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| **The diversity of relations between felsic magmatism and uranium deposits**GéoRessources, CNRS, CREGU, Université de Lorraine, BP 23954506, Vandoeuvre les NANCY Cedex, France**Michel Cuney** |

**Abstract.** The strongly incompatible behaviour of uranium in silicate magmas results in its concentration in the most felsic melts and a prevalence of granites and rhyolites as primary U sources for the formation of U deposits. Despite the incompatible behavior of uranium, U deposits directly related to magmatic processes are quite rare. Generally, U is mobilized by hydrothermal fluids or ground water well after the emplacement of the igneous rocks. Only a few granite or volcanic rock types, have U contents and physico-chemical properties that permit the crystallization of accessory minerals from which U can be leached for the formation of a large diversity of U deposits. Four types of granites or rhyolites can be sufficiently enriched in U to represent a significant source for the genesis of U deposits: peralkaline, high-K metaluminous calc-alkaline, L-type peraluminous, and anatectic pegmatoids. The evaluation of the potentiality for igneous rocks to represent an efficient U source represents a critical step during the early stages of exploration. A wider use of magmatic inclusions to determine parent magma chemistry and its U content is of utmost interest to evaluate the U source potential of sedimentary basins that contain a felsic volcanic acidic tuff contribution.

# INTRODUCTION

Uranium in silicate magmas exhibits a strongly incompatible behavior because of its large ionic radius and high valence which prevents its incorporation into the structure of the main rock forming silicates. As a result, during partial melting and crystal fractionation, U is preferentially fractionated into silicate melts. Such a behavior has several major geochemical, geophysical and metallogenic consequences: [i] through geological time U has been continuously transferred from the mantle to the Earth crust, and within the continental crust towards its upper part together with other incompatible elements and more particularly Th and K, [ii] consequently radiogenic heat production is maximized in the upper crust, and thus radiogenic heat flux production distribution may be used to delineate radioelement enriched crustal blocks, [iii] the most felsic melts tend to be the most enriched in U, and [iv] granites and rhyolites represent the primary sources of uranium for the formation of U deposits. However, despite, the strongly incompatible behavior of U, deposits dominantly resulting from magmatic processes are rare. In an average granitoid, with a U enrichment of 3 to 4 ppm, uranium is dominantly held within the crystal structure of accessory minerals [zircon, apatite, monazite, titanite, xenotime …] from which U cannot be leached by most geological fluids. Only some specific granites have higher U contents permitting the crystallization of other types of accessory minerals from which U can be more or less easily leached for the formation of U deposits. On this basis [1] was the first to define the notion of “fertile granites”, which has been very fruitful for U exploration.

# THE DIFFERENT TYPE OF URANIUM-RICH MAGMATIC ROCKS

The three main types of magmatic rocks which can be enriched in U above their Clarke values, are distinguished according to in the A/CNK - A/NK plot [2]. They are the peralkaline, metaluminous, and peraluminous felsic magmatic rocks (Fig. 1). The use of an Al saturation index is particularly relevant for understanding the behavior of U and associated incompatible elements such as Th, Zr, REE … in magmatic rocks because the fractionation of these elements is controlled by the solubilities of the accessory minerals such as monazite, zircon, uraninite, which in turn depend on the degree of Al saturation of the silicate melt [2, 3, 4, 5], as well as of temperature and volatile element content. The A-B diagram of [6] is also used to further refine the distinction between the different types of magmatic rocks, and to track the evolution of the Al saturation index during magmatic fractionation. Each of these rock types is characterized by a specific magmatic fractionation of Th and U and a specific accessory mineral paragenesis [7]. The distribution of U between the different accessory minerals is of critical importance for the genesis of U mineralization. A fourth type of magmatic rock enriched in U corresponds to weakly peraluminous granitoid rocks occurring in migmatitic domains. They are generally referred to as alaskite or anatectic pegmatoid.



Figure 1: Diagram discriminating peraluminous, metaluminous and peralkaline igneous rocks [2]. Only the U-rich igneous rocks are indicated with their corresponding designation in the alphabetic classification. The arrows indicate the effect of an increasing abundance of the minerals at the origin of the Al, Ca and Na-K excess with respect to their ratios in feldspars.

## Peralkaline magmatic rocks

These rocks are characterized by an excess of alkalies, either Na or K, with respect to the amount Al bound to the feldspars (Fig. 1) and they are always enriched to variable degrees in high field strength elements and especially in U. They can be quartz saturated [peralkaline granites or rhyolites] or undersaturated (syenites or trachytes).

## Metaluminous high-K calc-alkaline magmatic rocks:

Certain metaluminous calc-alkaline magmatic rock family are enriched in K and other incompatible elements. Such magmatic rocks are called high-K calc-alkaline (HKCa) granite or shoshonitic granite when K-enrichment reaches levels above 5-6 wt%. These rocks are metaluminous because of an excess of Ca not balanced by Al as in the plagioclase structure. This Ca is hosted by Ca-rich minerals devoid of, or poor in Al, such as clinopyroxene, amphibole, titanite and allanite.

## Peraluminous magmatic rocks:

All magmatic rock with an A/CNK index greater than 1.1 have been defined as S-type by [8], because of their presumed derivation from the melting of metasedimentary rocks. However, in the A-B plot of [6] (Fig. 2) several types of contrasting fractionation trends can be differentiated within the peraluminous field composition: S-type granites [8] with a positive trend, Guéret-type granites (G-type) [9] with a negative trend, metaluminous calc-alkaline magmatic rocks also with a negative trend but rooted in the metaluminous field, L-type magmatic rocks corresponding either to biotite+muscovite±garnet±cordierite leucogranites, as exemplified by the peraluminous leucogranite complexes of Limousin (French Massif Central) [10] or the felsic volcanic rocks of Macusani in Peru [11].



Figure 2 : Peraluminous [A=Al–[Na+K+2Ca]] versus differentiation [B= Fe+Mg+Ti] indices diagram [6], discriminating various magmatic fractionation trends for igneous rocks. Composition fields of the major types of sedimentary rocks are represented. And=Andalusite, Sill = Sillimanite, Mus = Muscovite, Qz = Quartz, F[K] = K-feldspar, Plg = Plagioclase, Hyp = Hypersthene, Mgt = Magnetite, Ilm = Ilmenite, Hnb = Hornblende. Modified from [20].

## Anatectic pegmatoids:

They always occur in high grade metamorphic rocks submitted to partial melting, generally injected as dykes of various size, more or less conformable with the foliation of the enclosing metamorphic rocks, with variable grain size and occur commonly as coarse grained unzoned pegmatoids. At Rössing in Namibia, they are referred to as alaskites. They are commonly weakly peraluminous due to their composition dominated by quartz and feldspars. [12].

#  RELATIVE URANIUM FERTILITY OF U-RICH MAGMATIC ROCKS

Despite their U -enrichment, the magmatic rocks described above have not all the same potential to represent an efficient source of U for the genesis of U deposits.

## Peralkaline magmatic rocks

In peralkaline melts the excess of alkalies with respect to Al (Na+K/Al > 1) and their high temperature, favor a strong melt depolymerization and consequently a high solubility of the incompatible elements, such as U, Th, Zr, Nb, Ta, REE [3, 4, 5]. Hence, U is continuously enriched with the other incompatible elements during magma fractionation, which all reach the highest concentrations in the most fractionated peralkaline melts. Typically, in peralkaline complexes Th (as well as Zr, REE is positively correlated with U and their Th/U ratios remain close to the average crustal ratio during fractionation (Fig. 3). When crystallizing highly fractionated peralkaline plutons, abundant and complex Zr, REE, Th, Nb, and Ta minerals form with U as a minor element substituted in all these minerals [7]. Uraninite is generally not able to crystallize despite the strong enrichment in U of these melts. Extreme fractional crystallization of peralkaline melts may lead to U, Th, Zr, and REE concentrations in the silicate melts up to several hundreds to thousands of ppm and more rarely up to several wt%. However as U is hosted in accessory minerals it cannot be leached by fluids, and they do not represent a favourable U source. However, fractionated peralkaline rocks may represent significant U sources when the U hosting silicate minerals become metamict.

By contrast, peralkaline felsic volcanic rocks are excellent U sources, because most of the U is in the glass. When the glass is devitrified during alteration, U can easily be mobilized.



Figure 4. Evolution of Th and U contents and Th/U ratios during fractionation in U-rich peralkaline, peraluminous and metaluminous HKCa igneous rocks. Main U-bearing minerals specific of each igneous rock type are indicated according to their average Th/U ratio

## High-K calc-alkaline magmatic rocks

HKCa magmas have intermediate to high temperatures and metaluminous to slightly peraluminous compositions which confer them variable degree of polymerization. Consequently, the solubility of Th-, Zr-, and REE-bearing accessory minerals is variable and the behaviour of Th, Zr and REE relatively to U will likewise vary (Fig. 3). Because of the high CaO contents of HKCa magmas, Ca-rich minerals such as amphibole, titanite and allanite crystallize. These minerals incorporate REEs as well as minor quantities of Th and U, that substitute easily for Ca together with in their structure. In a melt with a Th/U ratio close to 4, most of the U will be incorporated in the structure of uranothorite [13, 7]. This mineral, being refractory, U will not be leached by fluids circulatating soon after the granite emplacement and thus will not represent a source for U deposits [7]. However, uranothorite and other Th- and U-rich silicate phases may become efficient U sources during later fluid circulation events when their structure becomes metamict [13].

Allanite is the main U bearing host for high REE/Th ratios in HKCa melts, whereas Nb and Nb-Ti oxides become the main U bearing mineral for high Nb/Th ratios. When the Ca-content of HKCa melts is sufficiently low, monazite become stable and uraninite may crystallize. However, the proportion of uraninite is generally small and such fractionated granites generally represent only small volumes of HKCa complexes.

In conclusion, U-rich high-K calc-alkaline granites may become a significant U source for the formation of U deposits, when their U-bearing accessory minerals have become metamict or when they contain a significant proportion of uraninite. High-K calc-alkaline volcanic rocks may represent a significant U source if the magmas are fractionated and if the U is dominantly hosted by the glassy matrix rather than in accessory minerals.

## Peraluminous magmatic rocks

Peraluminous magmatic rocks cover a wide spectrum of magmas generated in different conditions. They can be distinguished in a A-B chemical-mineralogical diagram (Fig. 2).

### S-type peraluminous magmatic rocks

S-type peraluminous magmatic rocks derive from high degrees of melting of source rocks which do not permit selective partial melting of protolith(s) enriched in U above the crustal Clarke value. During restite unmixing and magma fractionation, decreasing temperature leads to the fractionation of the U-bearing accessory minerals. Consequently, the enrichment in U in the residual melts is moderate, to a level insufficient to crystallize a significant proportion of uraninite. This type of granite is not known to be associated with U deposits.

### G-type magmatic rocks

For G-type magmatic rocks, even if the crustal protolith undergoing partial melting was enriched in U, the U content of the melt will decrease because of its mixing with a metaluminous melt poor in U. Such granite is not known to be associated with U deposits.

### L-type magmatic rocks

When enriched in U, they represent highly favorable U sources because U is dominantly hosted in Th-poor uraninite, easily leachable by hydrothermal fluids or ground waters. Crystallization of U - dominantly as uraninite - results from a succession of processes: [i] The protoliths submitted to melting have to be enriched in U significantly above the Clarke value for the upper continental crust [> 2.7 ppm], in order to have a large proportion of U hosted outside the structure of the accessory minerals, [ii] the degree of partial melting has to remain low, to have preferential melting of quartz-feldspar-rich lithologies [meta-arkoses, felsic magmatic rocks] which are the ones the most likely enriched in U, [iii] during fractional crystallization Th, REE and Zr contents decreases because of monazite and zircon fractionation. As these minerals incorporate only minor amounts of U, U continues to be enriched during fractionation until Th-poor uraninite crystallizes [7]. Uraninite represents a very easily leachable source of metal. In a Th [Zr, REE] versus U diagram, these elements are reversely correlated (Fig. 3), and the Th/U ratio of such granite decreases during fractionation.

Felsic highly peraluminous volcanic rocks equivalent to two-mica peraluminous leucogranites are rare. A typical exemple is the U-rich pyroclastic tuffs at Macusani in Peru [11]. They represent a very good source of U via devitrification of the glass by oxidizing fluids.

# RELATIONS BETWEEN U DEPOSITS AND FELSIC MAGMATIC ROCKS.

U can be enriched either by magmatic processes, or by a variety of hydrothermal processes disconnected with the magmatic activity, or by a combination of magmatic and hydrothermal processes, as well as by leaching by ground waters at low temperature. These relations are reviewed following the genetic classification proposed by Cuney [14, 15].

## Magmatic deposits.

### Fractional crystallization

Peralkaline magmas leads to formation of the only type of U deposit related to extreme magmatic fractionation. Such an association can be extended to carbonatites such as Palabora, South Africa [16]. The U mineralization always occurs within the most fractionated intrusions of the peralkaline complexes, located in their apical part or at their margins. These deposits may represent very large, low-grade U and Th resources, such as the Kvanefjeld deposit at Ilímaussaq, Greenland (> 250,000 tU at 200 ppm) [17, 18]. However, even if the U content of some deposits of this type are relatively high, they are not mined because of the high U extraction cost from refractory minerals. Locally, U, Th, and the REE may be transported by hydrothermal fluids up to several kilometres [Th veins, USA] from the intrusion.

### Partial melting

Low degrees of partial melting of U-rich metasediments or felsic meta-magmatic rocks leads to the formation of anatectic pegmatoids with disseminated uraninite associated with highly variable amounts of other U-bearing accessory minerals. The type example is the Rössing deposit in Namibia (246,500 t U at 300 ppm) whose mineralization is hosted by granitic pegmatite sheets and small plutonic bodies, called alaskites [19; 20].

## Hydrothermal deposits.

### Hydrothermal granitic U deposits

They derive their U mostly from L-type peraluminous leucogranites [21]. The European Variscan Belt which extends for over 2,000 km is the largest province of this type is with U deposits associated with Carboniferous leucogranites. Similar U provinces are known in southeastern China and in Argentinia. The U deposits are dominantly located within the granites in the French Variscides, or in their enclosing metamorphic rocks, in the Erzgebirge district. They occur as veins or as disseminations in de-quartzified granite (episyenite) [22].

HKCa granites may also represent a major source of U for hydrothermal deposits. At Hotagen, in Sweden, U-deposits occur within a Paleoproterozoic HKCa granite but where U was subsequently precipitated during a Caledonian tectonic-hydrothermal event [23]. The large time difference between granite emplacement and hydrothermal circulations resulted in metamictization of uranothorite, the main U-bearing mineral, which becomes an easily leacheable source of U. The U sources for the Olympic Dam deposit in Southern Australia are the host HKCa granites and overlying synchroneous HKCa volcanic rocks [24].

### Hydrothermal volcanic uranium deposits

They are mostly related to peralkaline volcanic rocks. The world largest U district of this type [280,000 t U at 0.2 %] is the late Jurassic Stretsovkoye caldera in Transbaikalia, Russia. The exceptional size of the resources results from the juxtaposition of 4 main U sources: [i] liparitic tuffs which represent 30 to 35 vol% of the volcanic pile, [ii] Variscan U-rich high-K calc-alkaline granitoids in the basement, [25], [iii] Ordovician U mineralization in the basement [26], and [iv] fluids expelled from the volcanic melts or from underlying magma chamber. A quantitative estimate of the amount of U liberated by the liparites is obtained from mass balance calculations between the U content of the melts inclusions from quartz trapped in the liparites and the average present U content of these rocks [25].

HKCa volcanic rocks are generally a less favorable U source because a significant portion of the U in these rocks tends to be trapped in accessory minerals [27]. Most deposits related to this type of volcanism have a relatively small size.

Highly peraluminous acidic volcanics, mineralized in U are known in the Macusani district, Peru. Pitchblende and autunite occur in fractures in Pliocene tuffs. A resource of 30,000 tU has been estimated for the whole Macusani district at an average grade of 0.1 % U [28].

### Hydrothermal diagenetic U deposits od tabular or tectonolithologic type

They may derive a large part of their U from volcanic tuffs, commonly of peralkaline origin, deposited within continental sandstone units of sedimentary basins and leached by diagenetic fluids as in the Arlit-Akouta U district in Niger with more than 100,000 t U at 0.3 % [29].

### Hydrothermal diagenetic U deposits with basement/basin redox control,

They are generally called unconformity related deposits They are characterized by large resources [631,000 t U] and comprised the highest grade U deposits in the world. The source of U for these deposits is debated. For the Athabasca, it is proposed that U is derived either from the sandstone basin [30] or from the basement lithologies and especially the abundant U-rich peraluminous granites and pegmatites [31, 32]. Other possible sources include U-rich HKCa granites mainly emplaced in Taltson Belt, to the west of the Athabasca Basin. It has been shown that, highly saline, acidic, and oxidizing hydrothermal diagenetic fluids [33, 34], were able to liberate U from a refractory mineral like monazite [35].

### Hydrothermal metasomatic U deposits

Na-metasomatic U deposits are associated to regional scale alteration characterized by dequartzification, albitization and later Ca- and K-metasomatism [36]. The largest resources of this type are located in central Ukraine (280,000 t U at 0.08 to 0.13 %). The alteration may affect HKCa granites as at Lagoa Real in Brazil [100,000 t U at 0.12%] [37] and at Kurupung in Guyana [38], or HKCa metavolcanics in Labrador [36,800 t U at 840 ppm], Canada [39].

### Hydrothermal metasomatic U deposits associated with skarns

In the Mary Kathleen U-REE deposit, Australia (8550 t U at 0.11 %), the skarns result from an interaction between the U-Th-rich HKCa Burstall Granite (1737 ± 15 Ma) and enclosing calc-silicates, involving highly saline fluids derived from evaporites [40].

### Olympic Dam, a iron oxide-copper-gold [IOCG-U] deposit

This is by far the world’s largest U resource [2,200,000 t U at 230 ppm]. The deposit occurs in the highly fractionated HKCa Roxby Downs granite (1.59 Ga) emplaced at a very shallow structural level [41]. U source is probably the Roxby Downs granite and the Gawler Range volcanic rocks of similar geochemistry and age. However, the genetic model for the genesis of the U mineralization in the Olympic Dam deposit is still not very well understood, but the most likely U source is the enclosing granite and overlying volcanic rocks.

## Other deposit types

The first U deposits of the Earth, the Archean to Early Paleoproterozoic Quartz Pebble Conglomerates [QPC] of the Dominion Reef and the Witwatersrand Basin in South Africa and the Elliot Lake district in Canada, derive their uraninite crystals of detrital origin, from Archean HKCa granites and pegmatites [42].

Many other types of U deposits may derive their U from spatially related U-rich granites or volcanic rocks. Such an origin is proposed for many roll front deposits from the Wyoming district, with an U derived either from U-rich Archean HKCa granites [43] or from interstratified volcanic ash [44]. Similar processes apply to the calcrete hosted deposits formed by evapotranspiration processes, with the type example represented by the Yeelerie deposit in Western Australia [45]. The relation between granites is still more direct in the paleovalley U deposit type from the Vitim district in Russia which occur in organic matter bearing sandstone deposited in narrow valleys directly incised into HKCa granites [46].

For other U deposit types their relation with a magmatic U source is more tenuous. Large U grade differences exist between black shales and phosphorite-hosted deposits over the world. The high U content of the Cambrian Alum shale of Sweden [47], compared to other occurrences, may be explained from a provenance comprising the alteration of the U-rich Svecofennian granitic basement. Similarly the high U content of the Moroccan phosphorites may derive from the presence of U-rich Hercynian granite in their source area. However, no specific study has yet been carried out to confirm such relationships.

# CONCLUSIONS

U deposits may derive directly and dominantly from magmatic processes in the case of deposits related to extreme fractional crystallization of peralkaline rocks , or to partial melting of U-rich crustal protoliths. However most deposits are related to U leaching by hydrothermal and surficial fluids, much later than the emplacement of the U-rich magmatic rocks. U enrichment above the Clarke value is necessary for a granitic or volcanic rock to represent a viable U source for the formation of U deposits, but is not sufficient. In addition, U has to be hosted in a site from which it can be leached by oxidized hydrothermal fluids or ground waters. Uraninite represent the most easily leachable U source occuring mainly in highly fractionated peraluminous leucogranite and related pegmatites, in weakly peraluminous anatectic pegmatoids and less commonly in highly fractionated high K metaluminous granite and related pegmatites. U hosted in silicates, such as uranothorite, allanite, U-rich zircon and Nb-Ti-oxides can be leached easily only when the structure of these minerals is metamict.

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