Uranium as a renewable for nuclear energy

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ABSTRACT

Uranium extraction is the first step of the nuclear fuel cycle. Currently, uranium is only extracted from solid ores such as uranium rich minerals (% level) or minerals such as phosphates (ppm level). For some years extraction of uranium from sea water (ppb level) has been the topic of investigations particularly in Japan due to its national interest. In the huge oceanic volume the amount of uranium is constant, regulated by its river input (soluble) and balanced by its scavenging (particulate) on the sea floor. This work shows that the uranium extraction with parsimony from sea water could be carried in a renewable way if its concentration remains quasi constant. Recommendations for the extraction with use of gel panels or with braid of fabric grafted by sorbing groups in high tide or oceanic pelagic current environments are suggested along with a reduction of the uranium consumption.

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1. Introduction

Presently, the uranium needs are evaluated for today's reactor fleet totaling 430 units to be 65,000 t of originally natural uranium a year (Emsley, 2014). This current demand is mostly filled by uranium production which is mostly issued from the mining exploitation of ores whose contents range from say about 50 (or more) to 0.05 wt% e.g. Frondel and Fleisher, 1950; Nininger, 1954.

The International Atomic Energy Agency (IAEA) assigns uranium deposits as types, based on their geological setting and genesis of mineralization. These deposits are arranged according to their approximate economic significance from high to relatively lower uranium fraction.

Primary uranium minerals are uraninite or pitchblende UO_2, coffinite $U(SiO_4)_{1-x}(OH)_{4x}$ and brannerite (UTi_2O_6) with uranium fractions of 88.1, 70.4 and 59.8 wt% respectively. The secondary uranium minerals are autunite $(Ca(UO_2)_2(PO_4)_2 \times 8-12\ H_2O)$, carnotite $(K_2(UO_2)_2(VO_4)_2 \times 1-3\ H_2O)$, torbernite $(Cu(UO_2)_2(PO_4)_2 \times 12\ H_2O)$ and uranophane $(Ca(UO_2)_2(HSiO_4)_2 \times 5\ H_2O)$ and with respective uranium fraction below 50 wt%.

Deposits also include uranium containing formations of sandstone, quartz, phosphorite, lignite and black shale deposits, locally in form of breccia, veins metamorphic, intrusive or volcanic. Here the uranium fraction decreases below 1 wt% and exploitation can be extended to fractions dropping to the 0.01 wt% level.

The extraction practice is widely established with various strategies ranging from the excavation in open pit (OP) e.g. in Namibia, Niger and Australia to deep tunneling (underground, UG) such as in Canada or Russia, through ore rich formations. In addition to physical exploitation, chemical treatments such as in situ leaching (ISL) may be used as in Kazakhstan. The largest-producing uranium mines in 2013 were: McArthur River, Canada (13%, UG), Olympic Dam, Australia (6%, UG), Somair, Niger (5% OP), Tortukuduk, Kazakhstan (4%, ISL), Priargunsky, Russia (4%, UG) Langer Heinrich, Namibia (4%, OP) and Ranger, Australia (4%, OP), producing a total of 59 kt (100%). The global nuclear fuel market reached a total world production of 59,370 t of U (or 70,015 t of U_3O_8) that is 92% of the worlds demand (WNA, 2013).

The uranium in term of U_3O_8 production has in the meantime increased steadily from 47 kt in 2006 to 70 kt in 2013 (See Fig. 1). The demand could, however, reach 100 kt with today's nuclear fuel cycle practice i.e. open cycle (mostly) with light water reactors (LWR).

Uranium exploitation may be performed as long as the extraction is economically viable whilst also considering ecological issues. The known recoverable uranium resources were evaluated reaching a total of 5.5 Mt (OECD, 2011, 2014). Basically countries such as Australia, Kazakhstan, Russia, Canada and Niger are key players while 9 other countries well spread in the world have significant potential (above 1%).

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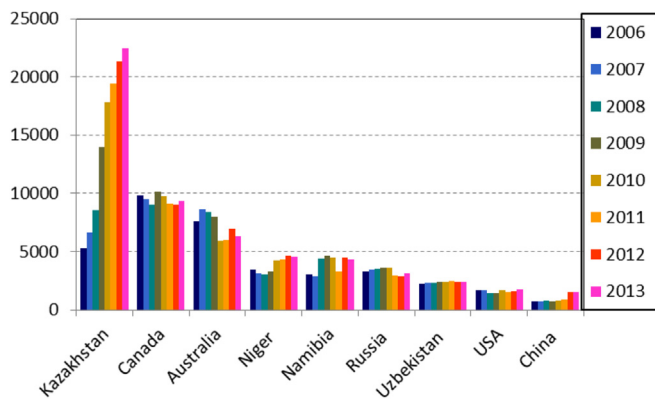
The reasonably assured resources, at US\$ 130 per kg of U, (Australia 31%, Kazakhstan 12%, Russia 9%, Canada 9%, Niger 8%, South Africa 5%, Brazil 5%, Namibia 5%, USA 4% and China 3%), would be 5,327,200 tons of U (OECD, 2011). The total at US\$ 260 per kg of U is 7,096,600 tons U, and Namibia moves up ahead of Niger. The total resources were reevaluated recently (OECD, 2014) suggesting a 10% increase compared to 2011.

In addition to the 5.4 million tons of uranium in known recoverable resources, there are substantial amounts comprising what are known as "unconventional resources".

For about 50 years the extraction of uranium from large **phosphate** deposits has been proposed as an alternative production. The U content in the phosphate ores currently ranges from 10 to 100 ppm and the U extraction has been applied as a purification step (decontamination) e.g. Hurst 1989. Prayon, Belgium (Davister and Martin, 1989) has applied this extraction over about 20 years (1978−1998). In 1998 the extraction was discontinued because the uranium price was too low. Consequently, Prayon changed the import source of phosphate towards less U rich ores. The world phosphate annual production includes China (65 Mt), Morocco & W Sahara (26 Mt), USA (26 Mt), Tunisia (7.6 Mt), Jordan (6 Mt), Brazil (5.5 Mt) and Egypt (5 Mt) as top producers. The world production reached 176×10^6 t per year in 2010. With an extraction of the 30 ppm level of uranium the production could reach potentially 5×10^3 t a^{-1}. Estimates of the available total uranium amounts range from 9 to 22 (average 15) Mt.

Also among the "unconventional resources" are **coal** derived products such as fly ash. The best estimates of the total amount of coal (including undiscovered coal) in the world are about 12 trillion tons (Devereux Carter, 1976) i.e. 12×10^{12} t. The total of U resource includes several categories of coal with various degrees of geologic assurance and data reliability. If one estimates the average uranium concentration to be 5 ppm then a total amount of 6×10^7 t of U may be derived from these values. Uranium from coal residues is a topic of R&D in various countries. As an example Mongolia uranium recovery from coal ash dumps has been recently investigated (Maslov et al., 2010). Decontamination (U, Th and decay products) of the ashes for other uses (building material) is an important point and the recovery of uranium is also possible. With the coal mining rate of 10^9 t a^{-1} and an extraction of the 5 ppm level of uranium the production could also reach potentially 5×10^3 t a^{-1}.

These extractions from solid ores and as by-products cannot be labeled as renewable. In addition since the ore treatment induces potential releases of natural radioelements and of chemical reagents they generate tailings that must be managed carefully.
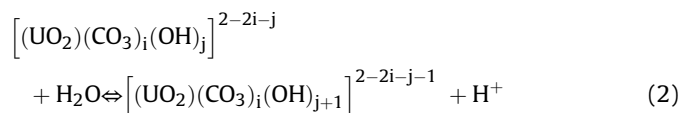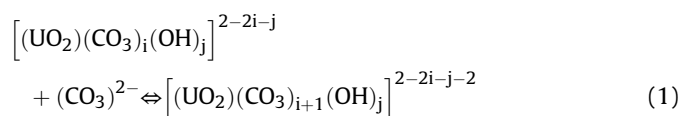
Renewable energy is derived from natural processes that are replenished constantly. Included in the definition is heat generated, electricity and derived hydrogen from sunlight, wind, oceans, hydropower, biomass, geothermal resources and biofuels. From today's nuclear fuel cycle options it may be shown that with the practice of reprocessing and recycling as well as with the use of actinide as burnable poison, nuclear energy could be qualified sustainable. However, from sustainable to renewable the need of a refurbishable source of fuel is required such as for biomass. In this work uranium in sea water is revisited focusing on the very large source properties with assured replenishment. This concept was already suggested and approached in a semi-quantitative way for example by Cohen, 1983. The purpose of this review and contribution study is to investigate and assess the limitations on the extraction of uranium from the sea and on its use to gain the label renewable.

2. Phenomenological approach

To understand the behaviour of uranium in the sea water it is important to depict its speciation including its sorption on particles prior to evaluating the input and output fluxes from the dissolved state that yield a quasi-equilibrium state in the sea.

2.1. Uranium speciation in aquatic systems

Starting with U(VI) as uranium redox state and UO_2^{2+} as basic species, the complex species that form in aqueous solution are hydroxide and carbonate complexes Ref. Saito and Miyauchi (1982) and Djogic et al. (1986). Uranyl complexes with fluoride − sulphate − phosphate and organics, e.g. oxalate, are ignored because of the low ligand concentration or their low complexation constant. The discrete formation reactions of the hydroxide − carbonate mixed complexes are:

$$\left[(UO_2)(CO_3)_i(OH)_j\right]^{2-2i-j} + (CO_3)^{2-} \Leftrightarrow \left[(UO_2)(CO_3)_{i+1}(OH)_j\right]^{2-2i-j-2} \tag{1}$$

$$\left[(UO_2)(CO_3)_i(OH)_j\right]^{2-2i-j} + H_2O \Leftrightarrow \left[(UO_2)(CO_3)_i(OH)_{j+1}\right]^{2-2i-j-1} + H^+ \tag{2}$$

The carbonate-, hydroxide- and mixed complex concentration can be quantified using the stability constants (partial: K or cumulative: β) according to the following equations:

$$K_{(i+1,j)} = \left[\left[(UO_2)(CO_3)_{i+1}(OH)_j\right]^{2-2i-j-2}\right]\left[\left[(UO_2)(CO_3)_i(OH)_j\right]^{2-2i-j}\right]^{-1}\left[(CO_3)^{2-}\right]^{-1} \tag{3}$$