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Evolution of Uranium Fractionation Processes through Time: Driving the Secular Variation of Uranium Deposit Types

MICHEL CUNEY†

Nancy Université, UMR G2R 7566 CNRS-CREGU, BP 70239–54 506 Vandoeuvre Cedex, France

Abstract

Uranium deposit types have evolved considerably from the Archean to the Present. The major global drivers were (1) change of geotectonic conditions during the Late Archean, (2) strong increase of atmospheric oxygen from 2.4 to 2.2 Ga, and (3) development of land plants during the Silurian. Other significant variations of uranium deposit types are related to unique conjunctions of conditions such as those during phosphate sedimentation in the Cretaceous. Earth's uranium fractionation mechanisms evolved through four major periods. The first, from 4.55 and 3.2 Ga, corresponds to formation of a thin essentially mafic crust in which the most fractionated trondheimite-tonalite-granodiorite (TTG) rocks attained uranium concentrations of at most a few parts per million. Moreover, the uranium being essentially hosted in refractory accessory minerals and free oxygen being absent, no uranium deposit could be expected to have formed during this period. The second period, from about 3.1 to 2.2 Ga, is characterized by several widespread pulses of highly fractionated potassic granite strongly enriched in U, Th, and K. Late in this period peraluminous granite was selectively enriched in U and to a lesser extent K. These were the first granite and pegmatite magmas able to crystallize high-temperature uraninite. The erosion of these granitic suites liberated thorium-rich uraninite which would then be concentrated in placer deposits along with pyrite and other heavy minerals (e.g., zircon, monazite, Fe-Ti oxides) within huge continental basins (e.g., Witwatersrand, South Africa, and Bind River, Canada). The lack of free oxygen at that time prevented oxidation of the uraninite which formed the oldest economic uranium deposit types on Earth, but only during this period. The third period, from 2.2 to 0.45 Ga, records increased oxygen to nearly the present atmospheric level. Tetravalent uranium from uraninite was oxidized to hexavalent uranium, forming highly soluble uranyl ions in water. Uranium was extensively trapped in reduced epicontinental sedimentary successions along with huge quantities of organic matter and phosphates accumulated as a consequence of biological proliferation, especially during the Late Paleoproterozoic. A series of uranium deposits formed through redox processes; the first of these developed at a formational redox boundary at about 2.0 Ga in the Oklo area of Gabon. All known economically significant uranium deposits related to Na metasomatism are about 1.8 Ga in age. The high-grade, large tonnage unconformity-related deposits also formed essentially during the Late Paleoproterozoic to early Mesoproterozoic. The last period (0.45 Ga–Present) coincided with the colonization of continents by plants. The detrital accumulation of plants within continental siliciclastic strata represented intraformational reduced traps for another family of uranium deposits that developed essentially only during this period: basal, roll front, tabular, and tectonolithologic types. However, the increased recognition of hydrocarbon and hydrogen sulfide migration from oil or gas reservoirs during diagenesis suggests potential for sandstone-hosted uranium deposits to be found within permeable sandstone older than the Silurian. Large uranium deposits related to high-level hydrothermal fluid circulation and those related to evapotranspiration (calcretes) are only known during this last period of time, probably because of their formation in near-surface environments with low preservation potential.

Introduction

URANIUM ore deposits present the most extreme diversity of concentration processes for a metallic element (Cuney, 2009). Uranium deposits are known at nearly all stages of the geologic cycle but are not known prior to 3086 Ma. Also, types of uranium deposits vary greatly from Mesoarchean to Present (e.g., Bowie, 1979; Robertson et al., 1978; Meyer, 1981; Nash, 1981; Nash et al., 1981, Ferguson, 1987; Dahlkamp, 1993; Kyser et al., 2000), and uranium resources are unevenly distributed through geologic time (Figs. 1, 2). The variation of uranium deposit types through time has been related by most of these authors to two major changes in the geochemical cycle of uranium, one associated with the development of an oxygenated atmosphere at about 2.2 b.y. and the other with the development of land plants at about 0.4 b.y. Most of them also pointed out that a large proportion of the uranium deposits are hosted in Archean-Paleoproterozoic rocks anomalously enriched in uranium. However, it is now possible to better assess the mechanisms at the origin of the uranium enrichment of the Late Archean and Paleoproterozoic rocks, from the recent progress made in the understanding of the petrogenesis of these rocks. Moreover, the time distribution of the uranium resources proposed in the previous papers was based on drastically different numbers available during the 1980s or early 1990s which did not include the enormous resources of the Eastern countries (former USSR, Eastern Europe, and China). For example, the resources of the uranium deposits related to volcanic caldera systems have increased tenfold. The resources of sandstone-hosted deposits have increased by more of than 1 million metric tons (Mt) U with the addition of the Central Asia province including Kazakhstan and Uzbekistan, the second largest uranium province of the world after Australia. Also some major uranium deposit types were not very well known or even unknown, including the huge Olympic Dam (Australia) and Elkon (Russia) uranium deposits.

† E-mail, Michel.Cuney@g2r.uhp-nancy.fr

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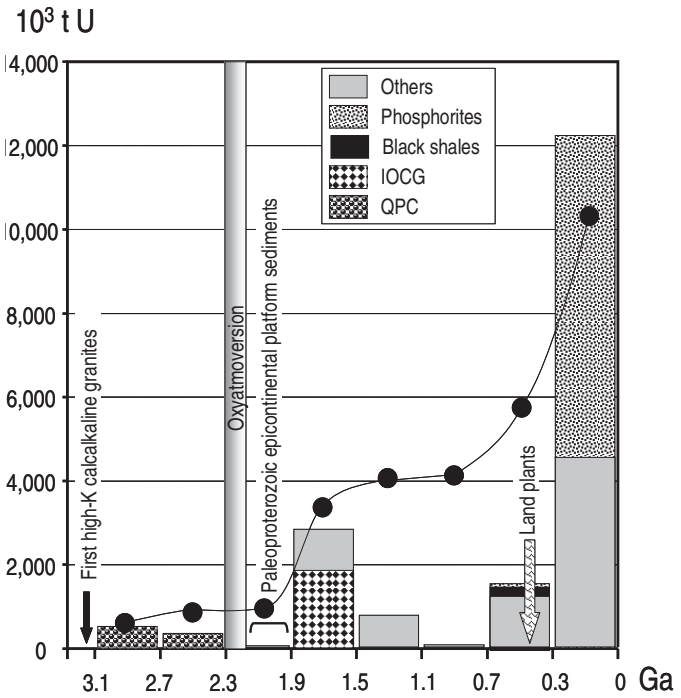


FIG. 1. The uneven distribution of the uranium resources through geologic time. Some deposits types with large resources have been distinguished (QPC = quartz pebble conglomerates; IOCG = iron oxide-copper-gold, mainly represented by Olympic Dam; black shales, mainly represented by the alum shales of Sweden; sandstone = regroup all hydrothermal diagenetic deposits with intraformational redox control and surficial deposits such as calcretes and lignite, which represent nearly 500 deposits; phosphorites and other types. Source of the data: International Atomic Energy Agency redbooks and database, including more than 1,200 deposits. The cumulative present-day age distribution curve of uranium resources is represented. The curve does not take into account the uranium resource from phosphate deposits for the 0.3 to 0 Ga period.

Dahlkamp (1993), who provided the most recent synthesis for uranium deposits, pointed out four parameters driving the time-related change of uranium deposit types: (1) uranium deposits are restricted to distinct epochs in the Earth's history, (2) uranium-rich source rocks prevailed during distinct geologic times, (3) sediment- and metasediment-hosted uranium deposits generally have a distinct spatial affinity with the uranium-rich source rocks mentioned above, and (4) uranium deposits can be grouped into five distinct generations. However, these generations are not always related to significant change in the global geologic conditions on the Earth affecting uranium fractionation. Generation one corresponds to the 2.8 to 2.2 Ga period with the quartz pebble conglomerates assumed to be of detrital origin because of the low oxygen level of the atmosphere. Generation two (2.2 to 1.9–1.7 Ga) corresponds to the increase of the oxygen level and the solubilization of hexavalent uranium leading to the formation of the deposits associated with the Franceville basin, to metamorphosed deposits and some vein-type deposits. Generation three (1.5–0.9 Ga) comprises mainly the unconformity-related deposits and minor deposits related to uranium dissemination in intrusive rocks (Bancroft and Kvanefjeld). Generation four (0.7–0.5 Ga) is considered as mainly synorogenic and includes the uranium deposits of the Damara-Katanga orogen: Rössing and the Shaba province. However these types of deposits are not specific to this period. Generation five (0.5-0.4 to 0 Ga), also recognized by most previous authors, is related to the development of land plants with the sandstone-hosted and surficial deposits. Most vein- and volcanic-related deposits are also included here.

The aim of the present paper is to review the fundamental parameters controlling variations in uranium fractionation processes leading to the formation of the various types of uranium deposits throughout Earth's history. Therefore, for each deposit type, focus is given on the major change in the uranium fractionation processes and not on a comprehensive presentation of the mechanisms leading to the formation of each type of deposit. Such information and more detailed references are available in Dahlkamp (1993) or Cuney and Kyser (2008). Most of the changes in the types of uranium deposits through time can be attributed to major changes in the geodynamic evolution of the Earth—in magmatic or fluid fractionation processes, in the composition of the atmosphere, and in the nature of life. However, for some uranium deposit types, such as those related to sodium metasomatism, the reasons for their appearance at a much higher frequency during well-defined periods of Earth history are still poorly understood. Only four major periods, corresponding to key steps in uranium fractionation processes, and covering geologic periods close to the ones proposed by Nash (1981), will be considered here. The limits of these periods have taken into account new age determinations on the sedimentation of the Dominion Reef Formation, the emplacement of the first potassic granites able to crystallize uraninite, and the development of land plants.

Uranium Fractionation in the Early Earth (>3.2 Ga)

Uranium in silicate magmas has a strongly incompatible behavior because of its large radius and high valence. Hence, during partial melting and crystal fractionation, uranium is continuously preferentially fractionated into high-temperature metaluminous melts, nearly the only type of melt on the early Earth. Thus, the first mechanisms of uranium enrichment were partial melting of the primitive mantle (U = 20.5 ppb, Javoy, 1998; Table 1) followed by crystal fractionation during ascent of melts to the surface. Fractionation of basic melts did take place during the Archean, but to a limited extent. Before the Mesoarchean (<3.2–3.1 Ga), these processes led to the formation of a relatively thin and dominantly mafic crust, composed essentially of komatiitic and tholeiitic basalt.

TABLE 1. U, Th, and K Abundances in the Earth's Main Envelopes[1]

	U (ppb)	Th (ppb)	K (ppm)	Th/U
Upper continental crust	2,800	10,700	28,000	3.8
Average continental crust	1,020	4,800	21,900	4.7
Primary upper mantle	20.5	85	40–80	3.92
Midocean ridge basalt	47	120	600	2.55
Ocean island basalt	1,020	4,000	12,000	3.92
CI carbonaceous chondrites	7.4	29	545	3.92

[1] Primary upper mantle and average continental crust: U and Th (from Javoy, 1995) and K (from Allègre et al., 1987), upper crust (from McLennan et al., 2001), midocean ridge basalt, ocean island basalt, CI: carbonaceous chondrites

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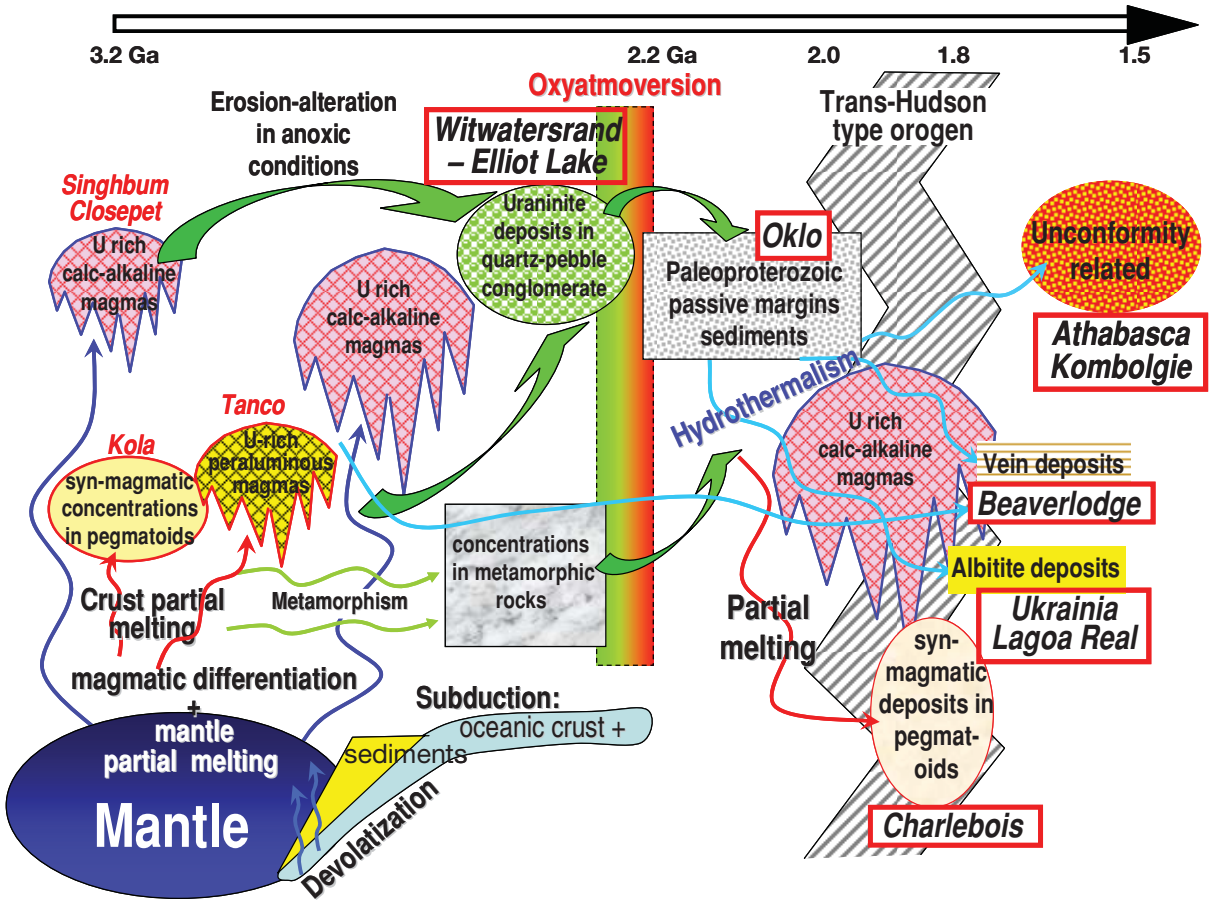


FIG. 2. Evolution of uranium fractionation mechanisms and the genesis of the different uranium deposit types from Mesoarchean to Mesoproterozoic. The fractionation mechanism is in bold, the major occurrences of the different types of ore deposits formed during this period of time are in italics and framed in red.

However, because of the continuous extraction of uranium-enriched material from the mantle by partial melting, mass-balance calculations indicate that 30 to 60 percent of the uranium initially present in the chondritic Earth has been now transferred to the continents (Ringwood, 1975).

At that time, the most felsic rocks belonged to the classic tonalite-trondhjemite- granodiorite (TTG) suite. The earliest known plutonic rocks, the Acasta gneiss, belong to this group and are ca. 4.0 Ga old (Iizuka et al., 2007). The uranium content of these early felsic rocks was generally less than a few parts per million. At such low levels of enrichment, uranium essentially enters into accessory minerals (e.g., zircon, titanite, monazite, allanite, apatite), and uraninite saturation of the melts was never attained. Uraninite has never been described in pre-Mesoarchean plutonic rocks.

The common accessory minerals represent refractory uranium sources from which uranium cannot be significantly extracted by common hydrothermal fluids. Even if a small fraction could have been extracted, the existence of a reduced atmosphere during this period of time would not have permitted the oxidation of uranium from the tetravalent state, its form within the accessory minerals, to the hexavalent state in order to form uranyl complexes, the most soluble form of U in geologic fluids (Langmuir, 1978). Hence, during this first period, uranium was never enriched sufficiently to represent a viable source for later magmatic or hydrothermal remobilization to form economic deposits. Thorium, which has the same ionic radius as uranium and which exists only in the tetravalent state in natural systems, behaves similarly to uranium in all fractionation processes, and the initial Th/U ratio of the primitive Earth of about 4 would have remained constant if other processes didn't operate.

U Fractionation during Mesoarchean and Early Paleoproterozoic (3.2–2.2 Ga)

During this period uranium continued to be essentially fractionated by magmatic processes but the conditions of fractionation became more complex and the first uranium-rich granites able to crystallize uraninite and the first uranium deposits associated with sedimentary rocks appeared on the Earth. The major change in the fractionation processes was probably related to the significant development of plate tectonics and subduction in particular, although the precise timing at which subduction started during the Earth history still remains highly debated and will be discussed later on.

The first granites sufficiently enriched in uranium to crystallize uraninite are 3.1 Ga in the Kaapvaal-Kalahari craton, South Africa (Robb and Meyer, 1990). They are highly potassic,

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possibly of calc-alkaline affinity, and simultaneously enriched in uranium, thorium, potassium, and other incompatible elements (Fig. 2). Such granites are also known in most other Archean cratons but are younger (from about 2.8 Ga and younger), as in the Yilgarn craton (Champion and Smithies, 2007) and in the basement of the East Alligator River Uranium district of Australia, in the Superior province of Canada, the Amazon craton of Brazil, the Dharwar and Singhbum cratons of India, and the Baltic shield. Despite their high uranium contents and the low Th/U ratios of some of them, uraninite has rarely been described. The nature and origin of these potassic granites and the other granite types which may also have crystallized uraninite are discussed below.

The first economic uranium deposits on Earth, the quartz pebble conglomerate type containing uraninite and variable amounts of gold (Figs. 1, 2), are those hosted by the Dominion Reef Group and the Witwatersrand Supergroup which were initially deposited at 3,083 ± 3 and 2,970 ± 3 Ma, respectively (Robb and Meyer, 1995) on the Kaapvaal craton, in Southern Africa. This type of deposit also represents the only significant uranium enrichment known before 2.2 Ga. Otherwise the highest uranium values at that time were a few tens of parts per million U in the most fractionated parts of some granite complexes. Therefore it is essential to first discuss the origin of uranium enrichment in the quartz pebble conglomerate type, before addressing the evolution of magmatic uranium fractionation during this period, to evaluate the possible genetic relationships between the two processes. In this respect, the level of oxygen in seawater and the atmosphere at that period of time will be first discussed, because it is crucial to know if the solubility of uranium in aqueous fluids was controlled by oxidized or reduced conditions. In reduced conditions uranous species (U^{4+}) have an extremely low solubility, equivalent to that of Th^{4+} (Fanghänel and Neck, 2002; Rai et al., 2003), whereas, in oxidized conditions, $U^{4+}$is oxidized to $U^{6+}$to form the uranyl ion (UO^{22+}) which is highly soluble in geologic fluids (Langmuir, 1978).

Oxygen level in the Archean and Late Paleoproterozoic atmosphere

The evolution of oxygen content in the atmosphere through time is a matter of intense debate. Ohmoto (1996) summarized the evidence for atmospheric oxygen levels of at least 50 percent of the present atmospheric level during the Archean, and Barnicot et al. (1997) among others, has defended a hydrothermal origin of the gold and uranium concentrated in the quartz pebble conglomerates of the Witwatersrand. We will not enter in the discussion of all the arguments provided by these authors, just those directly related to the uranium mineralogy and geochemistry which are within the scope of this paper.

The most compelling evidence directly provided by U mineralogy and geochemistry supporting the low level of oxygen before about 2.2 Ga and consequently the detrital origin of uraninite in the Archean and Late Paleoproterozoic quartz pebble conglomerates is as follows:

1. As noted by Ramdhor (1958), the quartz pebble conglomerates contain abundant detrital Th- and REE-rich uraninite, siderite, and pyrite, which are only known in significant abundance in strata older than 2.4 Ga, the most economically important being the Mesoarchean Dominion reef and Neoarchean Witwatersrand deposits in South Africa and the Early Paleoproterozoic Huronian deposits in Canada. A detrital accumulation of such minerals is only possible at low oxygen levels.

2. The detrital origin of such redox sensitive minerals in pre-2.3 Ga quartz pebble conglomerate beds has been further supported by the recent discovery of uraninite in the detrital formations of the Pilbara craton, Australia, where pyrite, uraninite, and gersdorffite, of unequivocal detrital origin, have been discovered in 3,250 to 2,750 Ma fluvial siliciclastic sediments devoid of any significant evidence of hydrothermal alteration (Rasmussen and Buick, 1999), unlike the Witwatersrand and Elliott Lake occurrences.

3. The uraninite grains in quartz pebble conglomerates in these deposits contain high Th (1–12 wt % ThO_2) typical of high-temperature crystallization, generally from a magma, because Th is only weakly soluble during low- to intermediate-temperature processes, such as in hydrothermal fluids. Moreover, the different uraninite grains in the conglomerate beds have different Th contents reflecting different conditions of crystallization of the uraninite in the different types of source rocks. Uraninite crystallizing from high K calc-alkaline melts has higher Th contents (~5–12 wt % ThO_2) than uraninite crystallizing from peraluminous melts (~1–12 wt % ThO_2, Cuney and Friedrich, 1987).

4. REE patterns of uraninite determined by SIMS on individual crystals from the Dominion reef, South Africa, and from Elliott Lake (Duhamel et al., 2009) present the same characteristics as those of the uraninite crystals having crystallized at high temperature from granite or anatectic pegmatite such as Rössing, Namibia (Fig. 3): (1) a high total REE abundance (×104 chondritic abundance for most of the REE) which results from their increased substitution into the uranium oxide crystals at high temperature compared to those crystallized at lower temperature (Bonhoure, 2007), (2) a moderate global fractionation of the REE pattern, reflecting a weaker crystal chemical control for REE incorporation relative to their ionic radius into the uraninite structure at high temperature, compared to those crystallized at lower temperature, as shown by comparison with the REE patterns of uranium oxides from unconformity related deposits (Fig. 3), and (3) a strong and variable negative europium anomaly reflecting the fractionation of plagioclase in the magma from which they have crystallized, similar to the Eu anomalies observed in the uraninite from Rössing and Finland pegmatoids. The term pegmatoid is used for leucocratic granitic veins or small granitic bodies, generally having a pegmatitic texture, but devoid of internal zoning and derived from the partial melting of metamorphic rocks (Cuney and Kyser, 2008).

5. The very low solubility of uranium in water during this period of time is also shown by the low concentration of redox-sensitive elements, such as U and Mo, in highly carbonaceous shale. Such shales older than 2.2 Ga are not enriched, because the oxygen content of the atmosphere and surface water was too low to increase their solubility and thus there was little to preferentially trap in reduced environments (Yang and Holland, 2002).

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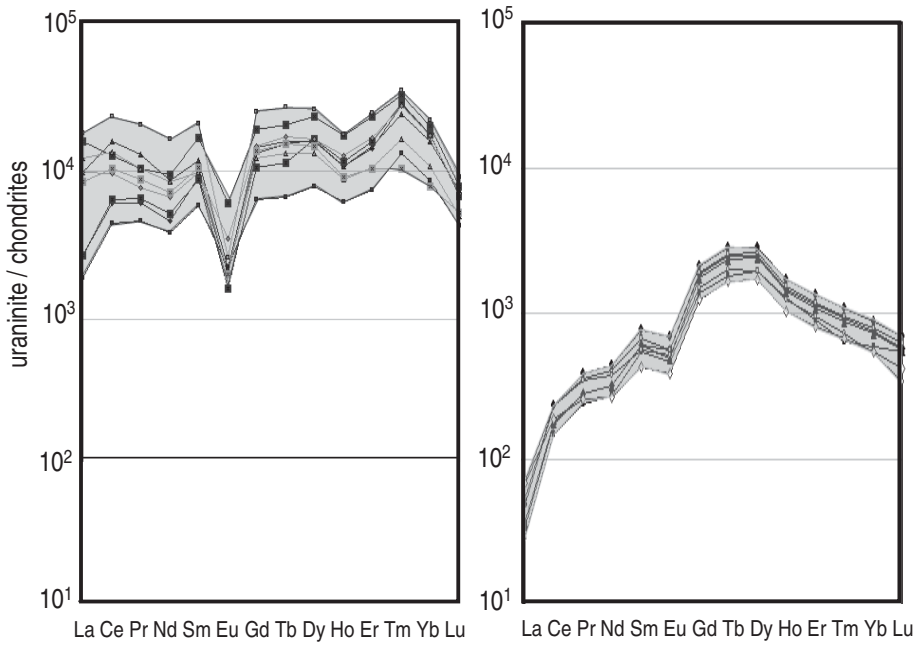


FIG. 3. REE patterns normalized to chondrites of uraninite crystals from the quartz pebble conglomerates of the Dominion reef (South Africa; I. Duhamel, pers. Commun.; Σ_{REE} = 28,771 ppm, $\Sigma_{HREE}/\Sigma_{LREE}$ = 0.77) compared to the patterns obtained on the diagenetic hydrothermal uraninite from the MacArthur River unconformity-related deposit, Canada (Σ_{REE} = 1,766 ppm, $\Sigma_{HREE}/\Sigma_{LREE}$ = 2.18). The uraninite of the Dominion reef has a typical magmatic signature (see text for explanation).

6. If redox contrasts at the level of those appearing after 2.2 Ga would have existed before this time, deposits with U enrichments higher than some thousands of parts per million, the highest U grades observed in the quartz pebble conglomerates, should have been found in the carbonaceous shale deposits. The first known major redox-controlled deposition of uranium was in the Oklo area where U grades exceed 10 percent in several parts of the deposits which were able to become supercritical as natural nuclear reactors.

7. Because of the absence of oxygen, Early Archean (3.46 Ga) hydrothermally altered sea-floor tholeiitic and komatiitic basalts have maintained the primary magmatic Th/U ratio of 4, in spite of their intense alteration (Kentaro and Yasuhiro, 2007). Hydrothermally altered modern basalt exhibits much lower Th/U (down to 0.03) resulting from the selective trapping of dissolved U from oxidized deep seawater during its infiltration into the oceanic crust on each side of the oceanic ridges.

Additional recent and strong evidence for a reduced atmosphere before 2.2 to 2.4 Ga, not directly related to uranium geochemistry, is provided by sulfur isotope mass independent fractionation. Mass independent fractionation requires the lack of an ozone shield in the atmosphere, allowing deep penetration of high energy ultraviolet light and photochemical dissociation of SO_2 into elemental and water-soluble S species (Farquhar et al., 2000). In an atmosphere with oxygen content larger than 10^{-5} times the present atmospheric level, sulfur species are oxidized to sulfate, exchange, and thus lose most of their mass independent fractionation signal. Bekker et al. (2004) have shown that by 2.32 Ga, atmospheric p_{O_2} was already $>10^{-5}$ present atmospheric level, from the sulfur isotope composition of synsedimentary or early diagenetic pyrites in black shales from South Africa.

All these results further support other long-standing evidence of a low oxygen level in the Archean and very Early Proterozoic (e.g., lack of red terrestrial strata), and then a rapid rise of the oxygen content in the atmosphere between 2.45 and 2.22 Ga during the so called Great Oxidation Event (Fig. 2). All of this strengthens the detrital origin of initial uraninite accumulations in the Early Paleoproterozoic quartz pebble conglomerates and the low oxygen level in the atmosphere at that time.

Archean to Late Paleoproterozoic uraninite-bearing granites

Possible sources able to deliver detrital uraninite into fluvial systems should be easy to identify if the above argument is correct. From Archean to the present, three main types of granites can be sufficiently enriched in uranium to crystallize uraninite: (1) highly fractionated high K metaluminous granite and related pegmatites, (2) highly fractionated peraluminous leucogranite and related pegmatites, and (3) weakly peraluminous pegmatoids resulting from low degree of partial melting of metasedimentary rocks (Cuney and Friedrich 1987; Cuney, 2009).

The oldest granitic bodies known to have crystallized uraninite are situated in the Kaapvaal craton, in the vicinity of the Dominion-Witwatersrand basin (Robb and Meyer, 1988, 1990). Let us now consider the nature of these granites and the geotectonic conditions permitted by their genesis at about 3.1 Ga. According to de Wit et al. (1992), after a long history of convergent-margin tectonics, several crustal blocks collided to form the Kaapvaal craton which functioned as a stable continent by ~3.1 Ga, at least in its central part. The first

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highly potassic granites (representing a part of the Granodiorite-Granite-Monzogranite suites [GGM] of de Wit, 1998) are intruded inside and along the boundaries of these crustal blocks between ~3.2 and 3.0 Ga and are attributed to a collision event which ended by 3.0 Ga. At least three subsequent generations of potassic granite were emplaced between ~2.75 and 2.5 Ga (Poujol et al., 2003). The Witwatersrand fluvial deposits were derived from the pre-3.1 Ga granitoid crust and composite terranes accreted after 3.1 Ga (Robb and Meyer, 1995).

Several hypotheses are proposed for the genesis of the 3.1 Ga, highly potassic, calc-alkaline granites. They are generally considered as derived by partial melting of a TTG crust and associated metasedimentary rocks at medium to deep crustal levels (Sylvester, 1994; Frost et al., 1998) or by melting of a mantle wedge peridotite previously metasomatized either through interaction with slab melts (Martin et al., 2005) or by fluids enriched in incompatible elements produced by the progressive dehydration of injected crustal material. The first and most widely proposed hypothesis does not explain the high uranium contents of many of these granites, because TTG are poor uranium sources at best, and at deep crustal levels under granulite facies conditions, the rocks are generally depleted in uranium. This hypothesis also fails to explain the enrichment of some of the most highly incompatible elements, especially K, U, and Th, in the locally associated mafic phases of high K calc-alkaline suites, such as monzonite and monzodiorite. The genesis of such rocks requires derivation from a subcontinental lithospheric mantle enriched in incompatible elements.

Condie and Kröner (2008) proposed that modern-style plate tectonics were already operational, at least in some places, by 3.0 Ga or slightly earlier and became widespread by 2.7 Ga. The development of subduction meant the injection into the mantle, at convergent plate boundaries of basaltic oceanic lithosphere and sediments deposited in accretionary prisms, enriched in U, Th, K, as well as other incompatible elements relative to the primitive mantle.

At that time, the mantle was predominantly primitive, with about 21 ppb U. The basalt derived from its partial melting should have been richer in uranium than the 70 to 100 ppb U of present average MORB (from MORB glasses, White, 1993) from the partial melting of the depleted mantle with only about 5 ppb uranium. Sediments were being generated by weathering and erosion of the upper part of the continental crust and thus represented the most U enriched material on Earth at that time. Enrichments in the sediments remained limited because uranium was not preferentially trapped in reduced organic-rich environments. As noted above, black shale older than 2.2 Ga is not enriched in uranium (Yang and Holland, 2002).

Incompatible element enrichment of the mantle wedge during subduction may occur through two different mechanisms: either by partial melting of the injected crustal material (slab melts), or by fluids enriched in incompatible element produced by the progressive dehydration of the injected crustal material. Today, the oceanic crust extensively produces incompatible element-enriched fluids before the metabasalt solidus is intersected because the angles of the subduction zones are generally too steep to induce hydrous slab melting (Martin, 1986). These fluids metasomatize the overlying mantle wedge and/or lithosphere; partial melting generates andesitic and basaltic magmas; and these with further fractionation give rise to the andesitic arc magmatism and associated juvenile incompatible element-enriched granitoid intrusions (Baker et al., 1994). However, for Late Archean granites, such as the uraninite-bearing Closepet granite, Moyen et al. (2001) proposed derivation by melting of a mantle wedge peridotite that had been previously metasomatized by slab melts in the same manner that the sanukitoids were generated. Sanukitoids have been first distinguished from TTG by Shirey and Hanson (1984), because of their major element geochemistry similar to that of Miocene high Mg andesite (Sanukite) from the Setouchi volcanic belt of Japan and they referred to them as "Archaean sanukitoids." The sanukitoids are meta-aluminous and moderately potassic, rich in Mg, Ni, Cr, in most LILE and in LREE, and their REE patterns are strongly fractionated. However, the Closepet-type granitoids are richer in incompatible elements (e.g., K, U, Th, Rb, Y, Zr, and REE) than the sanukitoids and are remarkably similar in composition to younger high K calc-alkaline granites. Hence, the occurrence of highly potassic calc-alkaline granites (possibly as part of a GGM suite) highly enriched in U, Th, and K as early as 3.2 to 3.1 Ga rather suggests that relatively steeply dipping subduction processes were already generating metasomatic fluids, because this process is the most efficient for quantitative selective transfer of incompatible elements into the overlying mantle wedges. Elliott et al. (1999) have estimated a flux of 500 t U/a from slab to arc, along present-day 37,000 km of active arcs. This includes contributions from both subducted sediment and altered oceanic crust, but many studies have shown that the sedimentary flux is dominant (e.g., Hawkesworth et al, 1993; Elliott et al., 1999). Hence, during the Late Archean and Early Paleoproterozoic, subduction of sediment may have been already sufficient to produce significant U enrichments in the subcontinental lithosphere. However the genesis of highly potassic highly fractionated granites enriched in U and Th requires further research.

In the Kaapvaal craton, relatively undeformed thick and extensive horizontal sheets of highly potassic GGM suites invaded 3.6 to 3.2 Ga TTGs at 3.1 to 3.0 Ga (de Wit et al., 1992). On the Early Archean Pilbara craton and the Late Archean Yilgarn, Rum Jungle, Wyoming, and Zimbabwean cratons, the highly potassic granitoids are far more voluminous than the TTG in present exposures (Bowring and Housh, 1995; Kinny and Nutman, 1996; Frost et al., 1998; Drüppel et al., 2009). 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 at least four periods, at ~2.8, 2.67, 2.63, and 2.55 Ga (Frost et al., 1998). These represent the uranium sources for the much younger roll front deposits (Stuckless and Nkomo, 1978). In the Rum Jungle province TTGs are nearly absent.

Despite high uranium contents and low Th/U ratios in these potassic granites, uraninite has been relatively rarely identified in petrographic descriptions and still more rarely analyzed. The main occurrences are reported in Table 2. For most of them no analysis of the uraninite crystals is available.

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TABLE 2. Pre-2.2 Ga Granitoids and Pegmatites in which Uraninite has been Identified

Rock type	Location	Typology	Age (Ba)	Th (ppm)	U (ppm)	Th/U	U minerals	Reference[1]
Johannesburg Granite	South Africa	High K Calc-alkaline	3.060 ± 30	1.09 – 17.5	0.19 – 12.6	–	Uraninite	1
Hinterland Witwatersrand Granite	South Africa	Peraluminous ?	>3.08	1.9 – 26.7	8.2 – 116.1	0.15 – 3.7	Uraninite	2
Kaduna orthogneiss	Nigeria	?	3.050 ± 23	–	–	–	Uraninite in zircon	3
Singhbum Granite	India	Peraluminous	3.0–2.9	–	–	–	Uraninite, allanite, titanite	4
Tanco Pegmatite	Manitoba, Canada	Peraluminous	2.640 ± 7	1	55	0.02	Uraninite, U microlite	5
Karimnagar charnockitic gneiss	India	High K calc-alkaline ?	2.6–2.8	-	-	-	Uraninite	6
Salobo IOCG	Brazil	High K calc-alkaline	2.573 ± 2	-	-	-	Uraninite	7
Clospet Granite	India	High K calc-alkaline	2.515 ± 18	-	-	-	Uraninite	8

Note: - = no data available

[1] References: 1 = Hallbauer (1984), 2 = Robb et al. (1990), 3 = Bruguier et al. (1994), 4 = Sarangi and Singh (2006), 5 = Duhamel et al. (2009), 6 = Jayananda et al. (2000), 7 = Salobo iron oxide-copper-gold deposit related to the old Salobo high K calcakaline granite, Requia et al. (2003), 8 = Santosh et al. (2004)

Uraninite of Archean age is also known in the iron oxide-copper-gold (IOCG) (±U) deposits of the Amazonian craton in Carajas, Brazil (Tallarico et al., 2005). They are certainly rare, but with U contents up to several hundreds of parts per million essentially hosted by uraninite, these deposits are much richer in uranium than surrounding highly potassic granite and pegmatite bodies. For example, the Igarapé Bahia-Alemão IOCG (U-REE) deposit is possibly related to fluids exsolved from high K calc-alkaline granites (A-type) of the Carajás belt, such as the Old Salobo (2573 ± 2 Ma; Requia et al., 2003) and Itacaiúnas (2560 ± 37 Ma, Souza et al., 1996) granites.

Thus, the highly potassic granites had generated even more highly fractionated granites and pegmatites able to crystallize uraninite. However, the most fractionated end members of such plutonic suites are generally emplaced in their apices and have been largely eroded. Moreover, the Th/U ratios of many of these granites are greater than 10—much higher than the average crustal ratio. The higher Th/U ratio may reflect the source but also results from two processes: faster decay of U relative to Th has increased the present Th/U ratio of Archean granites by about 1 (Robb et al., 1990), and oxidized meteoric waters have dissolved uraninite crystals from near-surface samples, which may also explain the paucity of uraninite reports for these granites (Table 2).

In conclusion, for the genesis of the uraninite-bearing highly potassic granites, possibly of calc-alkaline nature, the preferred model involves dehydration of subducted crustal material to enrich the subcontinental lithospheric mantle in incompatible elements, which are then extracted by partial melting and fractional crystallization (Fig. 2). Partial melts were derived from mantle enriched by subduction-related processes in addition to those derived from depleted and primitive mantle. During their ascents through the continental crust, any of these melts may also have assimilated variable amounts of U-Th-K–rich material. Thus, further research is required to constrain the mechanisms that enriched uranium and other incompatible elements in the first highly potassic granites able to crystallize uraninite from 3.2 to 2.4 Ga.

A second type of granite that crystallized uraninite began to appear by about 2.7 Ga. It corresponds to highly peraluminous leucogranite and pegmatite generated by limited partial melting of metasedimentary rocks in the continental crust. Some of these bodies are enriched in uranium but the majority are depleted in most other LILE elements (Th, REE, Zr) because of the low solubility of the Th-REE-Zr–bearing accessory minerals in low-temperature highly peraluminous melts (Cuney and Friedrich, 1987). The degree of magmatic fractionation reached in these Archean granites has not been exceeded since then. The best example is the peraluminous rare element Tanco pegmatite in Manitoba, Canada, which crystallized low Th-Y-REE uraninite at the magmatic stage (Duhamel et al., 2009). Significant fractionation of highly peraluminous, low-temperature melts is probably the only way to produce negative differential fractionation of thorium relative to uranium without requiring redox processes.

The third type of granite which can be highly enriched in uranium is peralkaline—rare in the Archean and Late Paleoproterozoic. The oldest well-documented examples are the 2.7 Ga highly potassic trachyte and leucite phonolite from the Kirkland Lake region of Canada (Blichert-Topf et al., 1996). Other examples of Archean peralkaline rocks are relatively common, but volumetrically insignificant, lamprophyric dikes and syenitic intrusions. Their rarity 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) or might be a result of poor preservation, because they are generally emplaced at a very high structural level (Blichert-Toft et al., 1996). In any case, despite local strong enrichment in uranium they rarely crystallize uraninite; instead uranium is distributed in the structure of abundant complex Zr-, REE-, Nb-, Ti-bearing minerals (Cuney and Friedrich, 1987).

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Also between 3.2 and 2.6 Ga, early photoautotrophic organisms in the hydrosphere were already producing oxygen, especially the rapidly expanding stromatolites. But atmospheric oxygen levels did not rise significantly because photosynthetic oxygen production was countered by sinks such as reduced volcanic gasses and the oxidation of Fe^{+2} to Fe^{+3}. From 2.6 to 2.2 Ga, extensive BIF and manganese oxide deposits captured the oxygen released by the early organisms. The oxygen level was able to rise sufficiently in some restricted areas to produce the first basins with red-bed sequences during the Late Archean (Eigenbrode and Freeman, 2006). The oldest evidence of extensive oxidative weathering is associated with 2.32 to 2.22 Ga glacial deposits and breakup of an inferred Late Archean supercontinent (Barley et al., 2005).

U Fractionation during the Early Paleoproterozoic to Paleozoic (2.2–0.4 Ga)

U enrichment of Paleoproterozoic epicontinental platform sediments

After the dramatic increase in oxygen fugacity of the atmosphere during the earliest Paleoproterozoic, by about 2.3 to 2.2 Ga (Farquhar and Wing, 2003; Bekker et al., 2004), uranium oxides began to be in direct contact with oxygenated water and passed into solution as uranyl complexes, dominantly as uranyl carbonates. Uranium was then dissolved and derived from uraninite accumulated in the pre-2.2 Ga paleoplacers or having crystallized in highly fractionated uranium-rich granite, from metamict uranium-rich silicate minerals and other oxide minerals in plutonic or sedimentary rocks, and from altered volcanic acidic glass.

At the same time, during the so-called Shunga event (Melezhik et al., 1999; Hannah et al., 2008), huge amounts of organic matter were incorporated by sediments in shelf and marginal sea environments. This event corresponds to an unprecedented increase in taxonomic diversity and expansion of stromatolites and to the occurrence of $\delta^{13}C$ excursions in carbonates between 2.25 and 2.06 Ga (Semikatov et al., 1999). At about 2.05 Ga, black shale incorporated up to 15 wt percent organic carbon in the nonmetamorphosed FB Formation of the Franceville basin, Gabon (Gauthier-Lafaye and Weber, 1989) and an average value of 25 wt percent C over a thickness of 600 m in the slightly metamorphosed upper Zaonezhskaya Formation at the northern shore of Onega Lake, Russia (Melezhik et al., 1999). This is in strong contrast to the median of 3.35 wt percent organic carbon in average black shales compiled by Vine and Tourtelot (1977). In equivalent Paleoproterozoic metasedimentary rocks metamorphosed to high grade, such as the Wollaston belt in Saskatchewan, Canada, and in the Cahill Formation in the Northern Territory, Australia, such organic-rich strata were metamorphosed to graphitic schist that during deformation became abundant graphite-rich fault zones. The abundance of graphite schist is particularly well documented by geophysical conductivity maps in the Wollaston belt, below the Athabasca Basin in northern Saskatchewan (Matthews et al., 1997).

The uranium content of these carbonaceous shales is anomalous, averaging 3.5 ppm (48 samples, Gauthier-Lafaye, 1986) to 10.8 ppm (6 samples, Mossman et al., 1998) for the Francevillian, and up to 31 ppm (12 samples with U contents from 12–84 ppm) for the Onega basin (Melezhik et al., 1999). The average U content of younger black shale is also 30 ppm (Vine and Tourtelot, 1977). Such high uranium contents indicate that by 2 Ga, oxidizing conditions were already sufficient to dissolve uranium as UO^{2+} and to deposit it in reduced environments. At a more global scale, Th and U analyses of shale from a wide range of ages show a significant decrease in their mean Th/U ratio with decreasing age, from 4 at the Archean-Proterozoic boundary to 0.55 in the late Phanerozoic (McLennan and Taylor, 1980).

It has been proposed that the widespread deposition of black shale at about 2 Ga was related to increased weathering fluxes of nutrients such as phosphorous into the oceans triggered by global changes after major glaciations (Condie et al., 2001). Increased availability of phosphorous would have stimulated photosynthetic oxygen production (Papineau et al., 2007). This hypothesis is consistent with the first large phosphogenic event at about 2 Ga in similar environments (Choudhuri and Roy, 1986) that are also enriched in uranium, such as the 2.1 to 1.92 Ga Ludicovian epicontinental carbonaceous strata of Karelia (Mikhailov et al., 1999).

U enrichment of oceanic crust and decrease of upper mantle Th/U

Another important process linked to the oxygenation of the atmosphere and hydrosphere, which further contributed to uranium enrichment of the subcontinental lithosphere, is the trapping of uranium from oceanic water in the upper part of the basaltic crust during convective hydrothermal circulation through midocean ridge basalt. The present level of uranium in seawater (3.22 ppb) was probably reached very soon after 2.2 Ga. The nearly complete trapping of seawater uranium within sea-floor basalts is evidenced by the composition of the reduced hydrothermal fluids escaping from the midoceanic ridge which contain two orders of magnitude less U (0.06–0.18 ppb U) than the seawater (Chen et al. 1986), and U enrichment directly measured in altered basalt sampled by sea-floor drilling: average U = 390 ppb and local enrichment up to 1.8 ppm U. The strongest enrichment, up to 50 ppm U, is in hydrothermal carbonate minerals (Kelley et al., 2005). The global amount of U trapped by hydrothermal circulation in the upper part of the oceanic crust is estimated at 1,500 to 4,000 t U/yr (Elliott et al. 1999), about 98 percent of the U from seawater (Chen et al., 1986). As Th is unaffected by this process, the Th/U ratio of altered continental crust has been strongly decreasing. Meanwhile, continental weathering is preferentially replenishing the ocean with hexavalent U through fluvial transport while Th remains in the detrital sedimentary residue.

Unless all the U in altered oceanic crust and sediment is directly returned to the continents by partial melting at subduction zones, then the net effect of the plate tectonic cycle would be to preferentially recycle continent-derived uranium back to the mantle relative to thorium, and this would lead to a continuous decrease of mantle Th/U, generally determined from the $^{232}Th/^{238}U$ ratio (kappa). Equilibrium appears to have been attained in present subduction systems wherein sediment with generally high Th/U (2.0–8.9) is combined with low Th/U-altered oceanic crust (~0.23), and thus bulk subducting slabs have Th/U ratios of about 2.6, similar to

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modern depleted mantle (~2.55; Elliott et al., 1999). Taking an intermediate estimate of the recycled excess U flux (1000 t U/a), assuming no new continental crust was formed during the last 2 Ga, Elliott et al. (1999) estimated that the Th/U ratio of an upper mantle MORB source (with 0.01 ppm Th) could have been lowered from 4.0 to 2.6 over only about 1.8 Ga. This process, labeled Post-Archean Uranium Recycling, has been proposed to explain the "kappa conundrum," that is, the discrepancy between the time-integrated mantle Th/U ratio of about 3.8, derived from radiogenic Pb isotopes, and the present-day upper mantle Th/U of about 2.55.

An independent confirmation of the decrease of the Th/U ratio in the upper mantle is also provided by Th/U ratios of kimberlitic zircons from 10 kimberlite occurrences, selected from four continents, and with ages spanning Archean to recent times (Zartman and Richardson, 2005). Assuming that Th and U entered melt in the same ratio as that of the asthenospheric mantle, and that zircon incorporated these elements with Th/U ratios one order of magnitude lower than for the melt, a clear trend appears with a decrease of the Th/U of the mantle from about 4.2 at 2.5 Ga to about 2 today (Fig. 4). The value of 4.2 is close to that of chondrite (Th/U = 3.92). The value of 2 is close to that of average modern MORB (Th/U = 2.5) and, therefore, should reflect the ratio of the asthenospheric mantle, if U is not fractionated relative to Th.

Implications for the formation of new U deposit types

The change of the Th/U ratio in the upper mantle: It is difficult to relate this change directly to an evolution in uranium mineralization processes. Theoretically a decrease of the Th/U ratio should favor the crystallization of a larger proportion of uranium outside the structure of accessory minerals in the most fractionated magmas, in a phase like uraninite, more easily leachable by common hydrothermal fluids. But, in mineralized magmatic associations which are the most likely to have developed from partial melting of the mantle, such as the Ilimaussaq and the Bokan Mountain peralkaline complexes, a Th/U ratio of about 4 is preserved even in the most fractionated body, except where fluid/rock interaction is documented (Cuney and Kyser, 2008). Similarly most high K calc-alkaline complexes, which may be at least partly derived from the subcontinental lithosphere and which are proposed as representing the source of uranium deposits, have average Th/U ratios close to 4. Conversely, the huge quantity of uranium accumulated in the post-2.2 Ga epicontinental platform sequences (Fig. 2) is illustrated by the large proportion of world uranium resources (Fig. 1) and subeconomic occurrences with low Th/U ratios that are hosted by or related to these sequences (as discussed further on).

Franceville basin deposits in Gabon: They represent the first type of uranium deposits formed after oxyatmoversion by redox processes. These are Boyindzi, Oklo, Okelobondo, Bangombe, and Mikouloungou (Gauthier-Lafaye, 1986). The sedimentary succession hosting these deposits was deposited at about 2.15 to 2.1 Ga and the deposits were formed at 2.0 Ga (Fig. 2). The P-T-X conditions prevailing during their genesis and the host sedimentary environments are similar to those of unconformity-related deposits (Mathieu et al., 2000), but deposition took place at the redox interface between oxidized basal sandstone of the FA Formation and overlying oil shale of the FB Formation. This contrasts with the redox interface located near the unconformity between oxidized sandstone and reduced basement rocks for the younger unconformity-related deposit type.

The Franceville basin in Gabon and the Zaonezhskaya Formation, Onega Lake, Russia, represent non- or weakly metamorphosed epicontinental platform successions that host

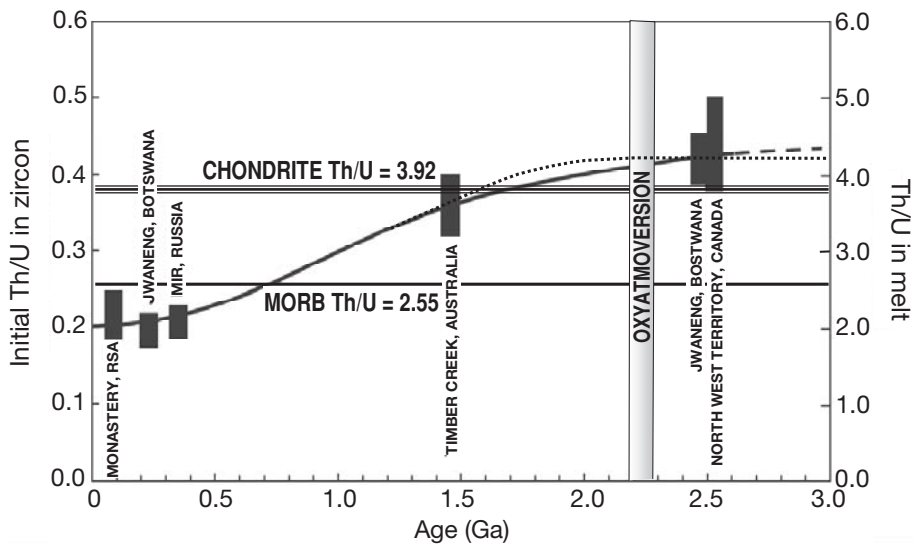


FIG. 4. Evolution of the Th/U of the mantle from Archean to present times estimated from the Th/U ratios of 10 kimberlitic zircons (modified from Zartman and Richardson 2005). The period during which the oxygen content of the atmosphere rose to nearly present concentration level is indicated. The dashed curve is preferred to the solid one proposed by Zartman and Richardson (2005) to maintain a constant Th/U ratio of the mantle before the oxyatmoversion, followed by a period of steadily decreasing Th/U from the middle of the Paleoproterozoic when the effect of preferential recycling of uranium in the mantle becomes noticeable.

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significant 2 Ga uranium deposits. A large number of other uranium occurrences are known in similar Paleoproterozoic formations, most of them having been metamorphosed to high grade during a worldwide orogenic event between 2.1 and 1.8 Ga (Fig. 2) that built Nuna (also named Columbia), the first relatively well characterized supercontinent (Zhao et al., 2002). The main primary Paleoproterozoic sediment-hosted uranium mineralization occurrences, or their remobilization as pegmatoids, include those summarized in Table 3.

Uranium deposits related to Na metasomatism: Most uranium deposits associated with this process, and especially the largest ones (the Central Ukrainian district and the Lagoa Real district, Brazil) were generated between 1.8 and 1.4 Ga in metasedimentary, metavolcanic, and granitic rocks in Paleoproterozoic epicontinental settings (Fig. 2). The roles of the host formations were probably critical as sources of uranium, but the geodynamic setting favoring the development of this type of deposit during that time is still poorly understood. A geotectonic model has to be able to explain the origin of the thermal events able to circulate large volumes of fluids at high temperatures (550°–350°C) to produce Na metasomatism along crustal-scale structures (several tens to hundreds of kilometers). Most puzzling, this event occurred some hundred million years after the last orogenic event (Cuney and Kyser, 2008). Another less important episode of uranium mineralization associated with Na metasomatism took place during the so-called Pan-African or Brazilian event (~500 Ma). Nearly all the uranium occurrences of this episode are located in the northeastern part of the Sao Francisco craton (Espinharas and Itataia deposits in Brazil) and its extension in Western Africa (Poli in Cameroon; Vels and Fritsche, 1988).

Unconformity-related uranium deposits: After ~1.75 Ga, in many parts of the Nuna supercontinent, or slightly earlier as in Central Ukraine and southern Africa, a long period of relative tectonic quiescence is recorded by numerous highly oxidized (red-bed) intracontinental siliciclastic basins of broad geographic extent, ranging from 1.5 up to 5 to 6 km thick (for a review, see Kyser et al., 2000; Cuney and Kyser, 2008; Fig. 2). Because of the absence of land plants, continental sediments, and soils, weathering of the rocks was extremely different (Erikson et al., 2001). Without the protection of plants, the fine-grained particles from the exposed soils were essentially removed by the winds. As a consequence the Proterozoic fluviatile sediments are poor in clay-rich layers, permitting the formation of vast aquifers. In the Athabasca (Canada) and Kombolgie (Australia) basins, the unmetamorphosed oxidized quartzose siliciclastic strata represent huge reservoirs for sodic brines derived from overlying evaporates (Derome et al., 2005, 2007). These brines became calcic when infiltrated into the basement (Derome et al., 2005) and leached U dominantly from Paleoproterozoic epicontinental sediments, from their anatectic derivatives, and from high K-U granites (Cuney, 2005). They finally formed the major hydrothermal-diagenetic uranium deposits with basement and/or basin redox control, generally called "unconformity related," of northern Saskatchewan in Canada and the Northern Territory in Australia. The uranium deposits are hosted either within the basement to these basins (post-2.2 Ga metamorphosed epicontinental platform sediments) or just above the unconformity in the overlying sandstone. The uranium-rich basement metasedimentary assemblages of Athabasca Basin are in the Wollaston and Taltson belts; those of the Kombolgie subbasin are in the Cahill Formation. The uranium deposits have

TABLE 3. Uranium Mineralization Occurrences Hosted by Paleoproterozoic Metasedimentary Rocks Deposited on Epicontinental Platforms

Occurrences	Type of mineralization	References
Wollaston and Mudjatik belts (Saskatchewan, Canada)		Parslow and Thomas (1982)
Duddridge Lake	Meta-arkose	Parslow and Thomas (1982)
Burbridge Lake and Cup Lake	Calc-silicate	Parslow and Thomas (1982)
Charlebois	Pegmatoids	Parslow and Thomas (1982)
Northern Quebec and Baffin (Canada)		
Steward Lake	Pegmatoids	Freewest Resources Canada
Ugava Bay	Pegmatoids, marbles	Neto et al. (2009)
Baffin Islands	Pegmatoids	Maurice (1977)
Colorado		
Wheeler basin	Pegmatoids	Young and Hauff (1975)
Norway		
Orrefjell	Pegmatoids	Lindahl et al. (1985)
Southern Finland		
Palmottu	Pegmatoids	Räisänen (1989)
Late orogenic potassic granites		Lauri et al. (2007)
Russia		
Kola Peninsula, Litsk district	Pegmatoids	Savitskii et al. (1995)
Ludicovian	P-rich carbonates	Shurilov et al. (2007)
South Australia		
Crocker Well	Pegmatoids	Thompson (1965)
Olary province	Pegmatoids	Thompson (1965)
Cloncurry: Six Kangarros	Pegmatoids	
Northern Territory: Nanambu,	Pegmatoids	Carter et al. (1961)
Nimbuwah in the Rum Jungle Complex		Ayers and Eadington (1975)

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been dated from about 1.65 to 1.3 Ga (Alexandre et al., 2009). Other uranium deposits in the world, smaller in size but also spatially associated with Paleo- to Mesoproterozoic intracontinental basins, may have been generated by similar processes. These include: the Baker Lake-Kiggavik and Sissons deposits located in the basement close to the Thelon basin (Miller and LeCheminant, 1985), the Mountain Lake deposit hosted by the Hornby Bay basin, Nunavut in Canada, the Karku deposit hosted by the Pasha Ladoga basin, Karelia in Russia (Shurilov et al., 2007), and possibly the metamorphosed Kintyre deposit, hosted within the basement in the vicinity of the Coolbro Sandstone, in Australia. Significant differences exist among these and the classic unconformity-type deposits. For example, the Mountain Lake deposit is a younger analogue of the Franceville deposits, formed at the upward interface from sandstone to carbonaceous shale at 1.2 Ga, driven by the Mackenzie Event (Davis et al., 2008). Further work is needed on some of them before inclusion in the hydrothermal-diagenetic unconformity clan with basement and/or basin redox controls.

The Olympic Dam iron oxide-copper-gold (IOCG)(uranium) deposit: It is by far the world's largest uranium resource and is hosted by the Roxby Downs granite dated at 1.59 Ga (Creaser et al., 1996). This granite was also emplaced in Paleoproterozoic formations similar to those listed above, at the margin of the Archean Gawler craton (Groves and Vielreicher, 2001). However, possible genetic relationships between the uranium enrichment of these formations and the genesis of the Olympic Dam deposit are not well understood. Compared to other IOCG deposits, Olympic Dam is entirely hosted within U-rich high K calc-alkaline granites and volcanics and is the richest in U.

Hydrothermal activity is related to the emplacement of the Roxby Downs Granite and the extrusion of contemporaneous Gawler Range volcanics (Reynolds, 2000). Dating indicates that magmatic activity, brecciation, and Cu-Au mineralization were synchronous (Johnson and Cross, 1995). The Olympic Dam ore genesis is related to the unmixing of a hot, highly saline fluid from a granitic magma which has mixed with an oxidized meteoritic fluid (Hitzman et al., 1992). The high Cl/S ratios of the fluids compared to other hydrothermal systems correlate with very low enrichments in Pb and Zn, elements with low solubilities in S-poor fluids.

Very little information is published concerning specifically the genesis of the U mineralization. Uranium minerals are small euhedral uraninite crystals dispersed within the mineralized breccia, which may correspond to an early higher temperature uranium mineralization event, and pitchblende, coffinite, and U-Ti oxides in veins which should correspond to a low-temperature hydrothermal event. For Hitzman and Valenta (2005), the leaching of U from the wall rocks by the hydrothermal fluids would produce the enrichment in the IOCG deposits. U enrichment in the ore is 10 to 40 times larger than in unaltered host rocks. A more detailed study of the uranium mineralization processes is clearly needed for IOCG deposits and especially in the Olympic Dam deposit.

The 1.4 to 1.3 Ga to 0.5 Ga period: This period corresponds to a nearly barren metallogenic interval, during which no economically significant uranium deposits were formed. Among the few occurrences of this time gap, the Kvanefjeld deposit is related to extreme fractional crystallization of the Ilímaussaq peralkaline intrusion (Sørensen et al., 1974), with 83,320 t U at 0.026 percent (International Atomic Energy Agency, UDEPO data base: http://www-nfcis.iaea.org), and the deposits related to partial melting of the Bancroft district with a total production + resources of about 10,000 t U at less than 0.1 percent formed at about 1 Ga, and several others of the same type in the Grenville orogen which as yet have not reached economic size and grade (Lentz, 1998). Ages of as old as 0.7 Ga have been obtained on the Shinkolobwe deposit (Cahen et al., 1971), Democratic Republic of Congo, but these determinations should be reevaluated because all the other deposits of the Katangan system have given ages close to 0.5 Ga.

The Pan African-Brazilian orogenic event (~0.5 ± 0.1 Ga): This event corresponds to the amalgamation of the Gondwana supercontinent (Meert and Van der Voo, 1997), and is related to the formation of a large variety of uranium occurrences. The Damara-Katangan belt is the most important one of this age for uranium mineralization. One of the major uranium sources for the deposits is probably the epicontinental platform strata comprising oxidized siliciclastic units at the base, overlain by organic carbon-rich shale, limestone, and evaporate deposited between about 850 and 700 Ma. The degree of metamorphism of these strata increases from Katanga (Democratic Republic of Congo) to the southwestern part of Namibia. In Katanga and Zambia metamorphic temperatures do not exceed 300° to 350°C, whereas partial melting temperatures were reached in the southwestern part of Namibia. In Katanga and Zambia the uranium deposits were produced by hydrothermal-metamorphic processes (Meneghel, 1981), whereas in Namibia they result from partial melting processes, with the large Rössing deposit and other similar deposits discovered in the vicinity (Roesener and Schreuder, 1992).

The highest grade synsedimentary redox-controlled uranium resources are hosted by black shale deposited during the late Cambrian (510 Ma; Fig. 1). The main example is the Alum shale of southern and central Sweden which contains several million tons of uranium at an average grade of 100 to 300 ppm (Andersson et al., 1985). However, other large accumulations of uranium in black shale, but at lower concentrations, are known from the Paleoproterozoic (Zaonezhskaya Formation, Russia: Mikhailov et al., 1999), to the Upper Devonian (Chattanooga Shale, United States; Conant and Swanson, 1961).

The second period of formation of deposits related to Na metasomatism also took place at about 0.5 Ga but with much smaller resources as discussed above. The Tranomaro uranothorite-bearing pyroxenite from the southern part of Madagascar represents a rather unique type of uranium-thorium mineralization related to metamorphic and/or metasomatic skarn developed in granulite facies conditions at 565 Ma (Andriamarofahatra et al., 1990).

More deposits are likely to be found in large underexplored domains that were deformed and metamorphosed during the Pan-African–Brazilian orogenic event, in areas such as northern Africa, Mozambique, and Brazil. Although no specific type of uranium deposit type can be related to the period 0.5 ± 0.1 Ga, the largest uranium deposits are related to synsedimentary redox-controlled (black shales), hydrothermal-metamorphic (Zambia-Democratic Republic of Congo) and partial melting (Damara Belt) systems as described above.

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U Deposition Controls after the Ordovician (>0.45 Ga)

Consequences of the development of vascular plants

The development of vascular plants on the continents is the last major change in the global environment which deeply affected the metallogeny of uranium. Plant emergence has been thought to take place during the Late Silurian (425 Ma), but recent discoveries in Saudi Arabia (Steemans et al., 2009) fix initial land colonization during the Late Ordovician (~450 Ma). After that time, siliciclastic continental sedimentation no longer led to the formation of homogeneously oxidized successions, best exemplified by the large intracontinental Proterozoic basins associated with unconformity-related deposits. The new sedimentary successions comprise alternating oxidized and reduced layers with inherent strong redox contrasts. Uranium began to be deposited at intraformational redox transitions and was related to the percolation of oxidized fluids. These new fluids were either of diagenetic hydrothermal origin for the tabular and tectonolithologic deposits, or of meteoric origin for the roll-front– and basal-type deposits. Nearly all such deposits were formed from Jurassic to recent times (Fig. 1) on all continents. Distal tectonic events induced the migration of diagenetic fluids within the basins or the infiltration of meteoric waters. Deposits of this style that are hosted by Carboniferous strata, such as the tectonolithologic deposits of the Arlit district in Niger, have Mesozoic to Cenozoic U-Pb ages of pitchblende (Pagel et al., 2005). The global-scale geotectonic process, contemporaneous which such fluid migrations on all continents, is the breakup of Pangea which started in the Early Jurassic to form the incipient Atlantic Ocean.

Most models for the sandstone deposits propose that uranyl-bearing fluids were reduced by the land plant detritus within the host permeable sandstone by humates derived by diagenesis evolution of the plant material (Hansley and Spirakis, 1992) or by biogenic sulfides under low-temperature conditions (e.g., Rackley, 1972; Reynolds et al., 1982). However, low-temperature experiments (Lovley and Philips, 1992; Abdelouas et al., 1998) indicate that such reactions are probably too slow. In addition, experiments demonstrate that iron- and sulfate-reducing bacteria may lead to the reduction of U(VI) to precipitate U(IV) mineral (e.g., Lovley et al., 1991). Despite those results, little direct evidence of biogenically precipitated U minerals, mainly of morphological nature, has been provided in uranium deposits (Milodowski et al., 1990; Min et al., 2005). Recently, robust mineralogical and geochemical evidence has been presented for the involvement of bacterial activity in the genesis of roll-front–type deposits of China (Cai et al., 2007). However, such bacterial activity cannot be invoked for most tabular or tectonolithologic deposits where basinal fluids are involved, with temperatures over 100°C recorded from fluid inclusion studies (e.g., Pagel et al., 2005 for the Niger deposits).

In contrast with a synsedimentary origin of the organic material within the sandstone units, Fisher et al. (1970) have been the first to suggest that the organic material in the South Texas roll-front deposits was hydrocarbons or hydrogen disulfide infiltrated from deep oil reservoirs along faults. Several recent studies support this model for other roll-front deposits: Aubakirov (1998) for Kazakhstan, Cai et al. (2007) for the Ordos basin of China, and Salze et al. (2008) for the Arlit-Akouta tectonolithologic deposits of Niger. Consequently, if continental-derived organic matter is not necessary for the reduction of uranium in the sandstones, intraformational redox-controlled deposits should have existed prior to the Silurian when oil and/or hydrogen disulfide migrated from marine strata and was trapped in porous siliciclastic sediments. This conclusion is of outmost importance for the exploration for uranium deposits in sandstone, because pre-Silurian sandstone sequences become interesting exploration targets for intraformational deposits. In the Oklo district, uranium deposition resulted from such a process, but the migration of hydrocarbons was limited to the vicinity of the contact between the reduced FB black shales and the oxidized FA sandstone. Also, uranium deposits hosted by quartzite or meta-arkose in Proterozoic metamorphosed domains, such as those hosted by the high-grade Mesoproterozoic Ascension Suite in the Mont Laurier area (Cuney, 1982), or the low-grade Paleoproterozoic siliciclastic Amer Lake Formation in Nunavut (Millar, 1996), may have been formed by such a reduction mechanism.

Until the opening of the Eastern Block countries it was also believed that the most important uranium deposits hosted in Phanerozoic rocks were adjacent to Archean-Proterozoic basement which was considered as the source of the uranium (Dahlkamp, 1993). For example, the Jurassic Morrison Formation, which hosts more than 50 percent of the United States reserves, was derived from the erosion of Paleoproterozoic rocks, however with the significant addition of synsedimentary ash-flow tuffs which are either present within the mineralized layers or in stratigraphic horizons above or below them (e.g., Granger and Finch, 1988). In contrast, the large sandstone- and volcanic-related uranium resources of Kazakhstan, Uzbekistan, Mongolia, northern China, and Transbaikalia do not seem to have been derived from significant domains of Archean-Proterozoic basement. Also, the mid-European Variscan belt which hosts the most important resources resulting from hydrothermal remobilization of uranium from granites (Marignac and Cuney, 1999) is essentially associated with Phanerozoic rocks. The genesis of such young uranium provinces, especially in middle Asia, requires further research for understanding the geotectonic conditions of their development.

Explosive volcanism during the Jurassic to Present

The oldest uranium deposits truly related to hydrothermal activity induced by the magmatic activity of a volcanic complex are probably the Devonian Ben Lomond of Queensland, Australia (Bain, 1977) and those in the Caledonian (Late Silurian) Chuili-Kandyktas uplift of northern Kazakhstan (Dahlkamp, 2009). However, these deposits are of relatively small size, with total resources on the order of 10,000 t U (IAEA, UDEPO data base: http://www-nfcis.iaea.org). A few deposits are also related to Carboniferous and Permian volcanism (Cuney and Kyser, 2008). The main hydrothermal-volcanic systems developed during Jurassic (Streltsov, Transbaikalia, Russia and the northeastern Mongolian Province), Cretaceous (Bulgaria, China), Eocene (Mexico), Miocene (Western United States, Peru, Bolivia, Argentina), and the Recent Latium province in Italy (Cuney and Kyser, 2008). The most mineralized districts (Streltsov, northeastern Mongolia and

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Western United States) developed in volcanic caldera, forming large depressions, usually resulting from the collapse of the magmatic chamber roof, during intracratonic basin and range-scale extensional events. The depressions are filled with alternations of basic and acidic volcanics, and subordinate amounts of siliciclastic sedimentary layers, but acidic volcanics generally represent the largest proportion of the extruded magmas. The pyroclastic rocks represent particularly favorable source rocks because they are relatively permeable and a high proportion of the uranium is hosted by the glassy matrix from which it can be easily mobilized during devitrification. Among volcanic rocks, peralkaline magmas represent the most efficient uranium source, as discussed above, and as shown by the association of the largest uranium districts with this type of volcanism. The genesis of large deposits requires also the existence of a relatively shallow magma chamber (a few kilometers to about 5 km), lasting over several million years. Such a situation is able to provide the heat flux and the fracturing necessary to promote focused and long-lasting convective fluid circulation, allowing the development of an important alteration of the rocks within the caldera and underlying basement and an intensive mobilization of the uranium from these rocks. The nature of the uranium-bearing fluids is still poorly known in this type of deposit. The uranium orebodies are mainly trapped along basement structures, which are reactivated after the major part of the caldera filling has been completed and may extend from the volcanic pile several hundreds of meters down into the basement as at the Antei deposit, Streltsovkoye, Russia (Chabiron et al., 2003). They may also occur along the ring structure of the caldera as at McDermitt, United States (Castor and Henry, 2000), at the margin of protruded domes as at Xiangshan, China (Hu et al., 2009), or still along sedimentary layers within the caldera as at Dornot, Mongolia (Petrov et al., 2003).

From the Jurassic to the Miocene, very large amounts of ash-flow tuff, deriving from such caldera systems, were also deposited in intracontinental basins and represent the source for many tabular and roll-front deposits such as in the Central Colorado Plateau of the United States. There is no reason for such volcanic systems and geotectonic settings not having developed more widely prior to Jurassic. Their scarcity may be due to weak preservation because of the continental setting and near-surficial hydrothermal systems, generally within the first hundred meters depth, exceptionally down to more than one kilometre in the case of the Streltsov deposit, and in postorogenic settings.

A series of Paleo- to Mesoproterozoic uranium deposits are associated with volcanic rocks, but the main mineralization process leading to economic-grade mineralization is associated with sodium metasomatism and some have been subsequently metamorphosed, for example, the Michelin and Jacques Lake deposits from Labrador, Canada (Sparkes and Kerr, 2008), the Arjeplog-Arvidsjaur district in northern Sweden (Adamek and Wilson, 1977), and the Vallalah deposit from Queensland, Australia (Polito et al., 2007). Hence they cannot be considered as uranium deposits related to hydrothermal-volcanic systems.

The Cretaceous-Eocene phosphate sea

Uranium concentrations associated with marine phosphatic strata are as old as the Early Paleoproterozoic, but the largest by far are Late Cretaceous to Eocene (90–45 Ma) in age (Fig. 1). These were all deposited at the same paleolatitude (8°–15° N) in carbonate platforms along the southern margin of the Tethys Ocean, from Turkey to Morocco, through Israel, Jordan, Syria, Iraq, Saudi Arabia, Egypt, Tunisia, Algeria, and beyond the Atlantic to Colombia and Venezuela. The phosphate deposition is related to the exceptional coincidence of four conditions. First, considerable rise in sea level during the Late Cretaceous resulted from a global warming episode. Second, the Late Cretaceous continuous east-west Paleotethys Ocean developed between northern (Laurasia) and southern (Gondwana) continental masses as Pangea broke up. This seaway merged with the Central Atlantic gulf already opened during the Late Jurassic, leading to the development of broad carbonate platforms along the southern margins of the Tethys Ocean and a circum-equatorial westward oceanic current in the Tethys. Third, the Tethys was located at the warmest possible equatorial latitudes. Fourth, dominant easterly winds produced a northward Eckman offshore transport of surface waters, inducing upwelling of cold nutrient-rich waters all along the southern Tethys shelf, supporting huge biogenic productivity (Soudry et al., 2004). Morocco, with geologic uranium resources of about 6.9 Mt U at 50 to 150 ppm (International Atomic Energy Agency, 2001), hosts nearly three-quarters of the world uranium resources associated with phosphates.

Deposits related to evapotranspiration

Uranium deposits related mainly to evapotranspiration and associated with calcretes are known only in late Tertiary to Quaternary settings. The uranium is hosted by surficial sediments deposited in fluviatile to lacustrine and/or playa systems which have not been subjected to significant burial. A number of mechanisms, including sorption, colloidal precipitation, changes in vanadium redox state and in CO_2 partial pressure, pH, have been proposed to explain the precipitation of carnotite (Boyle, 1984). Some of these changes may result from mixing of different ground waters and/or evaporation. However, deposition of the cementing minerals resulted mainly from fluctuation of the ground-water level and evapotranspiration in arid to semiarid climatic conditions in closed basins. Uranium was leached by meteoric waters from uranium-rich acidic igneous rocks. During migration downhill within valley-fill sediment the waters become more alkaline and saline due to progressive evaporation. Where the flow is stopped by basement highs, ground water is directed toward the surface where evaporation increases, inducing increasing salinity, CO_2 degassing, pH decrease, and finally breakdown of the uranyl carbonates (Mann and Deutscher, 1978). Uranium grade is generally higher close to the surface, indicating that evaporation plays a major role in precipitating uranium. For many of deposits carnotite is presently close to saturation, demonstrating that recent or even seasonal dissolution and remobilization of uranium may still occur in these deposits (Bowell et al., 2009).

Similar evapotranspiration processes may have taken place at any time since oxygenation of the atmosphere. Therefore, their absence before the Tertiary seems to be due only to the poor preservation of surficial deposits. The largest uranium deposits related to evapotranspiration processes are situated

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in tropical areas of the Southern Hemisphere: Langer Heinrich and Klein Trekkopje in Namibia, Yeelerie and Lake Mailand in Western Australia, and southern Argentina. As similar tropical conditions and uranium-rich granitic and volcanic rocks occur in tropical parts of the Northern Hemisphere, similar deposits should exist in addition to the few occurrences already known in Mauritania, Somalia, and the United States.

Conclusions and Perspectives

The evolution of uranium deposit types through time is subdivided into four periods. The first period from the Earth accretion to 3.2 Ga corresponds to a huge transfer of uranium during nearly 1.4 Ga by partial melting of the mantle, but the highest concentrations reached in the most felsic rocks (the TTGs) were limited to a few parts per million essentially hosted in accessory minerals, preventing further uranium mobilization to reach ore grade.

The transition from the first to the second period is marked by the change of geotectonic-thermal conditions during the Late Archean at about 3.2 Ga, permitting the generation of highly potassic and peraluminous granites able to crystallize uraninite and to form the first type of uranium deposit on the Earth: the quartz pebble conglomerates.

The third period began with the strong increase in oxygen content of the atmosphere at about 2.2 Ga, making possible the oxidation of U(IV) to U(VI) and the formation of an extreme variety of uranium deposits essentially related to redox processes. The largest ones are the unconformity related deposits occurring in the vicinity of the contact between a thick pile of continental to marginal marine oxidized sandstone, which has represented a vast diagenetic brine aquifer, and a Paleoproterozoic basement particularly rich in uranium.

The fourth period developed with the appearance of land plants during the Silurian, which created multiple redox barriers within siliciclastic continental successions. This change is responsible for the formation of the largest economic uranium resources on the Earth hosted by sandstones, deposited in epicontinental to marginal marine sediments, with more than 400 deposits. The exceptional conditions brought together during the Cretaceous along the southern margin of the Paleotethys Ocean, explain the genesis of huge phosphate resources which hosts nearly three-quarters of the world uranium resources associated with phosphates.

These four periods are relatively well established and linked to the preferential development of certain types of uranium deposits types at specific periods of the Earth's geologic history. Nevertheless, some major questions remain that require further research.

The first major problem is related to the appearance of large quantities of highly potassic granite able to crystallize uraninite at about 3.1 Ga. Were subduction processes able to produce the dehydration of the subducted sediments and selective enrichment of the mantle wedge in U, Th, and K by fluid metasomatism, or do we have to invoke another mechanism?

The geotectonic conditions responsible for the largest development of deposits related to Na metasomatism during the Paleoproterozoic is still unclear. In Ukraine, the genesis of such deposits requires the creation of huge thermal anomalies, with temperatures higher than 500°C, at the scale of more than 100 km, about 200 Ma after the end of the preceding orogenic event.

The Olympic Dam deposit, the largest economic uranium resource in the world, is also one of the least documented. Much more information needs to be published in order to understand the genesis of the uranium phases and their relationships to the iron oxide, copper, and gold phases.

The existence of intraformational deposits within continental sandstone older than Silurian should be reevaluated after increasing evidence of uranium reductants migrated from deep oil or gas reservoirs into these otherwise organic-free strata. In particular, the Amer Lake deposits in Nunavut hosted by weakly metamorphosed Paleoproterozoic (2 Ga) siliciclastic formations are worthy of study to identify the precipitation mechanisms for uranium in that setting.

Many believe that large uranium provinces are related to Archean and Proterozoic crystalline basement complexes. However, one of the largest uranium provinces of the world, mainly covering the territory of Kazakhstan, represents a very young uranium province, for which the ultimate source of the uranium is not very well constrained.

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