

The extreme diversity of uranium deposits

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Abstract Most available classifications of uranium deposits are based on the characteristics of the host rocks or on the morphology of the ore deposits. The aim of the present paper is to propose the basis for a genetic classification of these deposits. After a short introduction on the geochemical behavior of uranium in fluids and silicate melts and on the main uranium fractionation mechanisms operating in uranium-rich peraluminous, metaluminous, and peralkaline melts, the most recent metallogenic models of the main types of uranium deposits are shortly reviewed.

Introduction

Uranium deposits formed at nearly all steps of the geological cycle, from high-grade metamorphic conditions (up to 800°C, 5 to 7 kbar), plutonic, metasomatic, hydrothermal, basin diagenesis, metamorphism, volcanic to sedimentary and superficial environments (Dahlkamp 1993). They formed from Neoproterozoic times to Quaternary, but none is expected to exist before about 3.1 Ga. Large deposits exist on all continents, but the largest resources are known in Australia, dominated by the huge Olympic Dam deposit. The current world uranium resource (reasonably assured+inferred resources) is estimated at 5.5 Mt U (Table 1), of which 873 deposits with estimated resources over 500 t U are listed in the UDEPO database (<http://www-nfcis.iaea.org>). Among those, three types contain more than three quarter of the worldwide uranium resour-

ces: unconformity-related deposits, IOCG (iron oxide–copper–gold) deposits, and sandstone-hosted deposits (Table 1). Important past or current U production also comes from a variety of additional deposit types: quartz–pebble conglomerates, veins, volcanic-related, intrusive, metasomatic. Other types present either smaller resources such as the calcrete, breccia pipe, and metamorphic deposits, or very large, but low-grade resources (unconventional resources of the IAEA) with more than 7.6 Mt U (IAEA 2008), such as sedimentary phosphates and black shales. Uranium enrichment in coal and lignite represents only potential resources.

Uranium solubility in fluids and silicate melts

Uranium geochemistry is mainly governed by oxidation state. U is highly mobile as hexavalent uranyl ion (UO_2^{2+}) under oxidizing conditions. It may form more than 40 complexes with hydroxyl, carbonate, sulfate, chloride, phosphate, fluoride, and silicate anions (Langmuir 1978). Uranyl-carbonates predominate at high pH, low temperature, and intermediate to high $f\text{O}_2$. Phosphates predominate at neutral pH. Sulfates and chlorides are important at acid pH. In reduced conditions, U^{4+} solubility is extremely low and similar to that of Th. High U^{4+} and Th solubility is limited to high temperature and mainly controlled by fluoride complexes. Precipitation of U in most deposits is related to a decrease of $f\text{O}_2$, generally resulting from the interaction of oxidized U-bearing fluids with carbonaceous matter under various states: from anaerobic bacterial activity to graphite. Other potential reductants are H_2S , magnetite, ilmenite, and sulfides.

Thermodynamic data on U species are mostly available at low temperature (Grenthe et al. 1992).

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Table 1 Historic world uranium production up to 2007 (compilation from various sources), Reasonably Assured Resources (RAR) and Inferred Resources (IR) from IAEA (2008)

	Past production Including 2007	RAR <130 USD/kg U	IR <130 USD/kg U	Total
Unconformity	348,000	491,600	158,100	997,700
Sandstone	665,000	999,500	524,400	2,188,900
IOCG	42,000	499,400	401,500	942,900
Paleoplacer	246,000	163,600	138,300	547,900
Vein	376,000	800,000	167,700	1,343,700
Intrusive	95,000	183,700	104,200	382,900
Volcanic	234,000	157,800	53,500	445,300
Metasomatite	30,000	304,900	368,800	703,700
Other	171,000	284,300	154,400	609,700
Unspecified	70,000	96,700	59,700	226,400
TOTAL	2,277,000	3,338,300	2,130,600	7,745,900

Uranium and other large highly charged cations (Th^{4+} , Zr^{4+} , REE^{3+}) dissolve in silicate melts according to the degree of melt depolymerization (Peiffert et al. 1996). Depolymerization depends on the temperature and excess of alkalis and Ca relative to Al. Increasing one of these parameters enhances melt depolymerization and consequently the solubility of large highly charged cations. At 770°C, 2 kbar, and variable $f\text{O}_2$ conditions, haplogranitic melt and coexisting aqueous fluid compositions, melt apacity ($\text{Na}+\text{K}/\text{Al}$ in cations) is the most significant factor controlling U solubility in silicate melts (Peiffert et al. 1996). U solubility increases from ppb to percent levels with increasing apacity from 0.7 (peralkaline) to 1.6 (peraluminous). Oxygen fugacity increase from the Ni–NiO to Mn–MnO buffer increases U solubility only by a factor of 3. CO_2 or Cl in coexisting aqueous fluid has minimal effect on U solubility, because both CO_2 and Cl have a low solubility in felsic melts. However, fluorine is strongly partitioned into silicate melts, thereby depolymerizing them, and thus increasing U solubility.

Uranium fractionation in igneous rocks

Among felsic igneous rocks, three types may constitute “fertile” U sources for the genesis of uranium deposits with uranium contents well above the Clarke value (3–4 ppm): highly fractionated peralkaline and metaluminous high-K calcalkaline rocks as well as peraluminous igneous rocks derived from low degree of melting of supracrustals. A fourth type corresponds to granitoid dykes emplaced in migmatites, called alaskites, derived from still lower degree of partial melting of supracrustals, and of weakly peralu-

minous composition. Each of these rocks is characterized by a specific fractionation of Th and U, and a specific accessory mineral paragenesis.

Peraluminous leucogranites, classically referred to as S-type granites in the literature (White and Chappell 1977), result in fact from a variety of processes, and several subtypes have been distinguished (Debon and Lefort 1988). Only the peraluminous leucogranites, as defined thereafter are associated with large uranium deposits. They have a limited compositional range (biotite<10%) and display a strong increase of their peraluminosity index with fractionation, opposite to the S-type granites as defined by White and Chappell (1977). Early monazite and zircon fractionation, due to the low solubility of these two minerals in peraluminous low-temperature melts, leads to strong Zr, Th, and REE depletion. On further fractionation, U continues to be enriched, if the uranium content of the melt was significantly above Clarke abundance (Cuney et al. 1989), until uraninite saturation is reached. Th-poor uraninite then crystallizes which represents the most easily leachable uranium source (Cuney and Friedrich 1987). These granites are best exemplified by the peraluminous leucogranites from the mid-European Variscan belt. High-silica, highly peraluminous volcanic rocks equivalent to these leucogranites are quite rare. The main mineralized occurrences are the Cenozoic ignimbritic tuffs of Macusani in Peru (Arribas and Figueroa 1985).

Peralkaline granites, syenites and volcanic rocks (A1-type granites of Eby 1992) are always enriched in U simultaneously with Th and other large highly charged elements. Despite high U contents, peralkaline granitoids are not associated with significant vein-type deposits, but they may represent uranium sources when their U–Th minerals are metamict. However, peralkaline volcanic rocks represent excellent uranium sources because U is mainly hosted by glass which can be easily leached during devitrification.

High-K calcalkaline metaluminous granites and volcanic rocks (A2-type granites of Eby 1992) are enriched in U during magmatic fractionation, but Th, Zr and REE have a behavior either similar to that of peraluminous or of peralkaline melts, according to temperature and peraluminosity. Their high Ca contents (>1 wt.% CaO) induce the crystallization of Ca-rich minerals (amphibole, titanite, allanite) incorporating most REEs, but minor amounts of Th. Then, Th crystallizes as uranothorite with up to 30 wt.% UO_2 . Uraniothorite represents either a refractory uranium source for hydrothermal fluids circulating early after granite emplacement, or an easily leachable uranium source, when metamictized (Cuney and Friedrich 1987). When the Th/U ratio of the melt decreases sufficiently, small amounts of uraninite may crystallize in highly fractionated high-K calcalkaline granites. Deposits associated with such granites

and volcanic rocks generally have small resources, except for the Olympic Dam deposit (see below).

A genetic classification of uranium deposits

A classification of the different types of uranium deposits is presented according to formation conditions through the geological cycle (Fig. 1).

Uranium deposits related to surface processes

These deposits correspond to syn- to early epigenetic near-surface uranium concentrations formed during intracontinental sedimentation and weathering.

The paleoplacer uranium deposits are the first formed on Earth. The earliest ones are hosted by the Dominion Group (3.1 Ga) in South Africa and the latest ones by the Elliott Lake Group (2.3 Ga) in Eastern Canada. They correspond to syngenetic detrital accumulation of uraninite in fluvial to fluvio-deltaic environments (Mellor 1916), with more or less intense remobilization of detrital uraninite according to the modified placer theory (Pretorius 1961).

At the opposite end of time-bound ore formation are the uranium deposits associated with calcretes which formed from Tertiary to Present time, by evapotranspiration processes in fluvial to playa systems, in arid to semi-arid climatic conditions. The economically important U deposits of this category, Yeelirrie in Australia and Langer Heinrich in Namibia, are hosted by highly immature, porous, fluvial valley-fill sediments (Carlisle et al.

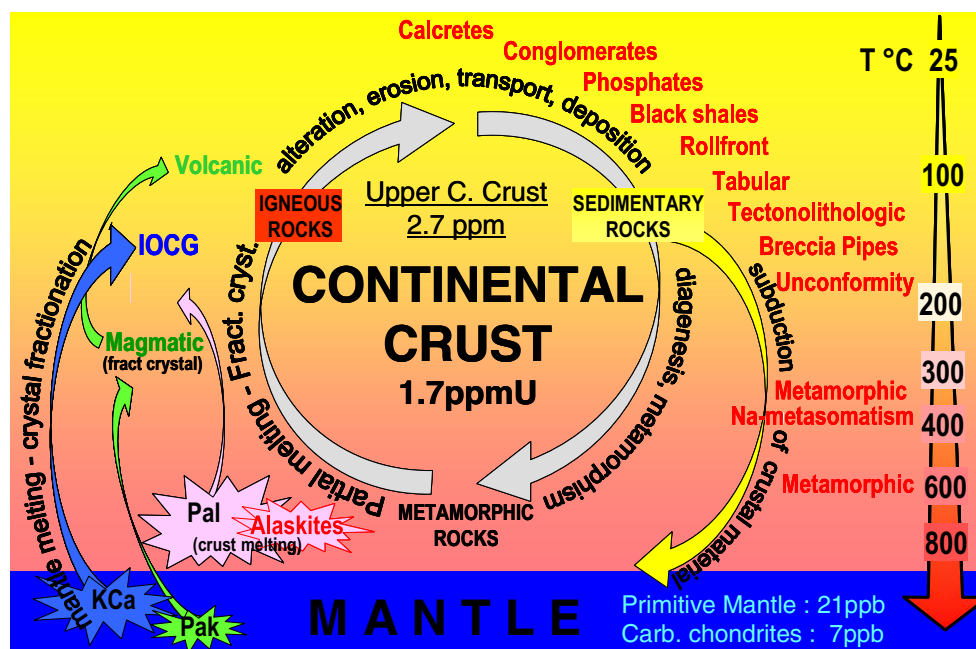
1978). Uranium is entirely deposited as (UO_2^{2+}) minerals. The predominance of vanadates (carnotite and tyuyamunite) in these deposits is due to their low solubility, two orders of magnitude lower than that of common (UO_2^{2+}) minerals in the pH range 5 to 8.5 (Langmuir 1978).

Lignite, organic-rich bog-peat, and closed anoxic lake and karst caverns represent other varieties of surficial uranium accumulation where uranium is deposited by adsorption on organic material and/or by anaerobic bacterial activity which produces gases (H_2S , CH_4 ...) capable of reducing (UO_2^{2+}).

Synsedimentary uranium deposits

Synsedimentary uranium deposits are formed during sedimentation in epicontinental platform environments and essentially correspond to the U-rich black shales and phosphorites. Phosphorite deposits formed along shallow continental shelves with restricted circulation. U^{4+} is a proxy for Ca^{2+} in the apatite structure, but biologic activity may create reducing environments where U may precipitate outside of the apatite structure. The largest episode of phosphate deposition occurred during the late Cretaceous to Eocene, under a common paleolatitude ($8-15^\circ \text{N}$), along the southern margin of the Tethys Ocean. The phosphorite belt extends from Turkey to Morocco, through Israel, Jordan, Syria, Iraq, Saudi Arabia, Egypt, Tunisia, Algeria, and beyond the Atlantic to Colombia and Venezuela. Morocco hosts three quarters of the world resources of this type, but most of the historical uranium production (17,150 t U from 1954 to 1992) was

Fig. 1 Position of uranium deposit types with respect to the main fractionation processes along the geological cycle. The different types of U-rich magmas are indicated. *Pak* peralkaline, *KCa* High-K calcalkaline, *Pal* peraluminous



extracted from Miocene–Pliocene phosphorites of Florida (Cathcart 1978).

Uraniferous black shales form in shallow marine environments, in which U is syngenetically deposited, adsorbed onto organic material and clay minerals. Preferential U enrichment occurs in the vicinity of paleo-shores where clastic input is limited and where vigorous bottom-water circulation promotes high rates of mass-transfer across the sediment/water interface (Schovsbo 2002). The largest deposits are in the Cambro-Ordovician shale of Ranstad, Sweden, with 254,000 t U at 170–250 ppm U, but the Silurian graptolitic shales of Ronneburg-Gera, Germany, with a resource of 169,230 t U at 850–1,700 ppm U, are the only ones to have been mined, because of their higher grades due to a combination of synsedimentary uranium deposition, late Variscan hydrothermal and recent superegene enrichment (Urban et al. 1995).

Uranium deposits related to hydrothermal processes

They correspond to a very wide variety of ore deposit subtypes in the uranium geology literature. They are typically epigenetic and formed during fluid circulation through porous and sometimes fractured fluvial, lacustrine, deltaic to near-shore siliciclastic formations, occasionally in limestones, or through fractured granitic, volcanic, or metamorphic rocks. Uranium can be transported by various fluids of meteoric, diagenetic, and/or metamorphic origin.

Basal-type uranium deposits are transitional between surficial and diagenetic–hydrothermal types. They occur in poorly sorted and consolidated, highly permeable, fluvial to lacustrine carbonaceous gravels and sands deposited as thin, 10 to 15-km elongated bodies along paleovalleys incised in basement rocks and capped by plateau basalts. U is leached from the granitic basement and precipitates by reaction with the organic matter during groundwater percolation in permeable sediments, between impermeable basement and basalts. The Blizzard deposit in Canada is a typical example of this type (Boyle 1982). The basal type is similar to the “paleovalleys or infiltration type” uranium deposits in Russia.

Tabular uranium deposits are transitional with the synsedimentary and diagenetic–hydrothermal deposits, because U minerals may begin to precipitate shortly after sedimentation and burial, but more commonly during diagenesis. Ore bodies form within sandy layers intercalated between non permeable clay horizons, generally at paleo-channel margins. Volcanic ash within the sandstone is a major source of U. They are generally also rich in vanadium. In the Grants region, Colorado, where over 240,000 t U at 0.09–0.21 % have been mined, Hansley and Spirakis (1992) propose that U transported by brines

expelled from underlying evaporitic sediments, precipitated on formerly deposited humates derived from terrestrial organic matter. A higher temperature evolution due to deeper burial may cause the apparent paucity in organic matter of some of these deposits as in the Henry Mountains district, Colorado.

Roll-front deposits represent the best example of epigenetic uranium deposition at a redox interface (front). Host rocks are commonly younger than Ordovician and were deposited in fluvial and lacustrine environments, or in marine marginal plains, in channel, lagoonal, and beach-bar settings. Both volcanic ash in host rocks and external U-rich granites or volcanic rocks may represent uranium sources. Oxidized low-temperature meteoric waters infiltrated permeable rocks, after some diagenesis had occurred (Finch and Davis 1985). Uranium ore location is mainly controlled by the interplay of the permeability of the sediments and the proportion of reducing components: detrital continental carbonaceous matter, sulfides, hydrocarbons, or hydrogen bisulfide migrated from deep oil reservoirs (Cai et al. 2007), and/or interbedded mafic volcanic rocks. The orebodies are crescent-shaped in cross section, and sinuous along the roll-front interface. In the Inkai deposit, Kazakhstan, they may extend laterally up to more than 100 km. The classical roll-front deposits occur in intermontaine basins, as in the Tertiary Powder River Basin, Wyoming.

Tectonic–lithologic deposits are also hosted by sandstones but fluid percolation is strongly controlled by faults. Typical examples are the deposits from the Arlit area in Niger (Pagel et al. 2005). The processes controlling ore deposition are similar to those described for the tabular uranium deposit type.

Solution-collapse breccia pipes correspond to near-vertical, 30–175 m large, cylindrical columns, located in flat-lying upper Paleozoic to Triassic marine platform sediments in the Grand Canyon region, USA (Wenrich and Sutphin 1989). Collapse may have propagated upward into overlying strata up to 1,000 m. Thousands of pipes are known but only about 100 of them are variably mineralized. The mineralization is associated with low temperature (80–173°C), saline (4 to 17 wt.% equiv. NaCl) oxidized solutions of diagenetic origin, derived from deeper parts of the basin.

Unconformity-related deposits are the most typical diagenetic–hydrothermal uranium deposits. Uranium deposition was focused at the interface between a thick, Paleozoic to Meso-Proterozoic sandstone cover and an Archean to Paleoproterozoic crystalline basement, where graphite-rich faults were reactivated. The debate is still open between the supporters of uranium extracted only from the basin (Fayek et al. 2002) and those making the U-rich metamorphic basement the largely dominating source (Hecht and Cuney

2000; Madore et al. 2000). Mineralogy and analysis of single fluid inclusions show that the slightly acidic (kaolinite–illite equilibrium), hot (160 to 220°C, about 1 kbar), oxidized (fO_2 in the hematite stability field) and Na–Ca-rich (up to 5 mol of chlorides) diagenetic brines, generated within Paleo- to Meso-Proterozoic continental, organic-free, sandstone formations, were progressively enriched in calcium during their percolation through the basement (Derome et al. 2005). The resulting Ca–Na brine was an exceptionally aggressive fluid even for highly refractory U-bearing accessory minerals. First analyses of U contents in fluid inclusions by laser ablation ICP-MS showed that the highest U-concentrations occur in Ca-dominated brines (Richard et al. 2008). Exceptional trapping conditions resulted from the strong redox gradient between the oxidized sandstone cover and the graphite-rich meta-sedimentary rocks of the basement and the openings created in the sandstone and the basement by the combined effects of reverse tectonics and quartz dissolution (Lorilleux et al. 2002). Dating of the least-altered samples, using SIMS or laser ablation ICP-MS, has given a wide spectrum of ages discussed by Alexandre et al. in this volume.

Synmetamorphic uranium deposits are formed during the circulation of metamorphic fluids in association with folding, faulting, and/or thrusting of the rocks. The most favorable conditions correspond to the lowest grade of metamorphism of epicontinental platform sediments, during which the most important release of fluids occurs, and which may expel both oxidized brines from evaporitic layers efficient for uranium transport, and hydrocarbons produced by black shales, efficient for precipitating uranium. Typical example are the Mistamisk veins from Labrador, Canada (Kish and Cuney 1981), and the Kansanshi deposit from Zambia (Kříbek et al. 2005) which both formed at $350 \pm 50^\circ\text{C}$.

At higher temperature the metamorphic origin of the deposits is rarely well constrained.

Metasomatic uranium deposits are mainly associated with Na-metasomatism. The alteration may result from a large variety of processes from the interaction of magmatic fluids exsolved from a peralkaline granite as at Bokan Mountain, Alaska (MacKevett 1963), to lower temperature fluids derived from a basinal brine or a magma as proposed for the Valhalla uranium deposit, Australia (Polito et al., this volume), or still to fluids with seawater composition as at Lagoa Real (Lobato et al. 1983). In the largest districts albitites form discontinuous occurrences over several tens of kilometers. Individual Na-metasomatic zones are several meters wide and several hundred meters long. Some authors consider that these deposits are related to metamorphic processes (Lobato et al. 1983), others argue for hydrothermal circulation occurring after granite emplacement (Turpin et al. 1988).

Uranium-mineralized skarns are another type of deposit related to metasomatic processes. The Mary Kathleen skarns are a typical example (Page 1983). An extreme type of metasomatic uranium deposit is represented by the pyroxenites of the Tranomaro region, Madagascar. The main stage of skarn formation and mineralization, with mainly meionite in the endoskarns and Al-diopside–spinel–corundum–uranothorianite in the exo-skarns records conditions of 5 kbar, 850°C (Moine et al. 1998). Fluid circulation responsible for metasomatism and U–Th mineralization is synchronous with granulitic metamorphism. Although CO_2 was a major fluid component in the metasomatic reactions, fluorine has played a major role in the complexation and simultaneous transport of Th, U, REE, and Zr in the skarns.

Vein-type deposits Uranium deposits related to granites are best exemplified by the mid-European Variscan uranium province, which extends over more than 2,000 km from Spain to the Bohemian Massif. The uranium deposits are essentially related to late Carboniferous peraluminous leucogranites. They are located either in the granites (French Massif Central) or in their metamorphic host rocks (Erzgebirge). Primary uranium deposition at the scale of the Variscan belt occurred 30 to 50 Ma after the emplacement of the granites, at the Stephanian–Permian transition (270–280 Ma; Holliger and Cathelineau 1986; Dolníček et al. and Kříbek et al., this volume) during a regional extensional event. The ore forming fluids are low-salinity and low-temperature fluids (Dubessy et al. 1987). Uranium deposition results from the mixing of oxidized meteoric fluids leaching uraninite from granites with fluids derived from an overlying basin and which have provided the reductants for uranium deposition (Turpin et al. 1990), a model similar to the one proposed by Dolníček et al., and Kříbek et al., this volume.

Uranium deposits related to volcanic rocks predominantly occur within wide calderas, filled with alternations of mafic and felsic volcanic rocks, and sedimentary layers. Rhyolitic pyroclastic tuffs represent the largest proportion of the extruded magmas. Peralkaline magmatism provides the most interesting uranium source as discussed above. The genesis of significant deposits also requires a relatively shallow magma chamber, lasting over several million years, and able to provide the heat flux necessary to promote focused and long lasting convective fluid circulation. The Streltsovskoye caldera (Transbaikalia, Russia), with 120,000 t U already mined and resources of 127,000 t U at 0.18% U (IAEA 2008) is by far the largest uranium ore field in the world of this type (Ishukova et al. 1991). The juxtaposition of two major U sources: U-rich peralkaline rhyolites and U-rich subalkaline granites in the basement, is one of the key parameters

contributing to the large size of the resources of Strel'tsovskoye (Chabiron et al. 2003).

High-K calcalkaline metaluminous volcanic rocks are less favorable because a large part of the U tends to be trapped in refractory accessory minerals (Leroy and George-Aniel 1992). But, Fe-rich highly fractionated high-K magmas, as the most fractionated volcanic rocks of the Gawler Range associated with the Olympic Dam IOCG deposit in South Australia, are more favorable. At Olympic Dam, hydrothermal activity is related to the ~1.6-Ga emplacement of the Roxby Downs granite and the extrusion of the Gawler Range volcanic rocks. Magmatic activity, brecciation, and mineralization are synchronous, and ore genesis is related to the unmixing of a hot, highly saline fluid from the felsic magma which mixed with oxidized meteoric water (Hitzman et al. 1992). Very little information is available about the genesis of the U mineralization. Leaching of U from the wall rocks is assumed to have produced uranium enrichment in the IOCG deposits, 10 to 40 times larger than in unaltered host rocks (Hitzman and Valenta 2005).

Deposits related to partial melting

Low-grade uraninite mineralization can occur disseminated in granitoids sheets and small plutonic bodies. They typically emplace in epicontinental sedimentary rocks (arkoses, quartzites, black shales, marlstone, limestones) metamorphosed to upper amphibolite facies with partial melting. The Rössing uranium deposit in Namibia is the largest deposit of this type (Berning et al. 1976). It has been proposed that these deposits result either from extreme fractionation of a deeper-seated granite body, as suggested for the mineralized pegmatites of the Grenville belt in Québec (Lentz 1996), or from partial melting of U-rich metasedimentary or metavolcanic rocks, as proposed for Rössing and the Mont Laurier pegmatoids, Québec (Cuney 1982). Although uraninite dominantly crystallized from the magma, part of the U enrichment results from the percolation of magmatic fluids and supergene processes.

Deposits related to crystal fractionation

This deposit style is only known in peralkaline complexes. The high solubility of U, Th, Zr, and REE in such melts leads to their continuous enrichment during magmatic fractionation, and finally to the crystallization of complex minerals that incorporate uranium together with these elements. Consequently, mineralizations associated with peralkaline rocks have rarely been mined because of the high cost of U extraction from refractory minerals. Another consequence is that the U mineralization is always

associated with the most fractionated units of peralkaline complexes, located at their apex or margin. The largest low-grade U resource related to fractional crystallization of peralkaline magmas is the Kvanefjeld deposit at Ilmaussaq, Greenland. U is mainly hosted by steenstrupine, a complex U–Th–REE silicophosphate, disseminated in peralkaline syenites (Iujavrite; Sørensen et al. 1974).

Conclusions

As a consequence of the considerable decrease of uranium exploration activities since the end of the 1980s, academic research of uranium metallogenesis has been mainly limited to high-grade unconformity-related deposits. New research developments in this field should allow a more advanced genetic classification of uranium deposits, than the one shortly proposed here, in order to integrate the exceptional diversity of processes involved in the formation of uranium deposits. Important progress in the understanding of the origin of uranium deposits is expected especially from in-situ analysis of trace elements and stable and radiogenic isotope composition of mineral phases and in fluid inclusions, and from experimental determination of uraninite solubility at high temperature and pressure with a variety of ligands.

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