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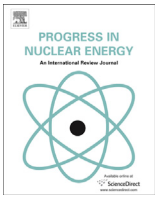

Could spent nuclear fuel be considered as a non-conventional mine of critical raw materials?

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A B S T R A C T

Each year, more than 10 thousand tons of spent fuels are discharged from nuclear power plants in the world. Heavy element nuclear fission reactions, at the origin of energy production, generate fission products of intermediary mass, some of them being considered nowadays as critical raw materials. The potential interest to treat these spent fuels in order to recycle these elements has risen recently following increasing international tensions on their supply for industry and energy. A study was carried out on the basis of the French nuclear fuel cycle scenario in order first to evaluate the inventory of such metals in spent fuel. The only elements of interest, since in significant amount, would be rare earth elements (REE) and platinum group metals (PGM). However, compare to the annual need of REE, the amount that would be recovered from spent fuels represent less than 0.01% of the annual world production. Because of the low price of these elements, there is no economic interest for such a recovery. The case of PGM, and specifically ruthenium and rhodium, is quite different. Even if a lower amount of these elements are in spent fuel, it represents 22% for Ru and 3.5% for Rh of the annual world production. The drawback is that these elements have numerous radioactive isotopes that forbid using them for industrial applications. 20–50 years of storage after separation would be necessary for ruthenium and rhodium to get a radioactivity level lower than potential clearance levels. Before any industrial use, very efficient separation processes would be required to selectively recover these elements. The physico-chemical forms of these elements in the spent fuel make the work tricky. Finally, such a use would require the official existence of a clearance level for nuclear materials as recommended by the IAEA.

1. Introduction

Over the last century, we observed a very strong diversification of raw materials used in industrial products: more than sixty elements are nowadays regularly used in the industrial production whereas only eight were regularly used at the beginning of the XXth century (Fe, Cu, Zn, C, Ce, Sn, Sb, Pb). They are playing a key role for the development of industry and the sustainability of the economy. They are used not only in information technologies (ICT) such as in microprocessors, smartphones, LCD screens, but also in the energy sectors, such as in batteries, low energy light bulbs, or alternative energies production techniques such as solar panels and wind mills (Table 1). Their consumption increases year after year with the development of technologies requiring more and more high specification materials. Many industrial sectors highly depend on them although they may be used in very small quantity (100–1,000t produced each year to answer the whole world needs). Some of these elements have very specific properties and they strongly modify the properties of the materials so that they are in many cases indispensable and not substitutable. Equilibrating their production and the industrial needs is therefore a challenging and mandatory task in order not to hinder the industrial and economic development.

These raw materials are unfortunately not homogeneously distributed on Earth and although many of them are not scarce on Earth, they are definitely not abundant in Europe and their supply highly depends on their importation. A great share of the worldwide production is concentrated in a few countries, among them China. For the past decade, many countries have realized how dependent they have become on foreign imports to access these raw materials, which represents a major threat to national industry and global competitiveness. It yields to define the category of

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Table 1
Critical materials found in emerging technologies (Harry Atwater and Otten, 2011).

Technology	Component	Material
Wind	Generators	Neodymium Dysprosium
Vehicles	Motors	Neodymium Dysprosium
	Li-ion Batteries (PHEVs and EVs)	Lithium Cobalt
	NiMH Batteries (HEVs)	Rare Earths: Cerium, Lanthanum, Neodymium, Praseodymium Cobalt
PV Cells	Thin Film PV Panels General	Tellurium Gallium Germanium Indium Selenium Silver Cadmium
	CIGS Thin Films	Indium Gallium
	CdTe Thin Films	Tellurium
Lighting (Solid State and Fluorescent)	Phosphors	Rare Earths: Yttrium, Cerium, Lanthanum, Europium, Terbium
Fuel Cells	Catalysts and Separators	Platinum, Palladium and other Platinum Group Metals, Yttrium

critical raw materials which corresponds to elements having a high economic importance combined with a high supply risk or a high dependence on a limited number of foreign countries. We will later refer to them as critical raw materials (CRM). Today's challenge lies in securing their supply at affordable prices to maintain manufacturing industries and support the necessary development of technologies in a sustainable way. Such a situation is specifically relevant in Europe which does not have any significant mining activities anymore. Many European stakeholders hence decided to address the question of the long-term sustainable supply of critical raw materials. European Commission regularly updates a list of CRM at the European level, the last release from 2014 being given in Table 2. For some of these elements, it is foreseen that as early as 2030, the demand will be higher than the production.

This international mobilization of experts also allows having a relatively precise assessment of the current and future needs as evidenced by the indicators presented in Table 3.

To anticipate this growing need, the recycling of raw materials and the research of new “non-conventional mines” are under study. Today, some metals are efficiently recycled (steel, copper, aluminium but also platinum group metals from mufflers, cell phones or lap-tops) (European Commission, 2011). The development of such a recycling strategy should be widely extended. In parallel, new mining fields are considered such as coal ashes, waste from metallurgical industry, or water from desalination plants …). In this global framework, spent nuclear fuel has also been questioned as being a potential non-conventional mine of critical raw materials (Hazelton et al., 1986; Hecht, 1986; Sano et al., 2004). Indeed, nuclear fission of heavy elements leads to the formation of most of the critical raw elements thanks to the fission reaction, and French industry AREVA has demonstrated since the 80's that treating spent nuclear fuel to recycle some of the elements of interest (in this case U and Pu) can be safely, properly and efficiently managed at the industrial scale. The opportunity of recovering critical raw materials in spent nuclear fuel can hence not be ruled out and needs to be precisely and exhaustively addressed, which is the aim of this paper.

Table 2
List of 20 critical raw materials at EU level (in alphabetical order) (European Commission, 2014).

Antimony	Gallium	Magnesite
Beryllium	Germanium	Niobium
Borates	Graphite	PGMs
Chromium	HREE	Phosphate rock
Cobalt	LREE	Silicon metal
Coking coal	Indium	Tungsten
Fluorspar	Magnesium	

This paper aims to address the different aspects of this complex and multi-face questions in order to assess the relevance of considering, or not, spent nuclear fuels as a potential non-conventional mine for critical raw materials. It will first identify what are the relevant critical raw materials to focus on based on the spent nuclear fuel inventory and their annual production. Second, it will assess their anticipated long-term radioactivity that is subsequently compared to the current regulations for radioactive materials handling and re-use. Finally, the feasibility and the efficiency of potential separation processes are discussed.

2. Methodology used to define the potential critical raw materials of interest in the spent nuclear fuel

In order to identify the potential elements of interest within the spent nuclear fuel, a rationale stepwise approach was implemented: (i) identification of the elements that are produced in a sufficient amount to be potentially of industrial interest by comparing spent nuclear fuel inventory with the European and world market for the elements of interest, (ii) selection of those relevant in terms of mid-term radioactivity (on a timeframe 1–100 y.), (iii) assessment of the relevant decay time allowing them to be handled and reused, (iv) review of the potential separation processes that could be implemented to recycle such materials. The precise methodology is detailed below while the results are presented in the next section.

2.1. Methodology used to assess the overall spent nuclear fuel inventory and the potential elements of interest

Nuclear fuel is composed of uranium oxide enriched up to 4–5% in ^{235}U, the fissile isotope of uranium (UOX fuel). Uranium oxide can also be mixed with plutonium oxide coming from the reprocessing step to produce MOX fuel. During the 4 years of irradiation in nuclear reactor, fissile isotopes are progressively fissioned. This fission (i) produces a tremendous amount of energy which is used to produce electricity and (ii) forms two lighter atoms of intermediary