

GRANITE-RELATED RARE-METAL MINERALIZATION: A GENERAL GEOCHEMICAL FRAMEWORK

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ABSTRACT: The magmatic evolution of rare-metal mineralized granitic intrusions during intracrustal fractionation is controlled by fractional crystallization. Ti-Ta distribution patterns with negative linear correlation in log-log space for sample suites from several granite provinces (western Europe, SE Asia, Bolivia) define extended magmatic Ta enrichment trends to a degree of fractionation $F \leq 0.01$, corresponding to ≤ 1 wt. % of residual liquid in a perfect fractionation model. Such a high degree of fractionation is required for the generation of primary Ta-Nb mineralization, due to the relative immobility of Ta and Nb during postmagmatic fluid overprint. In contrast, the hydrothermal mobility of Sn, W, Mo, and U allows hydrothermal redistribution of these elements in magmatically enriched granitic systems at a lower degree of magmatic fractionation. Scatter distributions with bulk deficiencies of these metals point to large-scale hydrothermal element depletion as a complementary feature of metal enrichment in small-scale physical domains, i.e. in ore deposits. Diverse metal mobility with oxidation state as an important controlling parameter causes the distinct individualization in the general Sn-W-Mo-U ore deposit spectrum according to specific redox environments. The hydrothermal solubility of uranium allows low-temperature redistribution totally disconnected in time and displaced in space from the magmatic uranium enrichment event. In contrast, hydrothermal Sn-W-Mo systems are set up within the time interval of a few Ma from multiphase granite suites and form ore deposits close to their igneous metal source.

GLOBAL GEOCHEMICAL BACKGROUND

Metal enrichment to ore grade is the ultimate outgrowth of efficient large-scale and long-term fractionation processes in a global context (Brimhall, 1987). The geochemical evolution of some metals during the Earth's history is schematically depicted in Fig. 1 and shows some trends of general importance. The earliest history of the Earth is characterized by core segregation and ongoing accretion which result in an element distribution pattern in the primitive mantle which is controlled by volatility (vapour-solid fractionation) and degree of siderophile behaviour (metal-silicate fractionation). The volatility of tin (50% condensation temperature at 10^{-4} bar total pressure: 720 K) is distinctly different from the volatility of the ore-paragenetically related elements W, Mo or Ta (50% condensation temperatures at 10^{-4} bar: $>1,400$ K). This results in a drastic decrease of tin content from ca. 1.8 ppm Sn in condensed solar material, as seen in C1 chondrites, to approximately 0.4 ppm in bulk Earth and 0.13 ppm in primitive mantle (moderately siderophile behaviour of tin). The refractory and moderately siderophile elements W and Mo partition also preferentially into the core, which leaves the primitive mantle depleted in

these elements when compared to solar composition. However, as deduced from the abundances of siderophile elements in the mantle, general chemical equilibrium between the Earth's mantle and largely metallic core is possibly not established, which is the reason for a large error margin for all bulk Earth chemical models. The refractory and lithophile Ta is affected neither by volatile depletion (such as Sn) nor by substantial partitioning into the core (such as Au, W and Mo), and provides an example of a continuous enrichment trend.

Archaean crust formation by crystal-liquid fractionation of primitive mantle material and dominantly intracrustal fractionation in the Phanerozoic led to the present-day element pattern of the continental crust. It is interesting to note that long-term geochemical fractionation restores in the Earth's crust approximately the Sn and Mo levels which were present in the condensed solar starting material before the onset of cosmochemical fractionation. This is different from the evolution pattern of Au, which is depleted in the continental crust relative to C1 chondrites by about two orders of magnitude, and of W and Ta, which are enriched by one and two orders of magnitude, respectively (Fig. 1).

Granite magmatism is the quantitatively dominating intracrustal fractionation mechanism. The enrich-

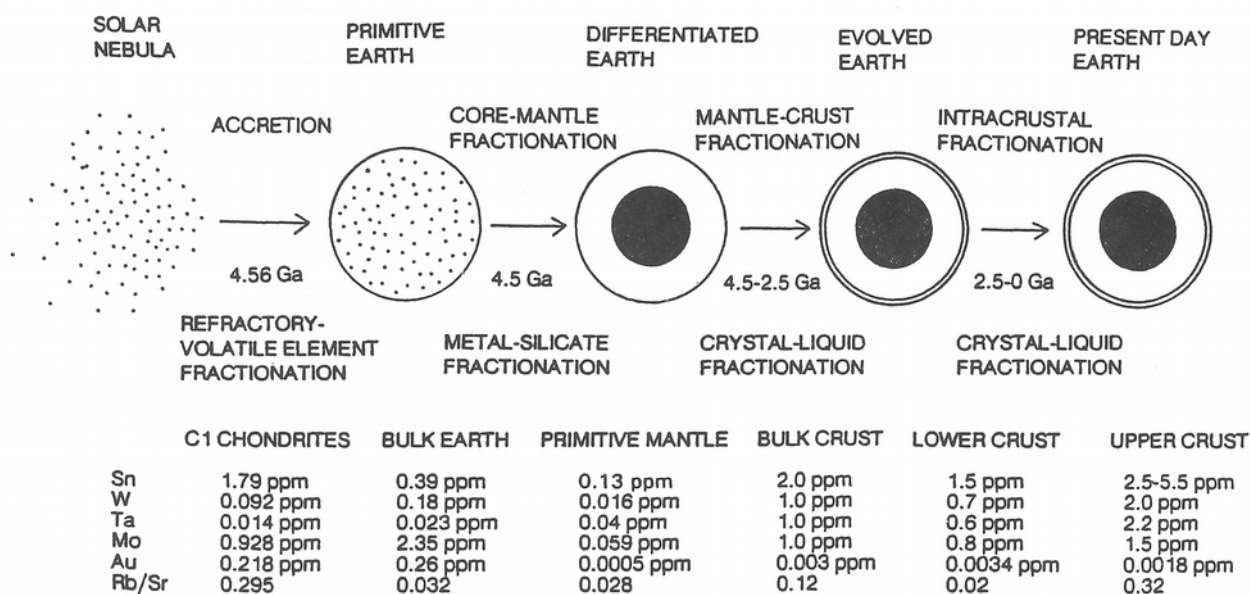


Fig. 1. Fractionation pattern of some metals during the Earth's history. C1 data are from Anders and Grevesse (1989) and Loss et al. (1989). The bulk Earth data are from Morgan and Anders (1980) and are calculated assuming chemical equilibrium between mantle and core. Primitive mantle data are from Anderson (1983), Wänke et al. (1984), Taylor and McLennan (1985), and Jochum et al. (1993). Data on continental crust are from Taylor and McLennan (1985).

ment of the elements Sn, W, Ta and Mo in the upper crust, compared to the lower crust, indicates the incompatible behaviour of these elements during partial melting of crustal material. The enrichment factor of 2-3 (upper crust/lower crust) corresponds to the mass balance constraint given by the mass ratio of bulk crust/upper crust.

MAGMATIC EVOLUTION

The element suite of Ta-Sn-W-Mo-U-Be-Li-Rb-Cs (commonly referred to as "rare metals" in economic geology) is geochemically characterized by generally incompatible behaviour during partial melting which results in enrichment in the most fractionated portions of the Earth's crust, i.e. in granitic (or rhyolitic) rocks. The magmatic evolution of granitic rocks is controlled by assimilation of wall rocks, restite unmixing, and a variety of little-understood internal fluid dynamical processes such as convective fractionation or sidewall fractional crystallization. However, without defining the exact physical nature of the igneous fractionation processes, the geochemical patterns of high-level granitic intrusions in association with rare-metal mineralization indicate evolution trends which essentially follow the fractional crystallization law (Lehmann, 1990).

Examples for this situation are given in Figs. 2-4 which are log-log plots of whole-rock Ta versus Ti content in granitic samples from the Bolivian Sn-W province, the central Thailand Sn-W-Ta belt, and some European Sn-W-Ta-bearing granitic rocks (northern Massif Central/France, Mittersill/Austria, Erzgebirge). The major granite units of the Erzgebirge Sn-W-U province are plotted as a reference suite in each diagram, because granite magmatism in this metal province has been studied in detail resulting in a good geochemical data base (Tischendorf, 1989). The Erzgebirge data points represent averages for major granite units from a multiphase Hercynian intrusion sequence, in which the systematic increase in Ta and decrease in Ti reflect successively younger and volumetrically smaller intrusion phases.

Tantalum and titanium are used as indicators of degree of fractionation because these two elements are relatively immobile during hydrothermal alteration of granitic rocks, and because both elements have a complementary geochemical behaviour, i.e. Ta is incompatible in granitic systems whereas Ti has a compatible behaviour. The negative linear correlation in log-log space of Figs. 2-4 suggests a magmatic evolution for these rock suites dominantly controlled by fractional crystallization. The most evolved rocks in each diagram characterize rare-metal mineralized alkali-feldspar aplogranite stocks, such as the Chojlla granite in Bolivia

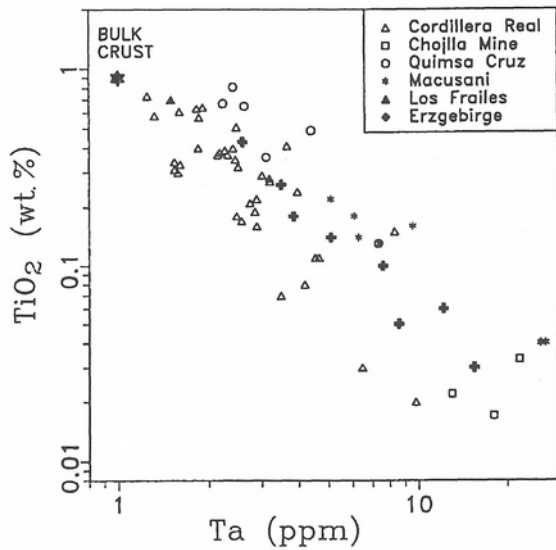


Fig. 2. Ta-TiO₂ variation diagram for granitic and rhyolitic rock samples from the Bolivian tin province (data from Lehmann et al., 1990; Lehmann, unpubl.; Noble et al., 1988, Pichavant et al., 1988). Reference data for the Erzgebirge granite suite are from Tischendorf (1989) and represent arithmetic means of a large sample population. Bulk crust composition from Taylor and McLennan (1985)

(tin-tungsten mine), the Altenberg granite stock (tin porphyry), the Beauvoir granite stock (tantalum-tin deposit, currently mined for kaolinite), and the Pilok/Thailand and Hermyingyi/Burma granite stocks (tin-tungsten mining).

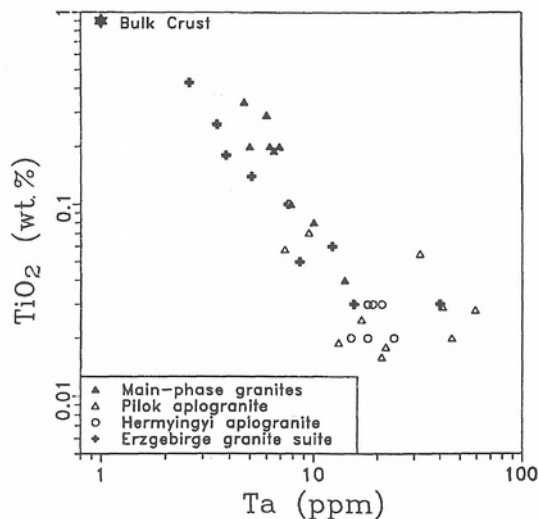


Fig. 3. Ta-TiO₂ variation diagram for granitic rock samples from the central Thailand/Burma border range (data from Cobbing et al., 1988; Lehmann, unpubl.), and the Erzgebirge reference granite suite (Tischendorf, 1989). Bulk crust composition from Taylor and McLennan (1985).

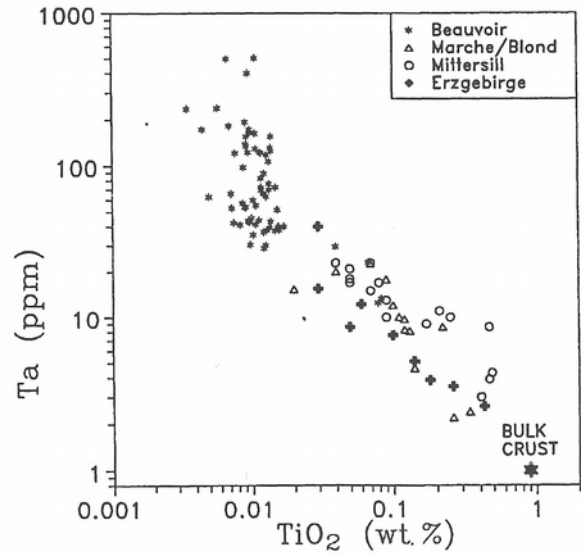


Fig. 4. TiO₂-Ta variation diagram for granitic rock samples from the northern Massif Central in France (data from Cuney and Raimbault, 1991; Raimbault et al., 1988), the K1 gneiss of the Mittersill scheelite mine in Austria (Lehmann, unpubl.), and the Erzgebirge reference granite suite (Tischendorf, 1989). Bulk crust composition from Taylor and McLennan (1985).

The granitic rocks from each metal province do not constitute cogenetic fractionation suites, because they are from spatially and in part also temporally separated intrusions. However, the fact that various individual rock suites follow a coherent chemical trend indicates that the underlying principles of their magmatic evolution are similar. Given the basic process of fractional crystallization with variable degree of fractionation, the linear correlation trends in Figs. 2-4 are defined by the bulk distribution coefficients D of Ta and Ti and by the initial Ta and Ti contents in the melt system. This corresponds to the definition of the slope m and term b in a general linear equation of the form $yD_{Ta-1} = mx + b$ with $m = D_{Ta-1}/D_{Ti-1}$ and $b = \log[Ta]_0/\log[Ti]_0$ (Lehmann, 1990). The identical position of Ta-Ti correlation trends from various areas suggests similar bulk distribution coefficients and similar initial Ti and Ta contents in the different melt systems. The last statement corresponds to the situation in Figs. 2-4 in which the extrapolated fractionation lines pass through bulk crustal composition.

A further argument for the importance of fractional crystallization in the igneous evolution of the rock suites discussed is the multiphase nature of the intrusion systems and their systematic enrichment in incompatible trace elements, and complementary depletion in compatible components towards the youngest and volu-

metrically smallest phases. This situation is well documented for the granite suites from the Erzgebirge (Tischendorf et al., 1987), Massif Central (Aubert, 1966), and SE Asia (Cobbing et al., 1986), and points to a crystallization process from the plutonic margins inwards and episodic tapping of an increasingly fractionated and buoyant magma chamber below the gradually deepening solidification front.

HYDROTHERMAL REDISTRIBUTION

The magmatic chemical inventory of high-level granitic rocks is invariably affected both by residual aqueous fluids of magmatic origin, and by ingressing meteoric and formation water. The metasomatic effect of hydrothermal overprinting on trace-element patterns in rocks depends on the reaction progress of fluid-rock interaction. Tantalum and, to a lesser degree also titanium, are relatively immobile in most granite-related hydrothermal systems, which is why the whole-rock abundances of these two elements can be used as an approximative indicator of magmatic fractionation of a given granite sample (Figs. 5-11). Titanium is used for those sample suites for which little or no Ta data are available.

Distribution patterns of Sn as a function of Ta or Ti are shown in Figs. 5-7. These patterns reflect both a magmatic tin enrichment trend and hydrothermal redistribution of variable degree. Relatively little tin redistribution is indicated for the Erzgebirge and Massif Central suites (Fig. 5) which give good linear correlation trends in log-log space, suggestive of element control dominantly by magmatic processes, i.e. fractional crystallization, as also deduced from the Ta-Ti plot of Fig. 2. It should be noted, however, that the invariable scatter in such logarithmic plots allows the identification of hydrothermal effects only when they are very pronounced with a more than about 100% change in element content. Compared to the Erzgebirge and Massif Central suites, there is a distinct increase in degree of scatter for the Bolivian (Fig. 6) and the Thailand/Burma rock suites (Fig. 7). The Thailand/Burma example gives a pronounced scatter for the most fractionated sample populations from the tin-tungsten mineralized Pilok and Hermyingyi aplogranite stocks, with the additional feature of a general tin deficiency when compared to the more or less magmatic tin enrichment trend of the Erzgebirge reference rock suite. This large-scale tin deficiency characterizes several of the SE Asian tin granite systems and is seen as a complementary feature of small-scale tin-ore deposition in the same system (Lehmann and Mahawat, 1989). The reason for the very pronounced tin redistribution patterns in SE Asian granite systems is probably their low oxidation state (Lehmann and Harmanto, 1990). This allows very

efficient leaching and hydrothermal transport of tin, because the strongly redox-dependent solubility of cassiterite is greatest in reducing environments (Eugster, 1986).

Corresponding distribution patterns for tungsten are shown in Figs. 8-9. The Erzgebirge reference suite indicates systematic tungsten enrichment towards the most evolved granite units. The Thailand/Burma sample suite displays strong scatter with a trend of increasing W contents in most fractionated rock portions. The position of the Beauvoir aplogranite stock (Massif Central) in Fig. 9 corresponds to the very high degree of fractionation of this sample suite and suggests only minor hydrothermal redistribution. This is opposite to the situation for granitic samples from the Mittersill scheelite mine in Austria. These rocks, the so-called K1-gneisses, give the typical pattern of extreme hydrothermal overprinting, i.e. depletion/enrichment of tungsten which was in this particular environment highly mobile, whereas the immobile Ta defines the high degree of fractionation of these rocks.

The behaviour of uranium is shown in Figs. 10-11. The examples from the Erzgebirge, Massif Central, and Thailand/Burma granite suites give scatter distributions with generally elevated uranium levels. There is a trend of increasing uranium contents with increasing degree of fractionation in the SE Asian rocks, which continues into unsystematic patterns with relatively constant U contents in the 10-60 ppm range in most fractionated rock portions. The same pattern is shown by the sample population from the Mittersill scheelite mine. This can be a result of a change in the bulk distribution coefficient of U during the magmatic evolution (for example by monazite crystallization) or of hydrothermal overprint. By contrast, the highly fractionated rocks from the Erzgebirge and Massif Central have relatively low U abundances in the 5-20 ppm U range (which is still 2-8 times the upper crustal average). This feature, together with their strong uranium scatter distribution, and the fact that these two granite provinces host the major uranium deposits of Europe, suggests a process of postmagmatic leaching of U in these rocks.

METALLOGENIC MODEL

The granitic phases/subintrusions in spatial, temporal, and chemical relationship to rare-metal ore deposits are highly fractionated. Systematic trace-element distribution patterns in these subunits and in associated larger granite systems point to fractional crystallization as the dominant petrogenetic process controlling magmatic evolution.

Degree of fractionation and oxidation state are the two parameters which control magmatic rare-metal en-

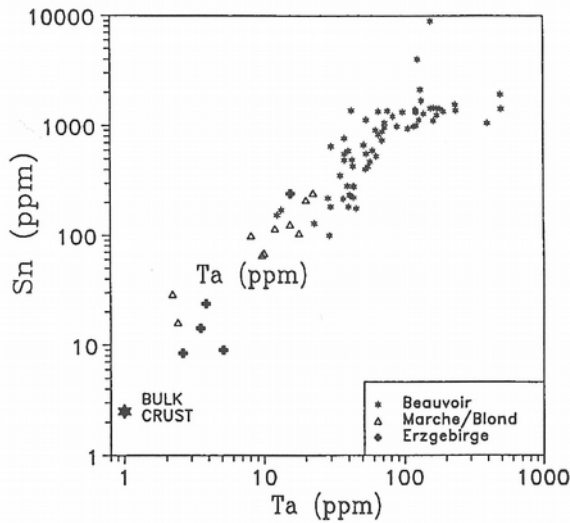


Fig. 5. Ta-Sn variation diagram for granitic rock samples from the northern Massif Central in France and the Erzgebirge granite reference suite. (For data sources see Fig. 4.)

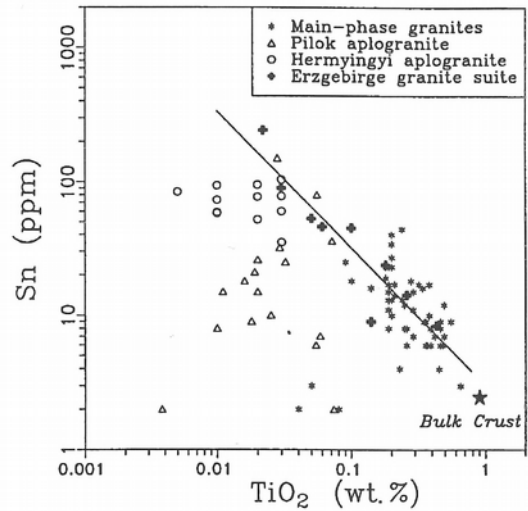


Fig. 7. TiO_2 -Sn variation diagram for granitic rock samples from the central Thailand/Burma border range. The correlation line through the Erzgebirge granite suite data defines the general magmatic tin enrichment trend. (For data sources see Fig. 3.)

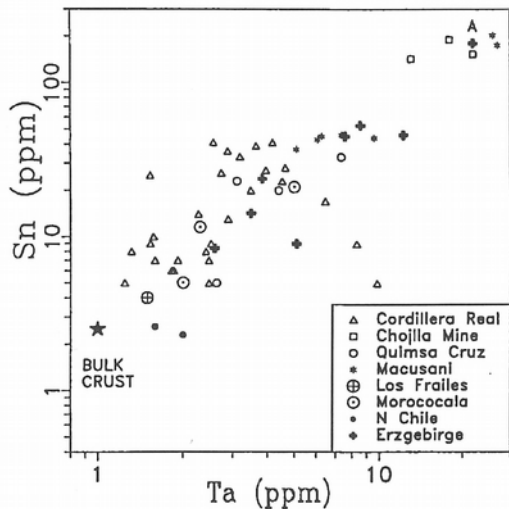


Fig. 6. Ta-Sn variation diagram for granitic and rhyolitic rock samples from the Bolivian tin province and its surroundings. (For data sources see Fig. 2.)

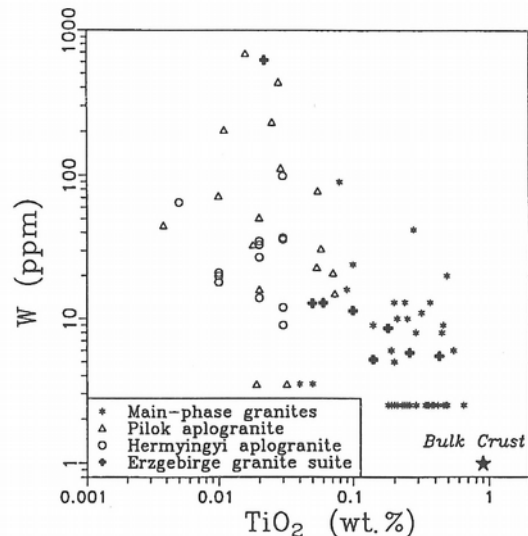


Fig. 8. TiO_2 -W variation diagram for granitic rock samples from the central Thailand/Burma border range and the Erzgebirge reference suite. (For data sources see Fig. 3.)

richment in granite suites. Systematic tin enrichment during magmatic fractionation is typical of ilmenite-series granitic rocks as opposed to a non-enrichment pattern in magnetite-series granitic rocks (Lehmann, 1990). This is opposite to the behaviour of molybdenum, and less pronounced of tungsten, which becomes more compatible with decreasing oxidation state of a melt (Tacker and Candela, 1987; Candela and Bouton,

1990). The behaviour of tantalum seems to be little or not affected by the oxidation state.

Postmagmatic hydrothermal redistribution is an essential feature for Sn, W, Mo and U mineralization, which is the reason why ore formation of these metals does not need the extreme degree of fractionation typical of Ta ore systems. The hydrothermal redistribution process is again dependent on the oxidation state of the

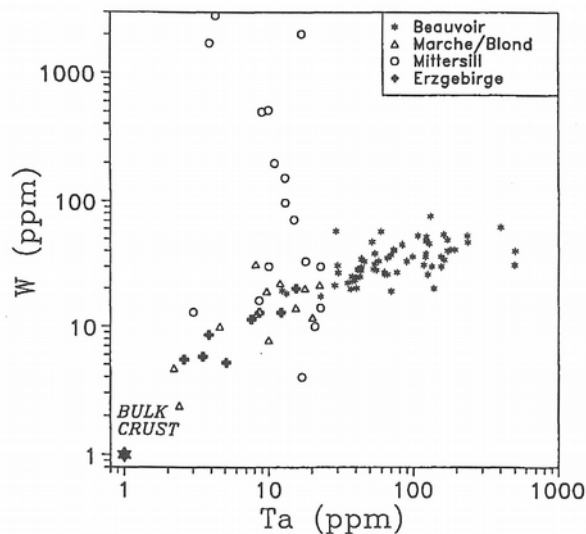


Fig. 9. Ta-W variation diagram for granitic rock samples from the northern Massif Central in France and from the K1-gneiss in the Mittersill scheelite mine in Austria. (For data sources see Fig. 4.)

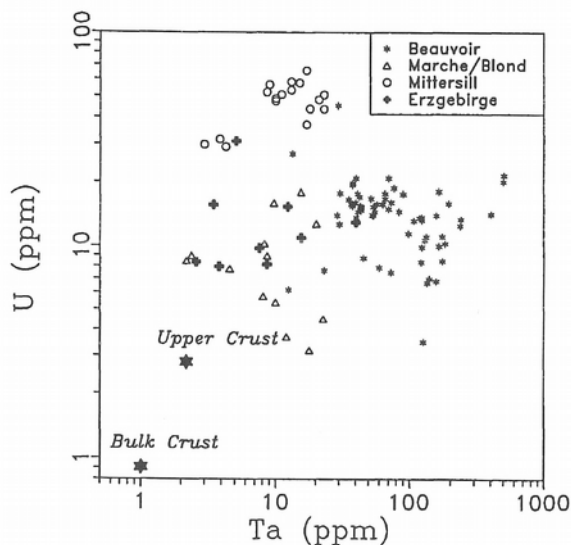


Fig. 10. Ta-U variation diagram for granitic rock samples from the northern Massif Central in France, from the Mittersill scheelite mine, and the Erzgebirge reference suite. (For data sources see Fig. 4.)

igneous system. For example, the hydrothermal solubility of tin is orders of magnitude greater under reduced conditions near the quartz-fayalite-magnetite buffer than under more oxidized conditions near the hematite-magnetite buffer (Eugster, 1986). Consequently, hydrothermal tin ore-forming environments have a low oxidation state. This situation is opposite to the behaviour of molybdenum which is mobilized in much more

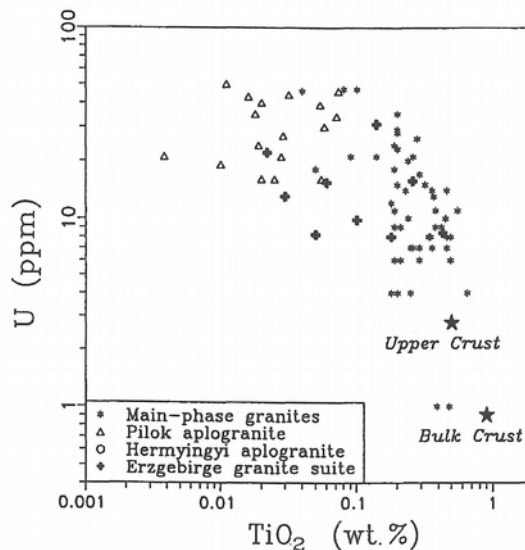


Fig. 11. TiO₂-U variation diagram for granitic rock samples from the central Thailand/Burma border range, and the Erzgebirge granite suite. (For data sources see Fig. 3.)

oxidized environments (Burnham and Ohmoto, 1980), or for gold which is transported preferentially in oxidized environments (Sillitoe, 1991). Tungsten appears to be less redox-sensitive and is redistributed in both reducing and oxidizing environments.

The high hydrothermal mobility of uranium in the U⁶⁺ valency state allows large-scale mobilization of uranium by oxidized fluids even at low temperature (Rich et al., 1977). Uranium ore formation is therefore not necessarily linked to the late magmatic hydrothermal evolution of fractionated granitic rocks, but often depends on structural reactivation which allows meteoric fluid circulation any time after granite emplacement. The multistage evolution of uranium mineralization has been well demonstrated in many parts of the world; the case of the Massif Central and Erzgebirge is discussed in Respaut et al. (1991), Turpin et al. (1990), and Lange et al. (1991). Multistage evolution is also a recurrent feature of many gold-bearing systems with similar redox-dependent solubility characteristics (Bonnemaison and Marcoux, 1990).

The establishment of rare-metal mineralization has two essential components: Magmatic metal enrichment through fractional crystallization and large-scale hydrothermal redistribution which acts upon magmatically pre-enriched rock volumes. Both processes have different importance for individual metals. The metallogeny of Ta is essentially controlled by the magmatic evolution, because of the low hydrothermal mobility of Ta. Tantalum ore deposits are therefore restricted to extremely fractionated rocks such as rare-metal pegmatites and alkalifeldspar granite stocks.

The importance of degree of magmatic fractionation is also well documented for the metallogeny of tin, with the additional feature that hydrothermal redistribution is an essential process for most tin ore deposits. Hydrothermal redistribution is an even more important ingredient in the formation of tungsten ore deposits, but the magmatic tungsten pre-enrichment stage can still be identified in many cases. The metallogeny of uranium is dominantly related to hydrothermal mobilization, often in no temporal relationship with magmatic events, and forms the transition towards the metallogeny of such components as gold and base metals for which fluid-rock interaction with a lesser degree of prior geochemical enrichment seems to be a characteristic feature. However, the existence of copper- and gold-rich porphyry deposits and their characteristic association with late phases of multiple intrusion sequences (Hollister, 1978; Titley and Beane, 1981), as well as the fact that base and precious metals often accompany tin-tungsten mineralisation, suggests even for these components some form of little understood magmatic enrichment.

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