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Felsic magmatism and uranium deposits

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Key words. – Uranium, Granite, Acidic volcanic rock, Geochemistry, Ore deposits, Accessory minerals

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 its incompatible behavior, U deposits resulting directly from magmatic processes are quite rare. In most deposits, U is mobilized by hydrothermal fluids or ground water well after the emplacement of the igneous rocks. Of the broad range of granite types, only a few have U contents and physico-chemical properties that permit the crystallization of accessory minerals from which uranium can be leached for the formation of U deposits. The first granites on Earth, which crystallized uraninite, dated at 3.1 Ga, are the potassic granites from the Kaapval craton (South Africa) which were also the source of the detrital uraninite for the Dominion Reef and Witwatersrand quartz pebble conglomerate 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. L-type peraluminous plutonic rocks in which U is dominantly hosted in uraninite or in the glass of their volcanic equivalents represent the best U source. Peralkaline granites or syenites are associated with the only magmatic U-deposits formed by extreme fractional crystallization. The refractory character of the U-bearing minerals does not permit their extraction under the present economic conditions and make them unfavorable U sources for other deposit types. By contrast, felsic peralkaline volcanic rocks, in which U is dominantly hosted in the glassy matrix, represent an excellent source for many deposit types. High-K calc-alkaline plutonic rocks only represent a significant U source when the U-bearing accessory minerals (U-thorite, allanite, Nb oxides) become metamict. The volcanic rocks of the same geochemistry may be also a favorable uranium source if a large part of the U is hosted in the glassy matrix. The largest U deposit in the world, Olympic Dam in South Australia is hosted by highly fractionated high-K plutonic and volcanic rocks, but the origin of the U mineralization is still unclear. Anatectic pegmatoids containing disseminated uraninite which results from the partial melting of uranium-rich metasediments and/or metavolcanic felsic rocks, host large low grade U deposits such as the Rössing and Husab deposits in Namibia.

The evaluation of the potentiality for igneous rocks to represent an efficient U source represents a critical step to consider during the early stages of exploration for most U deposit types. In particular a wider use of the magmatic inclusions to determine the parent magma chemistry and its U content is of utmost interest to evaluate the U source potential of sedimentary basins that contain felsic volcanic acidic tuffs.

Gisements d'uranium et magmatisme felsique

Mots clés. – Uranium, Granite, Roche volcanique acide, Géochimie, Gisements, Minéraux accessoires

Résumé. – L'uranium a un comportement incompatible élevé dans les liquides silicatés qui conduit à sa concentration la plus élevée dans les liquides les plus felsiques et à la prédominance des granites et des rhyolites comme sources d'U primaires pour les gisements d'uranium. Malgré son comportement incompatible très marqué, les gisements d'U résultant directement de processus magmatiques sont relativement rares. Dans la plupart des gisements, l'U est mobilisé par des fluides hydrothermaux ou des nappes phréatiques bien après la mise en place des roches ignées. Parmi les différents types de granites, seuls certains ont des teneurs en U et des propriétés physiques et chimiques permettant la cristallisation de minéraux accessoires à partir desquels l'U sera plus ou moins facilement lessivable pour la formation de gisements d'U. Les premiers granites sur la terre qui ont pu cristalliser de l'uraninite, apparus vers 3.1 Ga, sont les granites fortement potassiques du craton du Kaapval (Afrique du Sud) qui a représenté la source de l'uraninite détritique des conglomérats à galets de quartz du Dominion Reef et du Witwatersrand. Quatre types de roches ignées peuvent être suffisamment enrichies en U pour représenter une source pour des gisements d'U : peralcalines, métalumineuses calco-alcalines fortement potassiques, peralumineuses de type L, et les pegmatoïdes anatectiques. Les roches ignées peralumineuses plutoniques de type L dans lesquelles l'U est localisé de manière dominante dans l'uraninite ou dans le verre de leurs équivalents volcaniques représente les meilleures sources d'U pour les gisements hydrothermaux ou liés à la circulation d'eaux superficielles.

Les granites ou syénites peralcalines sont associées aux seuls gisements d'U magmatiques dérivant d'une cristallisation fractionnée extrême. Le caractère très réfractaire des minéraux d'uranium dans ce type de gisements ne permet pas leur exploitation dans les conditions économiques actuelles et en font des sources d'uranium peu favorable pour la genèse d'autres types de gisements. Par contre les roches volcaniques peralcalines felsiques, dans lesquelles l'U est principalement localisé dans la matrice vitreuse, représentent une excellente source pour beaucoup de types de gise-

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ments. Les roches plutoniques calco-alcalines fortement potassiques représentent une source d'U significative seulement lorsque les minéraux accessoires uranifères (U-thorite, allanite, oxydes de Nb) sont devenus métamictes. Les volcanites de la même géochimie peuvent également être une source d'U favorable si une large part de l'U est localisé dans la matrice vitreuse. Le plus grand gisement d'U du monde, Olympic Dam en Australie du sud est situé au sein de roches plutoniques et volcaniques fortement potassiques et très fractionnées, mais l'origine de la minéralisation en U n'est pas encore bien comprise. Les pegmatoïdes anatectiques avec de l'uraninite disséminée, aussi appelés alaskites, qui résultent de la fusion partielle de métasédiments et/ou de métavolcanites felsiques riches en U, renferment de grands gisements d'U à faible teneur tels que ceux de Rössing et Husab en Namibie.

L'évaluation de la potentialité des roches ignées à représenter une source d'U efficace dans une province donnée représente une étape critique, durant les premiers stades de l'exploration pour la plupart des gisements d'U. En particulier une utilisation plus systématique des inclusions magmatiques afin de caractériser la géochimie des magmas sources et de leur teneur en U est d'un intérêt majeur pour évaluer le potentiel source des bassins sédimentaires présentant une contribution volcanique sous forme de tufs felsiques.

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 the distribution of radiogenic heat flux production 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. Typical range of uranium concentrations measured in rock forming minerals and accessory minerals are given in the figure 1. 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 Moreau [1966] was the first to define the notion of "fertile granites", which has been very fruitful for uranium exploration.

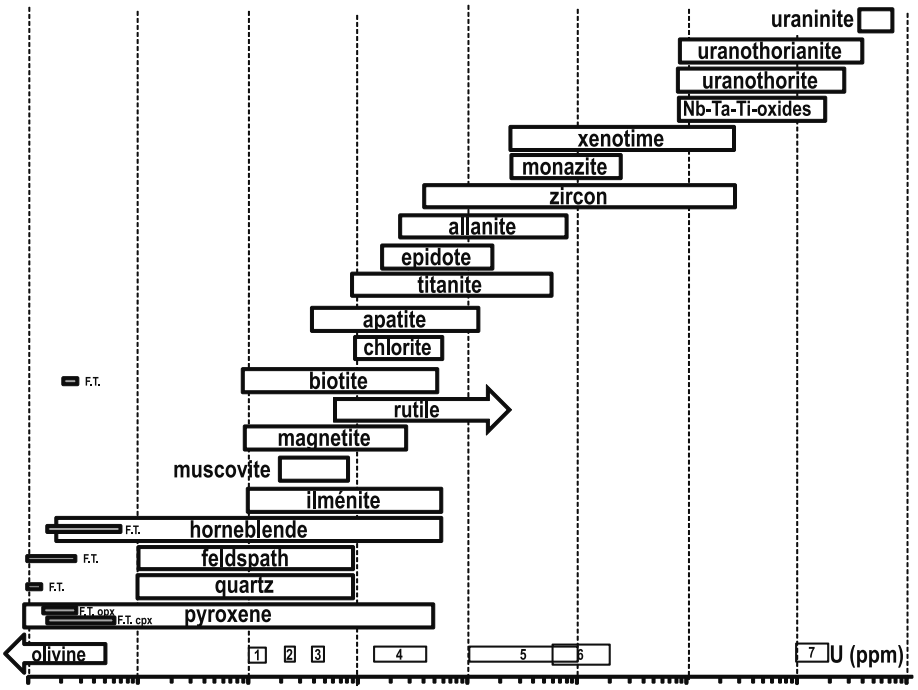

FIG. 1. – Typical range of uranium contents of the main rock forming minerals, accessory minerals and deposits. Values for the major silicates have been mainly measured on separated minerals, they are overevaluated because of the presence of mineral inclusions and/or uranium adsorbed on the surfaces. F.T. = value obtained by fission tracks which really represent the uranium incorporated within the mineral structure. For the accessory minerals concentrations are derived from microprobe analyses. Boxes of the lower part of the diagram: 1 = average crust; 2 = average upper crust; 3 = average granite; 4 = "fertile" granites; 5 = low grade uranium deposits (alaskites); 6 = average grade uranium deposit (500 to 2000 ppm U); 7 = highest grade uranium deposits (10 – 20 wt % U, e.g. unconformity related deposit of McArthur river). The data are derived from a large number of papers.

The aim of the present paper is to review the main types of igneous rocks that are sufficiently enriched in U to be of metallogenic interest, to define the processes leading to their genesis through time, this will facilitate an evaluation of the rock suites best suited as U sources, as well as the specific conditions under which U deposits can be formed by magmatic, hydrothermal or surficial processes from these sources.

THE DIFFERENT TYPES OF URANIUM-RICH IGNEOUS ROCKS

Proposed classification

The three main types of igneous rocks, which can be enriched in U above their Clarke values, will be distinguished according to the A/CNK versus A/NK plot of Shand [1943]. They are the peralkaline, metaluminous, and peraluminous felsic igneous rocks (fig. 2). The use of an aluminum saturation index is particularly relevant for understanding the behavior of uranium 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 are in turn dependant on the degree of aluminum saturation of the silicate melt [Peiffert *et al.*, 1994; 1996; Montel *et al.*, 1993; Watson and Harrison 1983, …], as well as other parameters such as temperature and volatile element content. The A-B diagram of Debon and Lefort [1988] will be also used to further refine the distinction between the different types of igneous rocks, and to track the evolution of the aluminum 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 [Cuney and Friedrich 1987]. The distribution of U between the different uranium hosting accessory minerals is of critical importance for the genesis of U mineralization associated with these rocks. This aspect will be discussed in more detail below. A fourth type of igneous rock which can be enriched in uranium corresponds to weakly peraluminous to metaluminous granitoid rocks occurring in migmatitic domains. They are generally referred to as alaskite or anatectic pegmatoid.

Peralkaline igneous rocks

These rocks are characterized by an excess of alkalies, either sodium or potassium, with respect to the amount of aluminum bound to the feldspars (fig. 2) and they are generally enriched to variable degrees in high field strength elements and especially in uranium. They can be quartz saturated (peralkaline granites or rhyolites) or undersaturated (syenites or trachytes). They are equivalent to the A1-type granite of Eby [1992]. Many hypotheses have been proposed for the genesis of peralkaline rocks [e.g., Macdonald, 1987]. In the present paper we will consider that peralkaline magmas derive from low degrees of partial melting of the mantle, which may itself have been previously enriched in incompatible elements, to produce alkali basalts, and followed by protracted magmatic fractionation to generate the most felsic magmas. This model provides an explanation for the fact that peralkaline igneous rocks are always enriched in U and other incompatible elements. The main parameters controlling their degrees of enrichment are: (i) the degree of partial melting of the mantle, i.e. the lower the melting rate, the richer in uranium the silicate melt, (ii) the degree of enrichment of the mantle by fluids derived from the subducted slab and sediments, (iii) the degree of fractionation of the magma after its extraction, and (iv) the importance of uranium fractionation in exsolved magmatic fluids.

Metaluminous high-K calc-alkaline igneous rocks

Some members of the metaluminous calc-alkaline igneous rock family (I- and A2-type in the alphabetical classification) are enriched in potassium and other incompatible elements (fig. 2). Such igneous rocks are called high-K calc-alkaline granite [A2-type granite of Eby 1992] or shoshonitic granite when K-enrichment reaches levels above 5-6 wt%. These rocks are metaluminous because of an excess of calcium not balanced by aluminium as in the plagioclase structure. This calcium is hosted by Ca-rich minerals devoid of, or poor in alumina, such as clinopyroxene, amphibole, titanite and allanite. Several hypotheses have been proposed for the genesis of these rocks: partial melting of K-rich meta-andesites [Roberts and Clemens, 1993], mixing of basaltic with felsic magmas derived from metasediments [Davis and Hawkesworth, 1993], assimilation of metapelites by high-Al basaltic magmas [Patiño Douce, 1995], and melting of metasomatized mantle, eventually accompanied by assimilation of crustal rocks, associated with fractional crystallisation [e.g., Schaltegger and Corfu, 1992]. But, at least for the basic to intermediate highly potassic magmas metasomatism of the mantle by subduction fluids would appear to be the main process for explaining their enrichment in K, U, Th.

Peraluminous igneous rocks

All igneous rock with an A/CNK index greater than 1.1 have been defined as S-type by White and Chappell [1977], because of their presumed derivation from the melting of metasedimentary rocks. However, in the A-B plot of Debon

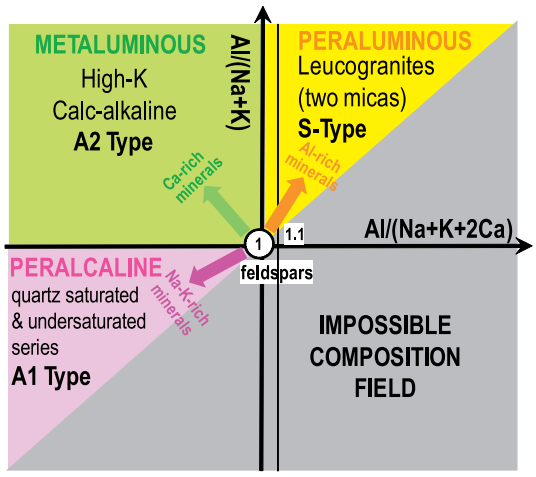

FIG. 2. – Diagram of Shand [1943] discriminating peraluminous, metaluminous and peralkaline igneous rocks. Only the uranium-rich igneous rocks have been indicated with their corresponding designation in the alphabetic classification. The arrows indicate the effect of an increasing abundance of the minerals at the origin of the excess of Al, Ca and Na-K excess with respect to their proportions in feldspars for each igneous rock type.

and Lefort [1988] (fig. 3) several types of contrasting fractionation trends can be differentiated within the peraluminous field composition.

S-type granites, have been first defined in the biotite-cordierite- bearing Kosciusko batholith (southeastern Australia) by White and Chappell [1977]. In the A-B diagram (fig. 3) they define a trend in which the most mafic compositions plot in the center of the greywacke field with the highest peraluminous indices. Such compositions result from a high degree of partial melting (> 50%) of metagreywacke or metapelite in dry conditions at temperatures of 800°C to 1000°C [Burkhard, 1991; Vielzeuf and Holloway, 1988]. During magmatic fractionation, with decreasing B value, the aluminous saturation index A typically decreases as a result of unmixing of femic and peraluminous restitic minerals (cordierite and or garnet) in accordance with the model proposed by White and Chappell [1977]. The composition of the magma evolves towards the eutectic (the origin of the A-B diagram) with a positive correlation between the A and B parameters.

Guéret-type granites [G-type], as defined in the French Massif Central [Stussi and Cuney, 1993; Turpin *et al.*, 1989] have the same range of mafic mineral proportions as the S-type Kosciusko Batholith, but the correlation between the A and B parameters is negative, implying a different genetic processes. The least fractionated granites, richest in mafic minerals (mainly biotite), are the least peraluminous. The most leucocratic bodies are the most peraluminous, with increasing cordierite ± muscovite proportions. Rb-Sr and Sm-Nd isotopic studies [Turpin *et al.* 1989] have shown that this type of geochemical trend can be explained by a mixing between a peraluminous leucocratic granitic melt derived from a limited amount of partial melting of crustal material and a metaluminous melt derived from the partial melting of metaluminous rocks at the base of the continental crust or from the partial melting of the mantle.

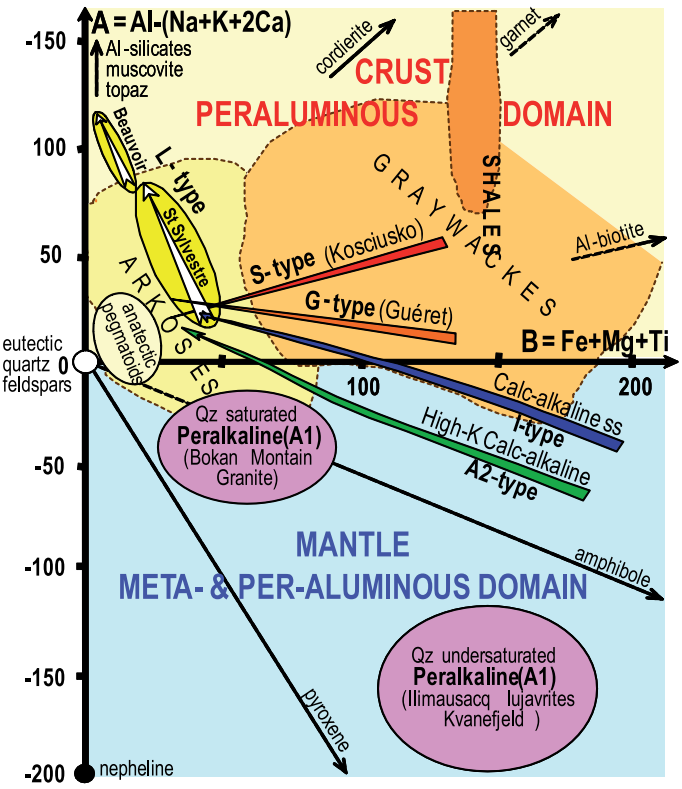

FIG. 3. – Peraluminous index (A = Al – [Na+K+2Ca]) versus differentiation index (B = Fe+Mg+Ti) diagram of Debon and Lefort [1988], used to discriminate various magmatic fractionation trends among peraluminous, metaluminous and peralkaline igneous rocks (Bokan Mountain granite, Ilimausaq lujavrites) and anatectic pegmatoids (Rössing). Composition fields of the major types of sedimentary rocks are also plotted. They are dominantly peraluminous. Graywacke and arkose may be metaluminous if they have a carbonate cement. Main rock-forming minerals are also plotted: And = andalusite, Sill = sillimanite, Mus = muscovite, Qz = quartz, F[K] = K-feldspar, Plg = plagioclase, Hyp = hypersthene, Mgt = magnetite, Ilm = Ilmenite, Hnb = hornblende. Modified from Cuney and Kyser [2008].

Metaluminous calc-alkaline igneous rocks (I and A2 type) may become peraluminous as a result of one or more of the following processes: (i) extreme magmatic fractionation of metaluminous minerals like amphibole or pyroxene which selectively depletes the melt in calcium but not in aluminium; (ii) assimilation of peraluminous material during ascent of the magma through the continental crust; (iii) late magmatic to hydrothermal alteration leading to the fractionation of Na, K, and/or Ca into Cl-bearing late magmatic to hydrothermal fluids. This may lead to the crystallization of muscovite in the miarolitic cavities of some granites when fluid oversaturation of the magma occurs at shallow levels, or to the sericitization of the feldspars during post-magmatic hydrothermal circulation. Generally, this type of peraluminous igneous rock represents a small volume of the whole calc-alkaline magmatic complex and its peraluminosity remains limited, except for the most fractionated portions of the suite, such as the low phosphorus Rare Metal Granites [Linnen and Cuney, 2005; Cerný *et al.,* 2005].

L-type igneous rocks correspond either to biotite+muscovite±garnet±cordierite leucogranites, as examplified by the peraluminous leucogranite complexes of Limousin (French Massif Central) [Cuney *et al.*, 1989] or the felsic sillimanite+muscovite±andalusite volcanic rocks of Macusani in Peru [Pichavant *et al.*, 1988]. Contrary to the typical S-type granites, mafic mineral contents remain below 10%, and the peraluminous index increases markedly during magmatic fractionation (fig. 3). L-type igneous rocks form from low degree of partial melting (< 30%) of quartz-feldspar-rich protoliths such as meta-arkose, felsic metavolcanic rocks, or metagranite [Turpin *et al.*, 1989].

Anatectic pegmatoids

Anatectic pegmatoids always occur in high grade metamorphic rocks submitted to partial melting. They are generally injected as dykes of various size, more or less conformable with the foliation of the enclosing metamorphic rocks. They exhibit 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 with rare garnet and muscovite. Biotite does not exceed generally two percents, but may locally occur as schlierens. The pegmatoids can be slightly metaluminous by reaction with enclosing marbles or calc-silicate rocks. They are interpreted to form as result of low degrees of partial melting of metasedimentary rocks [Cuney, 2009].

In early Earth history, igneous rocks enriched in U did not exist. They have appeared progressively through time. The following section will review the processes presumed

to be at the origin of the different types of U enriched igneous rocks though time.

URANIUM FRACTIONATION IN THE EARLY EARTH

Early Archean magmatic rocks

Before 3.2-3.1 Ga, the first processes by which U enrichment occurred was partial melting of the primitive mantle [U = 20.5 ppb, Javoy, 1998] to form the metaluminous komatiitic and basaltic melts. Limited fractional crystallization followed during ascent of these melts to the surface. These processes led to the formation of a thin and dominantly mafic crust. Partial melting of these rocks is considered to be largely responsible for the origin of the most felsic rocks formed in Archaean granite-greenstone belt terranes at that time: the tonalite-trondhjemite-granodiorite (TTG) association [Martin, 1994; Moyen, 2011]. The uranium content of these early felsic rocks is typically below 1 to 2 ppm. At such low concentration, U is nearly entirely hosted by the structure of common accessory minerals (zircon, titanite, monazite, allanite, apatite) and thus cannot have been significantly extracted by hydrothermal fluids. Uraninite or other U-minerals have never been reported in pre-Mesoarchean magmatic rocks. Even if a small fraction of U could have been extracted from the accessories, the reduced atmospheric conditions during this period of time would not have permitted the oxidation of U from the tetravalent state, its form within the accessory minerals, to the hexavalent state required to form the highly soluble uranyl complexes. Hence, before 3.1 Ga, U was never sufficiently enriched to represent a viable source for the formation of ore deposits. Thorium, which has the same ionic radius as U, and which exists only in the tetravalent state in natural systems, behaved at that time similarly to U in reducing conditions, and the initial Th/U ratio of the primitive Earth of about 4 has remained nearly constant under these conditions.

The first uraninite bearing granites

Between 3.1 and 2.2 Ga U continued to be dominantly fractionated by magmatic processes but new conditions of fractionation permitted the appearance of the first granites sufficiently enriched in U to crystallize uraninite. The oldest of these granites are known at 3.1 Ga in the Kaapvaal-Kalahari craton in South Africa [Robb and Meyer, 1988; 1990]. They are highly felsic and much richer in potassium than the TTG. They belong to the granodiorite-granite-monzogranite suites (GGM) of de Wit [1998]. They were intruded between ~3.2 and 3.0 Ga inside and along the boundaries of a series of crustal blocks which collided to form the Kaapvaal craton [de Wit *et al.*, 1992]. Then, between ~2.75 and 2.5 Ga, at least three other generations of potassic granite were emplaced in this region [Poujol *et al.*, 2003]. Aplites and pegmatites enriched in uraninite, associated with large high-K metaluminous to slightly peraluminous granitic complexes [Clemens *et al.*, 2010] also dated at about 3.1 Ga, have been identified in the Barbeton belt [Carroué *et al.*, 2012]. High-K metaluminous granites are also known in most other Archean cratons, as in the Early Archean Pilbara, the Late Archean Yilgarn cratons, and the basement of the East Alligator River Uranium district of Australia, in the Manitoba Superior province and Central Québec in Canada, Wyoming in the USA, in the Amazon craton of Brazil, in the Dharwar and Singhbum cratons of India, the Zimbabwean craton and others (table I). Generally, these granites cover much larger surfaces than

TABLE. I. – Pre 2.2 Ga granitoids and pegmatites in which uraninite has been identified. 1: Hallbauer, [1984]; 2: Robb and Meyer, [1990]; 3: Bruguier *et al.* [1994]; 4: Sarangi and Singh [2006]; 5: Duhamel *et al.* [2009]; 6: Jayayanda *et al.* [2000]; 7: Salobo Iron Oxyde Copper Gold deposit related to the old Salobo high-K calcalkaline granite, Requia *et al.* [2003]; 8: Santosh *et al.* [2004]; 9: Carroué *et al.* [2012]; 10: Drouet [2012]; 11: Stuckless *et al.* [1980]; 12: Som and Sai Baba [2005]; 13: Sharpe and Fayek [2011].

Rock type	Location	Typology	Age [Ga]	Th [ppm]	U [ppm]	Th/U	U minerals	Ref
Johannesburg Granite	South Africa	High-K Ca	3.060±30	1.1 -17.5	0.2 -12.6	-	uraninite	1
Hinterland Witwatersrand Granite	South Africa	Peraluminous ?	> 3.08	1.9 - 26.7	8.2-116	0.15 - 3.7	uraninite	2
Heerenveen and Mpuluzi aplite/pegmatite	South Africa	High-K Ca ?	3.105	-	-	-	uraninite	9
Kaduna orthogneiss,	Nigeria	?	3.050±23	-	-	-	uraninite in zircon	3
Singhbum Granite	India	Peraluminous	3.0-2.9	-	-	-	uraninite, all., titan.	4
Karimnagar charnockite	India	High-K Ca ?	2.6 – 2.8	-	-	-	uraninite	6, 8
Namlekonda granite	India	?	?				uraninite	12
Tanco Pegmatite	Manitoba, Canada	Peraluminous	2.640 ±7	1	55	0.02	uraninite, U-microlite	5
North-Central Québec	Canada	Peraluminous	2.580				uraninite	10
Huron claim pegmatite	Canada	anatectic	2 575	-	-	-	uraninite	13
Salobo IOCG	Brazil	High-K Ca	2.573±2	-	-	-	uraninite	7
Compeau Creek Gneiss	USA	High-K Ca ?	-	0.8 – 32.8	0.8 – 39.6	0.13-1.4	uraninite ?	11

the TTG in present exposures [Bowring and Housh, 1995; Kinny and Nutman, 1996; Frost *et al.*, 1998; Drüppel *et al.*, 2009]. For example, in the Wyoming province, TTG are restricted to rocks older than 2.8 Ga, whereas Late Archean highly potassic granites represent most of the plutonic rocks, and were emplaced during four periods, at ~2.8, 2.67, 2.63, and 2.55 Ga [Frost *et al.*, 1998]. In the Archean basement of the Rum Jungle province TTGs are nearly absent. Despite the high U contents and low Th/U ratios of these potassic granites, uraninite has been relatively rarely identified in petrographic descriptions and still more rarely analyzed. Also, the aplites and pegmatites, deriving from the extreme fractionation of these granites were probably the richest in uranium, as observed in the occurrences of the Barbeton belt [Carroué *et al.*, 2012]. However, these aplites and pegmatites have been probably largely eroded, because they were generally emplaced in their apices.

Some Archean uraninites may derive from magmatic hydrothermal processes related to the unmixing of a fluid phase from highly fractionated high-K calc-alkaline magmas, such as those of the iron oxide-copper-gold (±U) deposits of the Amazonian craton in Carajas, Brazil [Tallarico *et al.*, 2005]. For example, the Igarapé deposit is spatially and probably genetically related to the high-K calc-alkaline granites of Old Salobo [2573 ± 2 Ma; Requia *et al.*, 2003] and Itacaiúnas [2560 ± 37 Ma, Souza *et al.*, 1996].

Genetic models for the Archean high-K granites

Several hypotheses have been suggested to explain the genesis of Archean potassic granites. One suggestion is that they originated through partial melting of the TTG's and associated metasedimentary rocks at medium to deep crustal levels [Frost *et al.*, 1998]. It has been also suggested that they were derived from the partial melting of a mantle wedge peridotite previously metasomatized either by melts derived from slab melting [Martin *et al.*, 2010] or by fluids enriched in incompatible elements produced by the dehydration of sediments injected along the subduction zone, as proposed for post-Archean high-K granites.

Partial melting of early Archean crust does not explain the high U contents of high-K granites and in particular those which are able to crystallize uraninite. Very small increments of partial melting of TTG and/or associated sediments (typically containing less than 1 ppm U) would be required to generate granites enriched in U by 1 to 2 orders of magnitude. Another option would be to consider the partial melting of a mantle wedge peridotite metasomatized through interaction with slab melts as proposed to explain the origin of the sanukitoids [Martin *et al.*, 2010]. However, these rocks are metaluminous monzodiorites to granodiorites rich in Mg, Ni, Cr, in most LILE and in LREE, and have U contents that are too low to permit the crystallization of uraninite. A similar mechanism followed by mixing with crustal melt is proposed for the Late Archean Closepet metaluminous porphyritic monzogranite, which is richer in incompatible elements [Moyen *et al.*, 2001]. The latter characteristic is explained by a higher enrichment of the mantle source by slab melts [Jayananda *et al.*, 2000]. However, the highest enrichment of radio-elements in the Closepet granitic complex is reached in its northernmost part with 4.5 wt% K_2O, 40 ppm Th, and 6 to 7 ppm U [Kummar and Ready, 2004]. Such high Th and U contents and Th/U ratios is explained by the occurrence of allanite as the main U and Th bearing mineral [Jayananda *et al.*, 2000], but not of uraninite which requires lower Th/U ratios to crystallize.

Hence, the genesis of uraninite bearing high-K magmas probably requires a greater initial enrichment of U and Th in the subcontinental lithospheric mantle. These elements are particularly mobile through a fluid phase and may derive from the dehydration of subducted sediments rather than from the subducted oceanic slab in which water in bound to more refractory minerals such as amphibole. During the Mesoarchean, therefore, subduction of sediment may have occurred locally to produce significant U and K enrichments in the subcontinental lithosphere. The partial melting of the metasomatized mantle wedge could have generated K- and U-enriched andesitic magmas with subsequent fractionation giving rise to granitoid intrusions enriched in incompatible elements as proposed for the modern high-K calc-alkaline granites. However, further research is required to constrain the mechanisms of U enrichment in the early high-K granites able to crystallize uraninite from about 3.1 Ga.

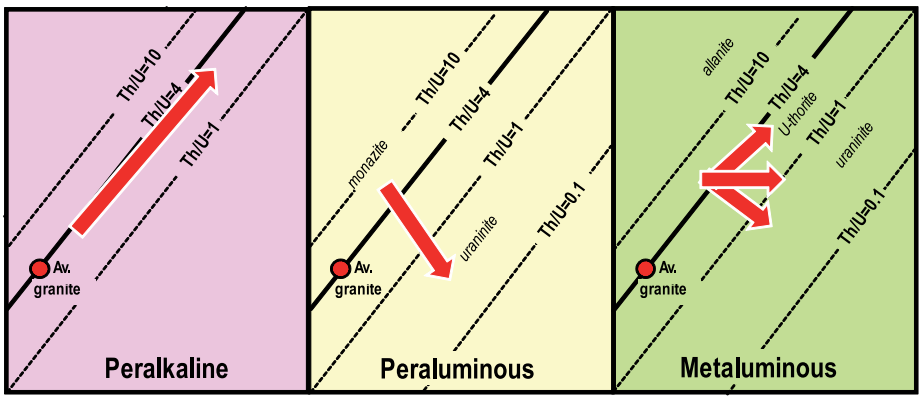

FIG. 4. – General evolution of Th and U contents and Th/U ratios during magmatic fractionation in uranium-rich peralkaline, peraluminous and metaluminous high-K calc-alkaline igneous rocks. The main U-bearing minerals specific of each type of igneous rock have been indicated in the diagram according to their average Th/U ratio.

The first peraluminous uraninite bearing igneous rocks

The first felsic magmas to crystallize a low Th-uraninite began to appear by about 2.7 Ga such as the Tanco pegmatite in Manitoba, Canada [Duhamel *et al.*, 2009]. These granites are enriched in uranium to 10-100 ppm (table I), but the most felsic members are typically depleted in most other HFSE elements (Th, REE, Zr) because of the low solubility of Th-REE-Zr-bearing accessory minerals in low-temperature highly peraluminous melts [Montel, 1993; Watson and Harrison, 1983; Cuney and Friedrich, 1987].

The first peralkaline igneous rocks

Although peralkaline rocks are commonly observed in Archaean terranes, they are volumetrically insignificant, and represented by lamprophyric dikes and syenitic intrusions. The oldest well-documented examples are the 2.7 Ga high-K trachyte and leucite phonolite from Kirkland Lake in Canada [Blichert-Topf *et al.*, 1996]. Their the volumetric insignificance during early Archean times may result from the fact that mantle temperatures were too high to obtain the low degree of partial melting needed to generate this type of magma [Hattori *et al.*, 1996]. It might also be that they were poorly preserved, because they emplaced at a very high structural level [Blichert-Toft *et al.*, 1996]. The degree of U-enrichment in these rocks was also moderate.

RELATIVE URANIUM FERTILITY OF URANIUM-RICH IGNEOUS ROCKS

Despite their U-enrichment relative to the Clarke value, the igneous rocks described above have not all the same potential to represent an efficient source of U for the genesis of U deposits. Beside their degree of U-enrichment, their fertility depends on the nature of the sites in which U is hosted and the ease with which U can be leached by hydrothermal or fluids of ground water. The following section examines the nature of the different U bearing mineral parageneses in various igneous rock types.

Peralkaline igneous rocks

In peralkaline rocks the excess of alkalies with respect to alumina (Na+K/Al > 1) and the high temperature of the silicate melts, favor both a strong melt depolymerization and consequently a high solubility of the large highly charged elements, such as U, Th, Zr, Nb, Ta and the REE's [Peiffert *et al.*, 1994, 1996; Montel, 1993; Watson and Harrison, 1983]. Hence, uranium 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 a Th (Zr, REE) versus U diagram, peralkaline complexes define a positive correlation (fig. 4) and their Th/U ratios remain close to the average crustal ratio during magmatic fractionation. Interaction with late-magmatic fluids may lead to a slight decrease of the Th/U ratio.

During crystallization of highly fractionated peralkaline melts (fig. 5), in plutonic bodies, abundant and complex Zr, REE, Th, Nb, and Ta minerals form (e.g. pyrochlore $(Ca,U,REE)(Nb,Ta,Ti)_2 O_6(O,OH,F)$, eudyalite $(Na,LREE,Ca,K)_{15}(Ca,Mn)_6(Mn,Fe)_3(Zr,Nb,Hf)_3 (Nb,Ta,Si) Si_{26}O_{74} (OH,Cl,F)_2,2H_2O$, steenstrupine $Na_{14}Ce_6Mn_2Fe_2(Zr,Th,U)(Si_6O_{18})_2(PO_4)_7,3H_2O$), with U as a minor element substituted in the structure of all these minerals [Cuney and Friedrich, 1987]. Individual crystals of uraninite are generally not able to crystallize despite the strong enrichment in uranium 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 weight percent. However as uranium is hosted in accessory minerals from which it cannot be leached by hydrothermal fluids or ground waters, they do not represent a favourable source of uranium. However, highly fractionated peralkaline rocks may become significant U sources when U is hosted by silicate minerals and when the latter become metamict.

By contrast with peralkaline plutonic rocks, peralkaline felsic volcanic rocks (e.g., liparites) represent an excellent U source, because most of the U is in the glassy matrix. When the glass becomes devitrified during alteration, U can easily be mobilized. These units form extensive and thick layers of pyroclastic tuffs (ignimbrites) within and outside volcanic calderas. The Latium Province in Italy, an area covering more than 2000 km^2, represents one of the most recent examples of such a pyroclastic tuff sheet strongly enriched in U [Villemant and Palacin, 1987].

High-K calc-alkaline igneous rocks

The intermediate to high temperatures of highly fractionated high-K calc-alkaline melts and their metaluminous to slightly peraluminous compositions lead to a variable and intermediate degree of polymerization of the silicate melts. Consequently, the solubility of Th, Zr, and REE bearing accessory minerals will be variable and lower than in peralkaline melts, and the behaviour of Th, Zr and REE relatively to uranium will likewise vary, with lower degrees of enrichment (fig. 4). With magmatic fractionation and increasing U concentrations, these elements may increase slightly in the magmas, remain constant, or decrease slightly, depending on the melt temperature and aluminosity. These magmas are also characterized by high Ca contents. When the CaO contents in the silicate melt exceeds about 1 wt.%, crystallization of Ca-rich minerals such as amphibole, titanite and allanite occur. These minerals incorporate REEs as well as minor quantities of Th and U, that substitute easily for Ca in their structure (fig. 5). If the Th/REE ratio of the melt is sufficiently high, Th and U will crystallize together as thorite (Th-silicate), which may incorporate up to 30 wt.% UO_2 in its structure [Pagel, 1982; Cuney and Friedrich, 1987]. Consequently, in a melt with a Th/U ratio close to 4, most of the uranium will be incorporated in the structure of uranothorite. This mineral, being very refractory, will not be affected by hydrothermal fluids or ground waters, which circulate soon after the granite emplacement and will not represent a source for uranium deposits [Cuney and Friedrich, 1987]. However, uranothorite and other Th- and U-rich silicate phases (allanite, zircon) may become efficient U sources during later fluid circulation events when their structure is destroyed as a result of alpha-recoil during U decay (metamictisation) [Pagel, 1982]. Significant metamictisation of the U-bearing silicates typically requires a time laps of 200 Ma.

Allanite may represent the main U bearing host for high REE/Th ratios in the melts, whereas Nb and Nb-Ti oxides become the main U bearing mineral for high Nb/Th ratios. When the fractionated high-K calc-alkaline melts become slightly peraluminous and/or when their temperature and Ca-content has decreased sufficiently, monazite may become stable and Th-bearing accessory minerals start to fractionate to induce a decrease in Th/U ratios, and permit the crystallization of uraninite. However, the proportion of uraninite is generally small and such highly fractionated granites generally represent only small volumes of high-K calc-alkaline complexes. So, the amounts of U available as uraninite remains limited. Moreover, when uraninite crystallizes in equilibrium with Th-rich minerals (uranothorite), it is characterized by high Th contents (8 to 15 wt.% ThO_2) [Pagel, 1982], which make it less soluble in hydrothermal or surficial fluids.

In conclusion, U-rich high-K calc-alkaline intrusions bodies 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. However, high uraninite contents in such granite suites are rare compared to peraluminous leucogranites, and associated secondary U deposits will be limited in extent.

High-K calc-alkaline volcanic rocks may represent a significant uranium source if the magmas are fractionated and if the U is dominantly hosted by the glassy matrix rather than in U-bearing accessory minerals.

Peraluminous igneous rocks

In S-type igneous rocks, the high degrees of melting of source lithologies does not permit any selective partial melting of specific protolith(s) enriched in uranium above

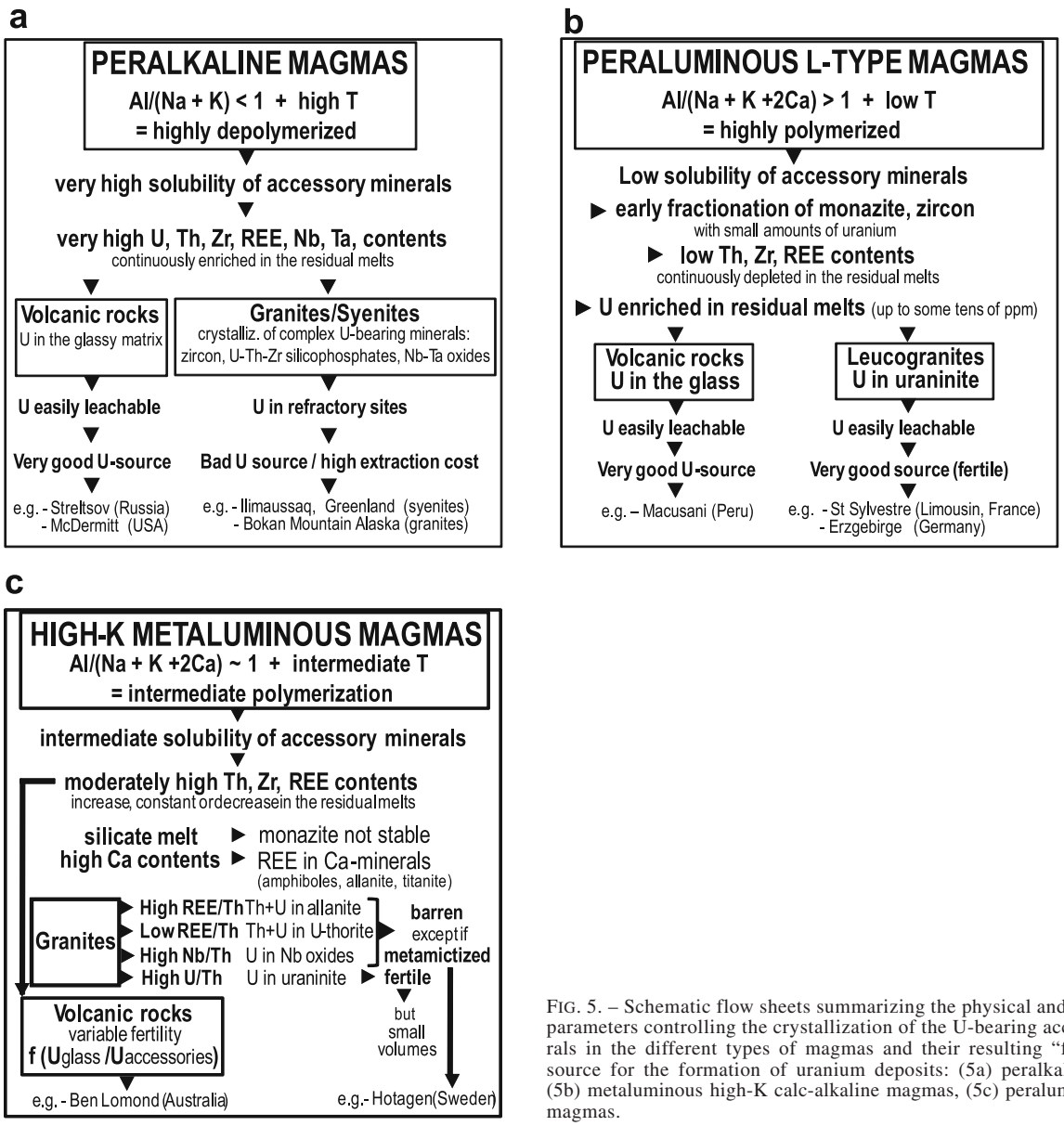

FIG. 5. – Schematic flow sheets summarizing the physical and geochemical parameters controlling the crystallization of the U-bearing accessory minerals in the different types of magmas and their resulting "fertility" as a source for the formation of uranium deposits: (5a) peralkaline magmas, (5b) metaluminous high-K calc-alkaline magmas, (5c) peraluminous L-type magmas.

the crustal Clarke value. Hence, the U concentrations of the various lithological layers are averaged and the U content of the melt remains moderate with U dominantly bound to the structure of the common accessory minerals. During magma fractionation, decreasing temperature during restite unmixing leads to the fractionation of the U-bearing accessory minerals. Consequently, the enrichment in U in the residual melts remains moderate, to a level never sufficient to obtain the crystallization of a significant proportion of U as uraninite. This type of granite is not known to be associated with U deposits.

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

L-type igneous rocks, when enriched in U, 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:

– 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. The fraction of uranium hosted in accessory minerals, such as monazite, zircon, and apatite, cannot contribute to the enrichment of the melts because accessory minerals are only sparingly soluble in low temperature peraluminous silicate melts [Watson and Harrison, 1983; Montel, 1993]. These accessory minerals will be enriched in the restite material together with U [Friedrich *et al.*, 1987];

– the degree of partial melting has to remain low to favor U enrichment in the resulting melt and to melt preferentially the lithologies dominantly composed of quartz and feldspar (meta-arkoses, felsic volcanic or plutonic rocks) which are the ones the most likely enriched in U;

– during fractional crystallization of the U-enriched peraluminous melts, decreasing temperature and increasing peraluminosity decrease accessory minerals solubility along a protracted liquid-line-of-descent. More particularly, the fractionation of monazite, the main Th- and REE-bearing mineral in peraluminous magmas, depletes melt in Th and REE. Uranium is not depleted because monazite and other accessory minerals (zircon, apatite) incorporate only minor amounts of uranium (fig. 5). As a consequence, the U remaining in the melt, not retained in the structure of the accessory minerals, continues to be enriched during fractionation until the silicate melt reaches uraninite saturation and Th-poor uraninite crystallizes [Cuney and Friedrich, 1987]. It is remarkable that the U-rich peraluminous granites associated with uranium deposits, have U concentrations in the order of 10-30 ppm, consistent with values obtained in the experimental studies of uraninite solubility in peraluminous silicate melts at about 800°C [Peiffert *et al.*, 1994; 1996].

U dominantly hosted by Th-poor uraninite, represents a very easily leachable source of metal [Cuney and Friedrich, 1987]. The most fractionated members of this type of peraluminous granite are represented by rare metal, high-P highly peraluminous granite or pegmatite (LCT-type), extremely depleted in Th [down to less 1 ppm], Zr (~20 ppm), and REE (close to or below chondritic abundances) [Linnen and Cuney, 2005; Černý *et al.,* 2005]. In a Th (Zr, REE) versus U diagram, these elements are reversely correlated (fig. 4). Hence, the Th/U ratio of such granite decreases during fractionation. Magmatic fractionation of highly peraluminous, low-temperature melts is probably the only way to produce differential fractionation of thorium relative to uranium without requiring a redox process.

Strongly felsic, highly peraluminous volcanic rocks equivalent to two-mica peraluminous leucogranite are quite rare. Such peraluminous melts are produced at relatively low temperatures and rich in water, two parameters favoring the intersection of the granitic solidus during their rise before they reach the earth surface. One of the rare occurrences of U-rich highly peraluminous volcanics is the Macusani volcanic field in Peru, where they occur as thick and extensive sheets of pyroclastic tuffs [Pichavant *et al.,* 1988]. These tuffs have the same composition as peraluminous leucogranite with about the same levels of U enrichment (up to about 30 ppm U), and very low Th, Zr, and REE contents. They represent a very good source of U via devitrification of the glass by oxidizing fluids.

RELATIONS BETWEEN URANIUM DEPOSITS AND FELSIC IGNEOUS ROCKS

Uranium can be enriched either by purely magmatic processes such as during extreme fractionation of peralkaline intrusions, 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 in the following paragraphs following the genetic classification proposed by Cuney [2010; 2011; 2013] (table II) and have been represented schematically along the geological cycle in figure 6.

Magmatic deposits

Two types of contrasted uranium deposits can be formed dominantly by magmatic processes: those resulting from extreme fractional crystallization and those resulting from very low degree of partial melting.

Extreme fractional crystallization of peralkaline magmas leads to formation of the only type of uranium deposit related to magmatic fractionation. As an outcome of such a process, the uranium mineralization always occurs within the most fractionated intrusions of the peralkaline complexes. These intrusions are generally located in the apical part of the complexes or at their margins, where low viscosity residual melts and associated exsolved fluids are emplaced. The fluid/melt partition coefficients are extremely low in peralkaline systems [Peiffert *et al.*, 1996]. However in some occurrences, U, Th, and the REE are transported by hydrothermal fluids a few hundreds of metres (e.g., Bokan Mountain) to several kilometres (e.g. Th veins of the Front range) from the intrusion, but none of these veins has sufficient uranium grade or tonnage to be mined.

These deposits may represent very large, low-grade U and Th resources, such as the Kvanefjeld deposit at

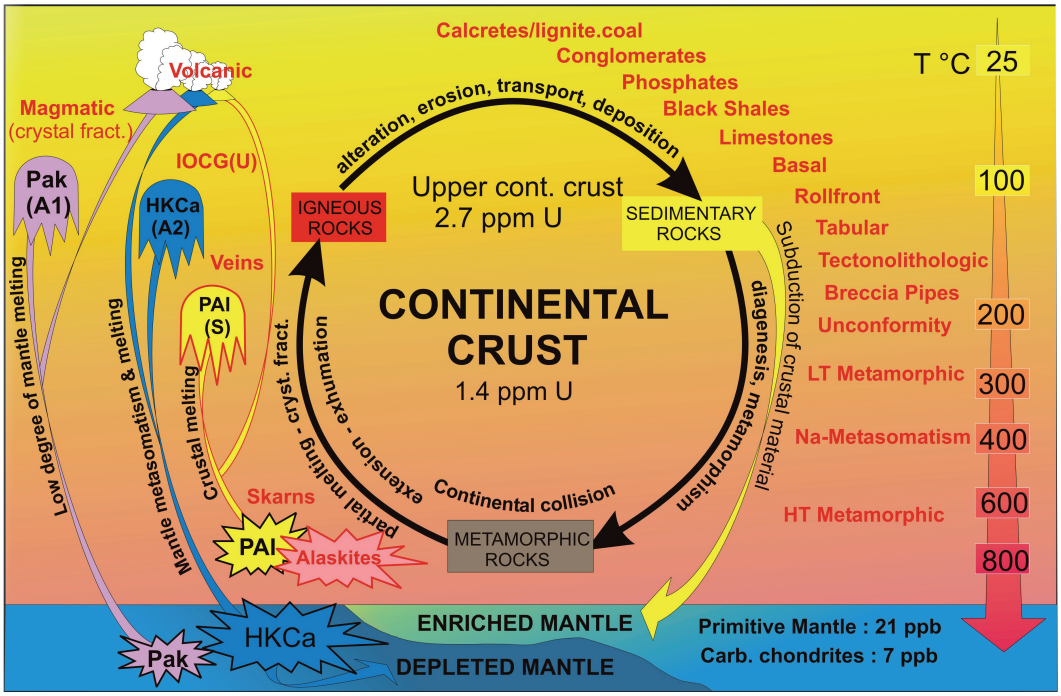

FIG. 6. – General geologic cycle along which the various types of uranium deposits may occur. Pak = peralkaline magmas, HKCa = high-K calc-alkaline magmas, Pal = peraluminous magmas; A1, A2, S correspond to the granite classification equivalents using letters (see the text explanations) [modified from Cuney, 2010, 2011, 2013].

Ilímaussaq, Greenland [Sørensen, 2001; Bohse *et al.,* 1974]. Other major occurrences of this type are: Pocos de Caldas, Brazil [Fraenkel *et al.,* 1985], Bokan mountain, Alaska [MacKevett, 1963], Lovozero massif, Kola peninsula, Russia [Balashov, 1968], and the Kaffo valley, Nigeria [Bowden and Turner, 1974]. Such an association can also be extended to the ultimate fractionation products of peralkaline complexes, namely carbonatite intrusions such as Palabora, South Africa [Verwoerd, 1986]. However, even if the U content of some deposits of this type may be relatively high, they generally not have been mined because of the high cost of uranium extraction from refractory minerals.

The Kvanefjeld deposit in the Ilímaussaq peralkaline complex [Sørensen *et al.,* 1974] represents one of the best examples of of a U deposit associated with the most strongly fractionated syenite of a peralkaline complex, where U is mainly hosted by steenstrupine, a complex silico-phosphate of U, Th, and REE. The resources of the deposit are over 250,000 tU at a grade around 200 ppm.

Low degrees of partial melting of uranium-rich metasediments or felsic meta-igneous 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 whose mineralization is hosted by granitic pegmatite sheets and small plutonic bodies, called alaskites [Berning *et al.*, 1976; Cuney, 1980, 1982; Cuney and Kyser, 2008]. They typically intrude into epicontinental sediments, possibly associated with acidic volcanic rocks, metamorphosed to a high grade with accompagning anatexis. The Rössing U deposit in Namibia (246,500 t) is one of the lowest grade (300 ppm) U deposits ever mined.

The large accumulation of alaskite dykes with relatively high U grade at Rössing results from the combination of the following parameters: (i) a U-rich source represented by intracontinental platform sediments (arkoses, quartzites, black shales, marls, limestones), probably associated with felsic volcanic rocks; (ii) a low degree of partial melting, dominantly affecting the quartz-feldspar-rich lithologies of the volcanosedimentary sequence which explains their weakly peraluminous character and high degree of U enrichment; (iii) a structural control of alaskite emplacement linked to the late kinematic evolution of the Rössing Dome [Basson and Greenway, 2004]; (iv) the existence of a chemical barrier, which was able to stop the rise of the alaskitic melts which reacted with enclosing marbles of the Rössing formation or calcsilicate rocks of the Khan formation to form skarns. This in turn led to the production of CO_2 shifting the solidus of the melts to high temperatures to facilitate their accumulation in the vicinity of the marble layers; (v) a reducing barrier represented by the sulfide- and graphite-bearing Rössing formation, which has prevented the fractionation of U in magmatic fluids at this level, and promoted the entrapment of U from magmatic fluids deriving from alaskites crystallized at deeper structural levels under oxidized conditions; and (vi) favorable climatic conditions which allowed the oxidation of uraninite in the weathering zone and precipitation of uranophane in fractures enriching the upper part of the deposit.

Many other occurrences of uraninite-rich pegmatoids are known worldwide but all deposits which are being mined or which will be mined in a near future (e.g., Usab)

TABLE II. – Genetic classification of the uranium deposits [modified from Cuney, 2010].

	Deposit type	size	grade %	type example	Reference
M	**MAGMATIC**				
MCF	Crystal fractionation	small to medium	0.04	Ilimaussaq	Sørensen [1974]
MPM	Partial melting	large	0.01-0.05	Rössing [Namibia]	Berning *et al.* [1976]
H	**HYDROTHERMAL**				
HV	Hydrothermal-Volcanic	small to very large	0.05-0.2	Streltsovkoye [Russia]	Chabiron *et al.* [2003]
HG	Hydrothermal-Granitic	small to large	0.1-0.6	Niederschema-Alberoda [Germany]	Golubev *et al.* [2000]
HD	Hydrothermal-Diagenetic				
HDIa	*- intraformational redox*				
HDIaTb	** Tabular*	small to large	0.1-0.2	Grants [USA]	Hansley and Spirakis [1992]
HDIaTl	** Tectonolithologic*	small to large	0.1-0.4	Arlit-Akouta [Niger]	Pagel *et al*. [2005]
HDIaCb	** Collapse breccia*	Small	0.1-0.9	[USA]	Wenrich and Titley [2009]
HDBb	*- Basin/basement redox*	small to very large	0.2-20%	McArthur River [Canada]	McGill *et al.* [1993]
HDIf	*- Interformational redox*	small to medium	0.1-0.4	Oklo [Gabon]	Gauthier-Lafaye [1986]
HMp	Metamorphic hydrothermal				
HMt	Metasomatic-hydrothermal				
HMtAm	*- Alkali-metasomatism*	small to large	0.1-0.25	Michurinka [Ukraine]	Scherbak and Bobrov [2005]
HMtSk	*- Skarns*	small	0.1	Mary Katheleen [Australia]	Maas *et al.* [1987]
E	**EVAPOTRANSPIRATION**	small to large	0.014-0.1	Yeliree [Australia]	Carlisle *et al.* [1978]
S	**SYN-SEDIMENTARY**				
SMs	*- mechanical sorting*	large	0.02-0.15	Witwatersrand [South Africa]	Frimmel *et al.* [2005]
SRt *SRtm* *SRtc*	*- redox trapping* ** marine [black shales]* ** continental [coal, lignite…]*	very large small to medium	0.005-0.03 0.001-0.05	Randstatt [Sweden] Kazakhstan	Andersson *et al.* [1985] IAEA [2006]
SCcr	*- crystal-chemical/redox [phosphates]*	very large	0.005-0.03	Morocoo, Florida	IAEA [2001]
G	**GROUND WATER INFILTRATION**				
MB MRf	** basal type* ** roll front*	small to large small to very large	0.01-0.1 0.01-0.2	Blizzard [Canada] Wyoming	Boyle [1982] Adams and Cramer [1985]
O	**OTHERS**	very large	0.01-0.04	Olympic Dam [Australia]	Hitzman and Valenta [2005]

are located in Namibia, except for the much smaller deposits of the Bancroft district in Canada which have been mined in the seventies and eighties.

Hydrothermal deposits

For many hydrothermal deposits, the source of U is represented by plutonic rocks, either L-type peraluminous leucogranite or high-K calc-alkaline granites or equivalent volcanic rocks. Peralkaline plutonic rocks do not represent a significant U source, but peralkaline volcanic rocks and especially peralkaline tuffs are the major U source for a variety of deposits.

Hydrothermal granitic U deposits derive their uranium mostly from L-type peraluminous leucogranites [Cuney, 1978; Friedrich *et al.*, 1987]. The largest province of this type is the mid-European Variscan belt with uranium deposits associated with Carboniferous granite plutons which extends for over 2,000 km from Morocco to the Erzgebirge. Similar uranium provinces are known in southeastern China with the Jurassic to Cretaceous granite plutons of the Yanshanian belt and in Argentina with the Achala Batholith of Devonian age.

In the French part of the Variscan orogen, peraluminous two-mica leucogranite emplaced between 335 and 310 Ma. They are derived entirely from the partial melting of the continental crust [Bernard-Griffiths *et al.,* 1985] during the collision between the Eurasian and African continental plates at about 400 Ma. The "fertile granites" in which U is dominantly hosted in uraninite are those emplaced along a high heat flow and heat producing (HHFHP) belt spreading from northern Brittany to the Limousin area. The (HHFHP) belt corresponds to a 15 km thick crustal block enriched in U, Th and K during the Neoproterozoic to lower Paleozoic [Vigneresse *et al.*, 1989]. Leucogranites located outside of the HHFHP belt have low U content and are not associated with U deposits. Therefore, the genesis of the fertile leucogranites clearly requires the partial melting of a U enriched source. The Sr and Nd isotopic studies of Turpin *et al.* [1990] have shown that Late Proterozoic to Early Paleozoic felsic orthogneisses represent a possible source lithology for the Saint Sylvestre leucogranite. More particularly, the potassic calc-alkaline orthogneiss of La Dronne from central Limousin, with an average of 6.9 ppm U represents a likely protolith [Bourguignon, 1988].

The U deposits of the French Variscides are dominantly located within the granites, or in their enclosing metamorphic rocks, in the Erzgebirge district. They occur as veins or as disseminations in de-quartzified granite (episyenite) [Leroy, 1978; Cathelineau, 1986]. They are predominantly of Permian in age. The deposition of U in veins is related to low temperature hydrothermal fluid circulation, which largely postdates the emplacement of the granites by 30 to 40 Ma [Leroy and Holliger, 1984; Cathelineau *et al.*, 1989]. Despite this large gap, the localization of the U deposits is strongly controlled by the magmatic structures which were active during the emplacement of fine grained U-rich granite intrusions which were reactivated as brittle structures to channel hydrothermal fluids [Cuney *et al.*, 1989; Cuney, 1990]. The importance of U leaching from peraluminous leucogranites by hydrothermal fluid for the formation of U deposits has been recently emphasized by an oxygen and $^{40}Ar/^{39}Ar$ isotopic study on the Questembert granite [Tartèse *et al.*, 2013].

High-K calc-alkaline granites may also represent a major source of uranium for hydrothermal deposits. At Hotagen, in Sweden, U-deposits occur within a Paleoproterozoic high-K calc-alkaline granite but where uranium was subsequently precipitated during a Caledonian tectonic-hydrothermal event. 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 uranium. Another type of relation between the granites and the uranium source is provided by the Bois Noirs-Limouzat deposit in France [Cuney, 1978], with 6,920 t U at grades of 0.27% U. The deposit is hosted within the Bois Noirs high-K calc-alkaline granite, but drill holes below the deposit have intersected uraninite-rich peraluminous leucogranites, which probably represent the major uranium source for the deposit [Poty *et al.,* 1986]. The Olympic Dam deposit in southern Australia represents an even more complex relation between the host high-K calc-alkaline granite and later fluids, and is discussed further below.

Hydrothermal volcanic uranium deposits are mostly related to peralkaline volcanic rocks. The world largest uranium district of this type (280,000 t U at 0.2%) is the late Jurassic Stretsovkoye caldera in Transbaikalia, Russia. Hydrothermal circulations occurred during the waning stages of magmatic activity within a large caldera. The exceptional size of the resources in the Streltsovkoye district results from the juxtaposition of four 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 [Chabiron *et al.*, 2003], (iii) Ordovician U mineralization in the basement [Chernyshev and Golubev, 1996], and (iv) fluids expelled from the volcanic melts or from underlying magma chamber. The latter parameter has a limited effect, however, because the U fluid/peralkaline melt partition coefficient is strongly in favour of the melt (KDU fluid/melt = 3.10^{-2} to 4.10^{-2} [Peiffert *et al.*, 1996]). A quantitative estimate of the amount of uranium, which has been liberated by the liparites has been obtained from mass balance calculations between the uranium content of the melts inclusions from quartz trapped in the liparites and the average present U content of these volcanic rocks Chabiron *et al.* [2003].

High-K calc-alkaline metaluminous volcanic rocks are generally a less favorable U source because a significant but variable portion of the U in these rocks tends to be trapped in accessory minerals [Leroy and George-Aniel, 1992]. Most deposits related to this type of volcanism have a relatively small size.

Highly peraluminous acidic volcanics, mineralized in U are essentially known in the Macusani district, Peru. Pitchblende and autunite occur in sub-vertical to sub-horizontal fractures in the top tens of meters of Pliocene crystal-rich flows / tuffs. A resource of 30,000 t U has been estimated for the whole Macusani district at an average grade of 0.1% U [IAEA, 2009].

Hydrothermal diagenetic U deposits of tabular or tectonolithologic type may derive a large part of their

uranium from volcanic tuffs, commonly of peralkaline origin, deposited within continental siliciclastic units of large sedimentary basins and leached by saline diagenetic fluids. In the Arlit uranium district in Niger, the evidence of an important contribution by volcanic tuffs in the sandstone of the Tim Mersoi basin is provided by the presence of volcanic shards in the sandstone, of magmatic inclusions in the detrital quartz of volcanic origin, and of analcimolite levels. The analyses of the magmatic inclusions indicate that a large part of the volcanics were peralkaline and rich in U [Forbes *et al.*, 1984; Pagel *et al.*, 2005]. The important uranium contribution from peralkaline volcanic origin in the Tim Mersoi basin probably explains the relatively high grade of the Arlit district deposits with more than 100,000 t U at about 0.3%.

For the Lodève hydrothermal-diagenetic deposit of tectonolithologic type, in France, Ahmadach *et al.* [1993] have also determined the parent magma chemistry and initial U content of the volcanics ash layers which are present through the Lodève basin from the study of magmatic inclusions in apatite. The volcanic ash layers are presumed to represent of major U source for the deposit.

Hydrothermal diagenetic U deposits with basement/basin redox control, which are generally called unconformity related deposits, are characterized by large resources (631,000 t U) and also comprised the highest grade uranium deposits in the world (e.g. 20% U average grade in the McArthur River deposit in the Athabasca, Canada). The source of uranium for these deposits is debated. In the case of the Athabasca, it is proposed that uranium is derived either from the sandstone basin [Ruzicka, 1996; Kotzer and Kyser, 1995; Fayek and Kyser, 1997; Kyser *et al.*, 2000] or from the basement lithologies [Tremblay, 1982]. A special emphasis has been put on the abundant uranium-rich peraluminous granites and pegmatites of the basement [Thomas, 1983; Madore *et al.*, 2000; Annesley et Madore, 1999; Hecht and Cuney, 2000; Mercadier *et al.*, 2013], which result from the partial melting of uranium enriched metasedimentary lithologies initially deposited in an epicontinental setting during the Paleoproterozoic [Cuney, 2010]. Other possible sources include a series of U-rich high-K calc-alkaline granites mainly emplaced in the Taltson belt, to the west of the Athabasca basin [Brouand *et al.*, 2003]. It has been shown that, due to the exceptionally aggressive nature of the hydrothermal diagenetic fluids, which are highly saline, very acidic, relatively high temperature and oxidizing [Derome *et al.*, 2005; Richard *et al.*, 2012], a refractory mineral like monazite can be totally destroyed and uranium can be liberated both in crystalline basement rocks and within the sedimentary basin [Hecht and Cuney, 2000; Cuney and Mathieu, 2000; Gaboreau *et al.,* 2007].

Hydrothermal metasomatic U deposits associated with Na-metasomatism correspond to regional scale alteration, controlled by deep crustal structures, and typically characterized by dequartzification, albitization and later Ca- and less commonly K-metasomatism [Cuney *et al.*, 2012]. 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 high-K calc-alkaline granites as at Lagoa Real in Brazil (100,000 t U at 0.12%) [Turpin *et al.*, 1988], at Novokonstantinovkoe in Ukraine [Cuney *et al.*, 2012] and at Kurupung in Guyana [Cinelu *et al.*, 2006; Alexandre, 2010], or felsic metavolcanics of high-K calc-alkaline typology in the Michelin deposit in Labrador (36,800 t U at 840 ppm), Canada [Gandhi, 1978], and in the U deposits of northern Sweden such as Pleutajok (4,000 t U at 0.10%) [Adamek and Wilson, 1977].

Hydrothermal metasomatic U deposits associated with skarns. The type example of this category is the Mary Kathleen U-REE skarn in Australia (8550 t U at 0.11%). The skarns result from interaction between the volatile- and U-Th-rich high-K calcalkaline Burstall Granite emplaced at 1737 ± 15 Ma, and the enclosing calc-silicates, as well as highly saline fluids derived from evaporites [Oliver *et al.*, 1999]. An early phase of U and REE enrichment is presumed to have occurred in the skarns, at or near the present orebody and related in time to the emplacement of the granite [Maas *et al.*, 1987]. However the main phase of U-REE mineralization was generated during a second episode dated at 1550 ± 15 Ma by Page [1983]. This episode has occurred under upper amphibolite facies conditions (600-650°C, 3.5-4 kb) with a new phase of highly saline hydrothermal activity producing intense scapolitisation of the sediments. Small, disseminated grains of uraninite enclosed in allanite or along veins are exclusively associated with zones of retrogressed garnet ± diopside skarns [McKay and Miezitis, 2001]. The present U-REE mineralization at Mary Kathleen seems to result from the recrystallization of an older, granite-related U-REE mineralization in skarns, that were upgraded during the later metamorphic hydrothermal event.

Olympic Dam deposit

The Olympic Dam iron oxide-copper-gold (IOCG-U) deposit is by far the world's largest uranium resource (2,200,000 t U at 230 ppm). Uranium is a co-product of copper and gold. The deposit occurs in the center of the high-K calc-alkaline Roxby Downs granite emplaced at 1.59 Ga at a very shallow structural level [Creaser, 1996]. This granite is the most fractionated intrusion, and thus the most enriched in U and Th, of the much larger Burgoyne batholith. Uranium in the Roxby Downs granite is dominantly hosted in uranothorite. Gawler Range volcanic rocks of similar geochemistry were extruded contemporaneously over large areas. However, the genetic model for the genesis of the uranium mineralization in the Olympic Dam deposit is still not very well understood. Unlike many other IOCG deposits, Olympic Dam is entirely hosted within U-rich high K calc-alkaline granites and volcanics. The most commonly accepted model links the multiple brecciation episodes and the hydrothermal activity at the origin of the deposit to the emplacement of the Roxby Downs Granite and extrusion of contemporaneous volcanics [Reynolds, 2000; Johnson and Cross, 1995]. Hitzman *et al.* [1992] attribute the genesis of the Olympic Dam ores to a hot, highly saline fluid derived from a granitic magma, which has mixed with an oxidized meteoritic fluid. However, the source of copper and gold requires other sources and hydrothermal circulation at a larger scale in the crust [Johnson and McCulloch, 1995].

Observations made by the author on a limited selection of Olympic Dam samples suggest that uranium minerals comprise minor amounts of euhedral uraninite crystals dispersed within the mineralized breccia, and a predominant pitchblende phase, either occasionally altered to coffinite or

primary, and U-Ti oxides, occurring mainly in veinlets [Cuney and Kyser, 1998]. The uraninite is interpreted as an early high temperature uranium phase of mineralization event related to the unmixing of a magmatic fluid derived from the Roxbydown granite. However, its limited abundance may only explain concentrations in the order of some tens of ppm as commonly observed in many IOCG deposits. The deposition of pitchblende and coffinite, which represent the major part of the uranium ore minerals, requires the circulation of oxidized low temperature hydrothermal fluids. For Hitzman and Valenta [2005], the leaching of U from the wall rocks by such hydrothermal fluids was considered to be the main source of U in the IOCG deposits, with an enrichment factor in the ore of 10 to 40. However, at the time of the early magmatic-hydrothermal fluid system, the main uranium bearing phase in the enclosing Roxbydown was represented by uranothorite generally considered to be a refractory uranium source in the presence of low temperature hydrothermal fluids. Hence, the associated hydrothermal fluid circulations have to have occurred at least in the order of 200 Ma after granite emplacement, the time necessary for metamictization of uranothorite and liberation of uranium. A more detailed study of the uranium mineralization processes is clearly needed for IOCG deposits in general and especially for Olympic Dam.

Other deposit types

The first uranium 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, accumulated placers alongside with other heavy minerals, from highly fractionated Archean high-K calc-alkaline granites and pegmatites described above. A controversy exists about the granitic versus hydrothermal orgin of the uraninite from these deposits, but the REE patterns recently obtained by in situ analysis on uraninite crystals clearly indicate their derivation from a granitic source [Cuney, 2010; Delpiné *et al.*, 2013].

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 high-K calc-alkaline granites [Stuckless and Nkomo, 1978] or from interstratified volcanic ash [Zielenski, 1983]. Similar processes apply to the calcrete hosted deposits formed by evapotranspiration processes, with the type example represented by the Yeelerie deposit in western Australia [Carlisle *et al.*, 1978]. The relation between granites is still more direct in the case of the U-deposits of paleovalley type from the Vitim district in Russia, which occur in organic matter bearing sandstone deposited in narrow valleys directly incised into high-K calc-alkaline granites [Kondrat'eva *et al.* 2004].

For other uranium deposit types their relation with an igneous uranium source is more tenuous. Large U grade differences exist between the different occurrences of black shales and phosphorite-hosted deposits of the world. The high U content of the Cambrian Alum shale of Sweden, 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

Uranium 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 uranium leaching by a variety of hydrothermal and surficial fluids, much later than the emplacement of the igneous rocks from which the uranium is derived.

Uranium enrichment above the Clarke value is necessary for a granitic or volcanic rock to represent a viable uranium source for the formation of uranium deposits, but is not sufficient. In addition, however, uranium has to be hosted in a site from which it can be leached by oxidized hydrothermal fluids or ground waters. Uraninite represents the most easily leachable uranium source – it occurs mainly in highly fractionated peraluminous leucogranite and related pegmatites, in weakly peraluminous anatectic pegmatoids resulting from low degree of partial melting of metasedimentary rocks 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, Nb-Ti oxydes can be leached easily only when the structure of these minerals becomes metamict. Leaching of uranium from other accessory minerals like monazite is generally not possible other than under exceptional conditions such as the hydrothermal diagenetic deposits related to Proterozoic unconformities, where highly saline, acidic and relatively hot oxidized diagenetic fluids circulate.

Further research is needed to clarify a series of questions concerning the relations between felsic igneous rocks and uranium deposits. A crucial point is the understanding of the mechanisms leading to the genesis of Mesoarchean high-K granites able to crystallize uraninite and containing uranium contents similar to those reached during later geologic times. The genesis of uranium mineralization at Olympic Dam –the world largest uranium deposit– would require a systematic study of the distribution of uranium within the deposits, in relation to the tectonic structures and the redox zonation, and a precise dating of the different generations of uranium minerals. At the mineral scale a better quantification of the time required for the metamictisation of uranothorite, and hence its ability to act as a viable source of uranium in secondary deposits, is also required so that the role that high K calc-alkaline granites play in uranium fertilization is better known.

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