation at high temperature remains one of the (relatively badly known) key for a correct understanding of the processes.

In the light of such data, careful mineralogical observations appear to be a necessary prerequisite to any interpretation of trace element geochemistry. Such study could ensure that the present-day geochemical features of granites were acquired at the magmatic, hydrothermal or supergene stages and may especially justify the use of purely magmatic models before any attempt at chemical modelling.

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