Unconventional uranium resources

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Note
In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all publications by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

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Unconventional uranium resources are deposits with low uranium concentrations, which. by virtue of their sheer size, constitute large, but very high-cost, uranium resources [IAEA 2001] Q149.
As NEA/IAEA [Red Book 2014] Q90 puts it:
Conventional resources are defined as resources from which uranium is recoverable as a primary product, a co-product or an important by-product, while unconventional resources are resources from which uranium is only recoverable as a minor by-product, such as uranium associated with phosphate rocks, non-ferrous ores, carbonatite, black shale and lignite. Most of the unconventional uranium resources reported to date are associated with uranium in phosphate rocks, but other potential sources exist (e.g. black shale and seawater).
Since 2009, a combination of expectations of rising medium-term demand and sustainability issues, have stimulated investigation of a variety of projects, extraction technologies and business models on the part of both governments and commercial entities. Interest in recovery of uranium from phosphates has been the primary focus for both economic and environmental reasons.
At present no unconventional uranium resources are being exploited. Some of these unconventional resources are reasonably well-known with respect to their size, grade and other qualities, but are not exploited yet for economic or other reasons: they are known, but unrecoverable resources.

This report briefly addresses some practical aspects of the recovery of uranium from the following unconventional resources:
- phosphorite deposits, also called phosphate rock
- black shales
- lignite & coal deposits
- granite
- carbonatite
- seawater.

Uranium from seawater is addressed in detail in a separate report L22p13 *Uranium from seawater*.

2 Uranium from phosphates

Phosphate rock

Phosphate deposits may be classified according to origin into the two main categories of sedimentary and igneous phosphate rocks, which, quite apart from their distinct characteristics as source of phosphate, also differ in terms of uranium content. Sedimentary deposits of marine origin have a uranium content in the range 40-300 ppm, and the igneous phosphate ores contain typically 10-100 ppm. The largest phosphate deposits of the world are in Morocco (55% of the world resources) and have an uranium content varying from 70-230 ppm [Bergeret 1979] Q47. Nearly all other known deposits contain less than 180 ppm uranium, most of them around 100 ppm or less (0.01% or less), although some small deposits could have a much higher uranium content. According to [Deffeyes & MacGregor] Q281 the phosphate rock occurrences in the earth's crust, together with shales, would contain some 600 000 Tg of uranium, at grades of 10-30 ppm.

According to [IAEA 2001] Q149, the world average uranium content in phosphate rock is estimated at 50-200 ppm (geometric mean 100 ppm). Marine phosphorite deposits contain averages of 6-120 ppm (geometric mean 29 ppm), and organic phosphorite deposits up to 600 ppm.

Table 1
Approximate uranium resources in phosphate rock, according to the [IAEA 2001] Q149, [Red Book 2014] Q90, see also [WISE-Uranium 2014] Q324. Tg = teragram = 10^{12} g = 1 million metric tons.

category	country	uranium content (Tg) 2001 [Q149]	uranium content (Tg) 2014 [Q90]
marine phosphorite	Morocco	6.9	6.5
	USA	1.2	0.14 - 0.33
	Mexico	0.15	0.24
	Jordan	0.1	0.06
	others (average)	0.65	0.39
	subtotal	9	7.33 - 7.52
organic phosphorite	Russia, Kazakhstan	0.12	0.058
	total	9.12	7.39 - 7.58

The IAEA [Red Book 2006] Q90 considers 22 Tg of uranium recoverable from phosphate rock; this figure is also cited in Red Book 2006 and following editions. The discrepancy of the figure of 22 Tg and those from Table 1 is striking. The Red Books 2006-2014 does not specify which deposits are concerned and at which grades those deposits are assumed to be recoverable. Data on phosphate rock deposits and their uranium content turn out to be extremely scarce in the open literature and the available data are not consistent.

A content of 1% phosphorus pentaoxide P_2O_5 in phosphate ore corresponds with 2.18% fluorapaptite $Ca_5(PO_4)_3F$.

The world phosphate rock reserves in 2006 were 18000 Tg, the world Reserve Base 50000 Tg, according to [USGS 2007] Q279. The Reserve Base includes the currently economic reserves (reserves), marginal reserves and subeconomic resources.

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Assuming a total global phosphate resource of 68 000 Tg the world average uranium content of phosphates would be:
according to [IAEA 2001] Q149: $G_{av} = 9.12/68000 = 134$ ppm
according to [Red Book 2006] Q90: $G_{av} = 22/68000 = 324$ ppm
The reasons why these figures are so widely divergent are unknown.

Extraction

Extraction of uranium from phosphate ores is not simple. Because of the low uranium content, the extraction of uranium has to be combined with the production of phosphoric acid, an essential chemical feedstock, e.g. for the production of fertilizers. The first stage of the process, the digestion of the ore, has to be carefully controlled to ensure a sufficiently high rate of extraction of the uranium from the phosphate rock into the phosphoric acid. Uranium is present in phosphate rock as a solid solution, without separate uranium mineral grains. The whole mass of phosphate rock has to be brought into solution to make uranium extraction possible.

After the acidulation process the raw phosphoric acid has to be purified and concentrated in uranium. From that concentrated solution, the uranium is extracted with solvents, highly toxic organophosphorus compounds. The process has high losses, partly due to reactions with hydrofluoric acid HF, needed in the extraction process. Reactions between HF and the organophosphorus compounds may yield very toxic organofluorophosporus compounds. In addition the formation of potent greenhouse gases is conceivable. No data are available on these aspects

To achieve a high efficiency of the extraction of uranium from the concentrated solution, a maximum of 70% is believed to be possible, the process conditions should be controlled within very narrow limits.
The overall yield of the extraction of uranium from the phosphate ore is difficult to estimate, because the yield of the each stage in the process is very susceptible to various process conditions. If a yield of some 70% is assumed in each of the three stages of the uranium extraction process, the overall yield would be around 30%.

Sulphuric acid consumption

Assumed that the most commonly mined phosphate rock is sedimentary phosphate consisting of fluorapatite $Ca_5(PO_4)_3F$, the quantities of sulphuric acid required to process the phosphate rock can be roughly estimated. The fluorapatite has to be treated with sulphuric acid to dissolve it:

$$Ca_5(PO_4)_3F(s) + 5\ H_2SO_4(aq) + 10\ H_2O \longrightarrow 5\ CaSO_4.2H_2O(s) + 3\ H_3PO_4(aq) + HF(aq)$$

Other metal ions, replacing some Ca^{2+} ions in the apatite, come into solution as well, or coprecipitate with the gypsum $CaSO_4.2H_2O(s)$. To bring the uranium ions into solution the conditions have to be carefully controlled.

From this equation we can calculate the stoichiometric sulphuric acid consumption at 0.97 Mg H_2SO_4 per Mg $Ca_5(PO_4)_3F$. Assumed that the phosphate in the ore consists of fluorapatite and that no other minerals in the ore are soluble in sulphuric acid, the minimum amount of sulphuric acid required to get 68 000 Mg/a uranium into solution would be $m(H_2SO_4)$ = 1692 x 0.97 = 1640 Tg/a. In practice the sulphuric acid consumption will be considerably higher than the stoichiometric minimum, due to incomplete reactions and reaction with other constituents of the phosphate rock. An amount of 1640 Tg/a is 8 times the world sulphuric acid production in 2006 of some 200 Tg [Peacock 2007] Q368. In 2006 nearly 70% of the world sulphuric acid went to the phosphate fertilizer production.

Energy balance

In phosphate rock no separate uranium minerals are present, so the whole mass of phosphate ore has to be brought into solution in order to extract the uranium.
The amount of phosphate processing needed to meet the current world uranium demand is 10-20 times the rate needed to supply the phosphorus demand for agriculture. In that case the energy consumption of the mining, milling and processing of phosphate rock should be attributed nearly fully to the recovered uranium. At a grade of 100 ppm U – most resources have lower grades – phosphate rock is beyond the energy cliff and beyond the mineralogical barrier, so phosphates cannot be regarded a net energy source: the nuclear energy system fed with uranium from phosphates will have a negative energy balance. For explanation see report m29 *Uranium for energy resources*.

If in the future phosphate rock would be exploited for uranium recovery, short-term economic and political considerations would prevail over thermodynamic arguments.
The high production costs, 50-200 \$/lb U_3O_8 (300-1200 \$(2000)/kg U), according to [Bergeret 1979] Q47, indicate a high specific energy and materials consumption even in coproduction with phosphoric acid. These costs likely will be much higher today.

Uranium and phosphate rock consumption

An amount of 68 Gg uranium, the world annual consumption in 2006, is present in 507 Tg phosphate rock at an assumed average grade of 134 ppm. The actual uranium production of 68 000 Mg/a would require the processing of some 1692 Tg/a (1.7 billion tons per year) of phosphate rock, assumed a recovery yield of 30% could be achieved. The world phosphate production in 2006 was 142 Tg.

The world uranium resources in phosphate rock would be sufficient for some 40 years of uranium supply at a consumption rate of 68 Gg/a, at an assumed recovery yield of 30%. So the uranium from these resources would run out within the lifetime of the present nuclear power stations.
If we assume the 22 Tg uranium stated by the IAEA would be recovered from phosphate rock with a uranium content of 127 ppm (the geometric mean of the Morocco deposits), 17300 Tg phosphate rock has to be mined and processed at a recovery yield of 100%. At a more realistic yield of 30%, some 57700 Tg of phosphate rock has to be mined and processed to extract 22 Tg of uranium.

To produce each year 68 Gg uranium, the current world uranium consumption rate, a theoretical minimum mass of 680 Tg/a phosphate rock must be processed at a uranium content of 100 ppm, a more realistic grade than 134 ppm in the previous example. The actual mass certainly will be much higher, due to an extraction efficiency considerably lower than 100%. At an extraction yield of some 30%, about 2000 Tg/a phosphate rock would have to be processed.

The world production of phosphate rock in 2006 was about 142 Tg. The world phosphate reserves are 18000 Tg, enough for 9-10 years of uranium supply at the current consumption rate. The world reserve base of phosphate is some 50 000 Tg, enough for another 25 years of uranium supply.
Even if the average uranium content would be twice as high as in this example, or if the extraction yield would be twice as high, the phosphate reserves would last roughly 20-50 years at the current consumption rate.

Marine deposits account for 80% of the world output of phosphate-based fertilizer products, and 70% of this total is converted into wet-process phosphoric acid, the base for the current uranium extraction process. Assuming an average recoverable content of 100 ppm of uranium, this scenario would result in an annual output of 3.7 Gg/a from 142 Tg/a phosphate rock, slightly less than 6% of the uranium consumption rate of 62 Gg/a in 2012.

No substitute for phosphorus in agriculture

The phosphate rock processing rate to meet the current world uranium demand would be 10-20 times the rate to meet the agricultural demand of phosphate.
Phosphate rock is the source of an essential agricultural fertilizer. There are no substitutes for phosphorus in agriculture. So the exploitation of phosphate rock should be tailored to the agricultural needs and to the demand for food production.

One of the deleterious effects of a fast exploitation of phosphate resources for uranium recovery is the fast degradation of the quality of the remaining phosphate resources. The richest and cleanest ores are always recovered first,so the ores with lower grades and more contaminants remain. This would result in fertilizers contaminated with more toxic and radioactive elements and a higher specific energy consumption per unit product.

3 Uranium from shales

Black shales

Black shales are a huge geologic reservoir of uranium, according to [Deffeyes & MacGregor 1980] Q281, containing some 30 000 Tg of uranium, at grades typically ranging between 30-100 ppm.
Uraniferous black shales are marine, organic-rich, commonly pyritic shale in which uranium (and other metals) is adsorbed on to the organic material and clay minerals. Average grades for the black shales deposits range between 50-400 ppm uranium, but because of their large areal extent they contain very large resources [IAEA 2001] Q149.

Examples are the Ranstad shales in Sweden, the Ronneburg shales in Germany and the Chattanooga shales in the USA. The Ranstad shales cover about 500 km² and contain approximately 254 Gg uranium at an average grade ranging between 170-250 ppm of uranium.
The Ronneburg shales cover about 164 km² and contain resources of 169.23 Gg uranium with grades between 0.085-0.17% U.
The Chattanooga shales in the southeastern USA cover some 80000 km² and contain uranium resources of 4-5 Tg at an average grade of 57 ppm [IAEA 2001] Q149.
For comparison: the annual consumption of natural uranium in 2012 was 62 Gg/a.

In 2013 no figures were reported on the Chattanooga shales and Ronneburg shales. Sweden reported a resource base of 968 Gg U in several shales/schist deposits and Finland reported 22 Gg U in shales and schists.

The uranium resources in the Chattanooga shales have been mapped by the US Bureau of Mines [BureauMines 1976] Q135, but found to be not economically recoverable. The Chattanooga shales consist of two principal uranoferous layers, each with a variable thickness of 1-2 meters. The richest layer (Gassaway Member) has an average grade of 60 ppm uranium and contains some 3 Tg (million tonnes) uranium. The other layer has grades in the range of 20-50 ppm and contains some 4.5 Tg uranium [BureauMines 1976] Q135. In these shales the uranium ions are bound to the organic material and for that reason these resources are beyond the mineralogical barrier (see report m29 *Uranium for energy resources*).

Practical aspects

Recovery from the rich layer of 162 Mg uranium – consumed by an advanced 1 GW reactor during 1 year and corresponding with 25.86 PJ gross electricity production – requires the mining and processing of 5.4 Tg shales at an average recovery yield of 50%. This amount would correspond with 1.4 km² to be mined per reactor per year, if the density of shales is taken at d = 2.6 Mg/m³ and the average layer thickness at h = 1.5 m. To feed the current world nuclear fleet some 560 km² of the shales have to be mined annually, a square of 24x24 km.
A coal fired plant consumes about 2 Tg/a of coal to generate the same amount of electricity.
Applying the average energy input of mining and milling (J_{m+m} = 2.33 GJ/Mg ore, see report m26 *Uranium mining + milling*) the total energy input per year per GW would be 12.6 PJ, or half of the energy output of the reactor. Likely the recovery yield would turn out much lower. At a value of say 25%, the energy balance of the uranium recovery alone would drop to zero. Obviously, the exploitation of the lower-grade layer of the Chattanooga shales would fall well beyond the energy cliff (see report m29 *Uranium for energy resources*).

The study of the Bureau of Mines [BureauMines 1976] Q135 itself also put some doubts on the energy input-output ratio of LWRs feeded by uranium from the shales and recommended further work that should concentrate on:

- investigating into the net change in energy balance that will occur if and when lower grade ores (60 ppm rather than 2000 ppm uranium) are used as a source of nuclear fuel
- developing more geologic data on the Chattanooga formation
- determining the sociological and environmental impact that will occur if the shale is to be mined and processed for its uranium.

As the authors of the study put it:

> “Currently, steady-state Light Water Reactors (LWR) have an energy input-output ratio of approximately 1 to 15 when using uranium derived from high-grade deposits. If the Gassaway [. . Member of the Chattanooga Shale . .] were utilized (60 ppm uranium) in the absence of more economical uranium deposits, this ratio would change. A preliminary study is needed to determine the increased ammount of energy input needed to mine, mill, and process the Chattanooga shale versus higher grade deposits and analyze how this would affect the energy ratio of LWR’s.”

From processing point of view shales can be taken as soft ores. Mining and milling of shales would be conventional processes. The Chattanooga shales would be mined underground. No pilot plant operations have been undertaken. In laboratory tests a recovery of uranium from the ore of 34-80% are achieved, depending on method, amount of reagent and number of extraction steps [BureauMines 1976] Q135. No data are given on the recovery from the resulting solution. So, the overall yield is unknown, but may be comparable to that from phosphates.

4 Lignite and coal deposits

Lignite and sub-bituminous coal deposits often contain uranium adsorbed on the carbonaceous material or as urano-organic complexes. The average uranium content is typically only a few 10s of ppm of U. Uraniferous lignite deposits are typically small, but deposits in Kazakhstan and China reportedly range between 20 and 50 Gg U.

Burning the lignite converts the uranium adsorbed on the organic material into a refractory uranium silicate, from which uranium extraction is complex and expensive.

There are no systematic resource calculations for uranium hosted in lignites. Speculative Resources are probably in the millions of Mg of uranium in lignites worldwide, but because of their high production costs these resources are of limited practical interest, according to [IAEA 2001] Q149.

5 Uranium from granite

The uranium content of granitic rocks typically varies from 3-20 ppm U (0.0003-0.002% U or 3-20 gram U per Mg rock), with an average value of 4 ppm (0.0004%). Roughly, the average composition of granite is 80% SiO_2 and 20% Al_2O_3. For the mining and milling process granite is a hard ‘ore’.

To fuel the reference reactor, each year about 162 Mg natural uranium has to be extracted from the earth’s crust. The mass of 162 Mg uranium is present in 40 Tg of granite, with an average uranium content of 4 gram U per Mg rock. The rock has to be dug up, ground to fine powder and chemically treated with sulfuric acid and other chemicals to extract the uranium compound from the mass. Assumed an extraction yield of Y = 0.50, an optimistic assumption, 80 Tg granite have to be treated. This is a block 100 meters wide, 100 meters

high and three kilometers long. Each year, for one reactor.
For comparison: a coal-fired power station of 1 GW(e) consumes about 2 Tg of coal each year.

Substituting c = 0.654 GJ/kg(U) for hard ores, we find a total energy (thermal + electric) requirement of:
$J_{th} + J_e$= 3.27 TJ/kg(U)
As the ratio thermal/electric energy is R = 1.6 for hard ores, the electric requirements alone are:
J_e = 1.26 TJ/kg(U)
To extract 162 Mg uranium from granite about 204 PJ electricity would be consumed, plus some 326 PJ thermal energy (fossil fuels). The gross electricity production possible from this amount of uranium is 25.86 PJ, ignoring the energy requirements of the other processes of the nuclear chain.

6 Uranium from carbonatites

Carbonatites are intrusive carbonate-mineral-rich igneous rocks, many of which contain distinctive abundances of apatite, magnetite, barite, and fluorite, that may contain economic or anomalous concentrations of rare earth elements, phosphorus, niobium, uranium, thorium, copper, iron, titanium, barium, fluorine, zirconium, and other rare elements. They may also be sources of mica or vermiculite [USGS 1995] Q88.

Minable carbonatite deposits typically range in size from approximately 6 to 300 million metric tons. Most carbonatite deposits are likely to be mined on a large scale in open pit mines. Potential impacts include radioactivity from uranium and (or) thorium in waste rock or sediment; dust from mining activities; acid- and (or) metal-contaminated drainage from pyrite-rich carbonatites; and possible water contamination from spillage or leakage of chemical solutions used to leach and process ore. Asbestiform amphiboles, such as riebeckite, that are present in waste dumps associated with some carbonatite deposits may pose another health risk [USGS 1995] Q88.

According to the NEA/IAEA [Red Book 2014] Q90 only Brazil and Finland have reported recoverable uranium from carbonatites with a total of 15.5 Gg U. This would be one quarter of the present annual consumption of 62 Gg U/a.

7 Uranium from seawater

Technically it is possible to extract uranium from seawater. The first stage of the extraction process is the adsorption of the complex uranium ions $[UO_2(CO_3)_3]^{4-}$ on solid adsorption beds. For reason of the extremely low uranium concentration (c = 3.34 mg/m^3) and the relatively high concentrations of a great many other dissolved chemical species in seawater, highly selective adsorbents are required.

Broadly two adsorption methods have been investigated, one is based on titanium hydroxide $Ti(OH)_4$, the other is based on a special graft polymer. The first method has been studied in the 1970s, leading to the conclusion in 1980 that it was not viable [INFCE-1 1980] Q226. The polymer-based method has been investigated in the 1990s and 2000s by a Japanese group, e.g. [Nobukawa 1994] Q72, [Sugo et al. 2001] Q298, [Seko et al. 2003] Q312, [Sugo 2005] Q303 and [JAERI 2005] Q304.
NEA/IAEA [Red Book 2014] Q90 reports:

> “Seawater has long been regarded as a possible source of uranium due to the large amount of contained uranium, over 4 Tg U. However, because of the low concentration of uranium in seawater (3-4 parts per billion), developing a cost-effective method of extraction remains a challenge.
>
> Research on uranium recovery from seawater was carried out in Germany, Italy, Japan, the United Kingdom and the United States from the 1950s through the 1980s and more recently in Japan. In 2012, researchers at the US Department of Energy’s Oak Ridge National Laboratory and Pacific Northwest National Laboratory reported encouraging results through the use of innovative improvements to Japanese technology tested in the late 1990s (Ferguson, 2012). Although not commercially oriented, the goal of the research is to determine the minimum cost of a virtually limitless supply of uranium in order to guide future fuel cycle decisions. Many Chinese research groups in universities and institutions have also shown interest in uranium extraction from seawater.”

The Japanese studies are based on gram-scale experiments with the adsorbent. As far as known no trials with a pilot system under realistic conditions have been performed, not even regarding a part of the required system, let alone with an integrated system. In addition, some crucial parameters have been ‘upgraded’ apparently without a solid empirical base.

The upscaling of a complicated, unproven technical and chemical process with a factor of ten billion might be a real challenge. Any cost estimate of uranium from seawater at the current state of technology and experience have anything but little meaning.

History shows that we should be prudent with technical optimism, especially regarding very large and very complicated systems of unproved technology. The studies [RAND 1979] Q127 and [RAND 1981] Q126 concluded that the cost of new large projects of energy technology always are underestimated. Their findings are far from outdated. The mechanisms leading to seriously underestimating the cost of a large technological project as described in the RAND studies, perfectly apply to the Japanese cost estimation of uranium from seawater.

Uranium from seawater is addressed in detail in a separate report m28 *Uranium from seawater*.

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