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

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ABSTRACT

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 world-wide 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

Table 3
Global demand in critical elements for emerging technologies in 2006 and 2030 compared to the current production (European Commission, 2010) from Ad-hoc working group on defining critical raw materials (2010)]. No reliable data has been found on Ru in the literature.

Raw material	Production 2006 (t)	Demand from emerging technologies 2006 (t)	Demand from emerging technologies 2030 (t)	Indicator[a] 2006	Indicator[a] 2030
Gallium	152	28	603	0.18	3.97
Indium	581	234	1911	0.40	3.29
Germanium	100	28	220	0.28	2.20
Neodymium	16,800	4000	27,900	0.23	1.66
Platinum	255	Very small	345	0	1.35
Tantalum	1384	551	1410	0.40	1.02
Silver	19,051	5342	15,823	0.28	0.83
Cobalt	62,279	12,820	26,880	0.21	0.43
Palladium	267	23	77	0.09	0.29
Titanium	7,211,000[b]	15,397	58,148	0.08	0.29
Copper	15,093,000	1,410,000	3,696,070	0.09	0.24

[a] The indicator measures the share of the demand resulting from driving emerging technologies in total today's demand of each raw material in 2006 and 2030.
[b] Ore concentrate.

mass, the fission products: first, a group of elements of atomic mass around 140 ± 15 (mainly the rare earth elements) and second a group of elements around 90 ± 15, among them the platinum group metals and the noble metals (Fig. 1).

Spent nuclear fuel composition depends on many parameters and varies from one fuel to the other. Among the main parameters, one can mention (i) the initial composition of the fuel and in particular the nature and the content of the initial fissile element, either a purely uranium oxide (UOX fuel), or a mixed uranium/plutonium oxide (MOX fuel) where the plutonium is recycled from previously irradiated fuel, (ii) the irradiation conditions, in particular the irradiation time, the neutrons fluxes and the reactor design and finally (iii) the cooling time after irradiation. Practically, spent nuclear fuel is characterized by its burnup, a global metric which quantifies the amount of energy produced per mass unit, and hence the extent of the fissile material consumption. In order to have a global assessment of the spent nuclear fuel inventory, a reference type of fuel has to be selected.

Based on the current industrial practice, we selected a representative UOX spent nuclear fuel with an initial U enrichment of 4% ^{235}U, a burnup of 47.5GWd/t, three cycles of irradiation of 400 days and a cooling time of 10 years after the end of the irradiation. Such a composition corresponds to the average of the spent fuel currently reprocessed in the French La Hague reprocessing Plant. The composition of this representative fuel was calculated by using the CEA DARWIN 2.4.0 code (Marimbeau, 1998; Tsilanizara, 1999; San Felice et al., 2013), with the JEFF3.1.1 database (Santamarina et al., 2009). This representative composition was subsequently used to assess the yearly amount of critical elements that is produced in reactor and could be recycled from the discharged spent nuclear fuels. The total amount of spent nuclear fuel discharged annually from reactors is considered to be 1,200t for France, and 10,500t worldwide (International Atomic, 2011).

Finally, a first selection of critical elements of interest was performed by comparing (i) the available inventory which could be yearly recovered from spent nuclear fuel with (ii) the annual world production. Annual production of critical elements of year 2014 was considered for comparison and we considered a threshold of 0.01% for selecting the elements of potential interest.

2.2. Assessment of the relevant critical elements in terms of radioactivity

Fission products can be either stable or radioactive, with half-

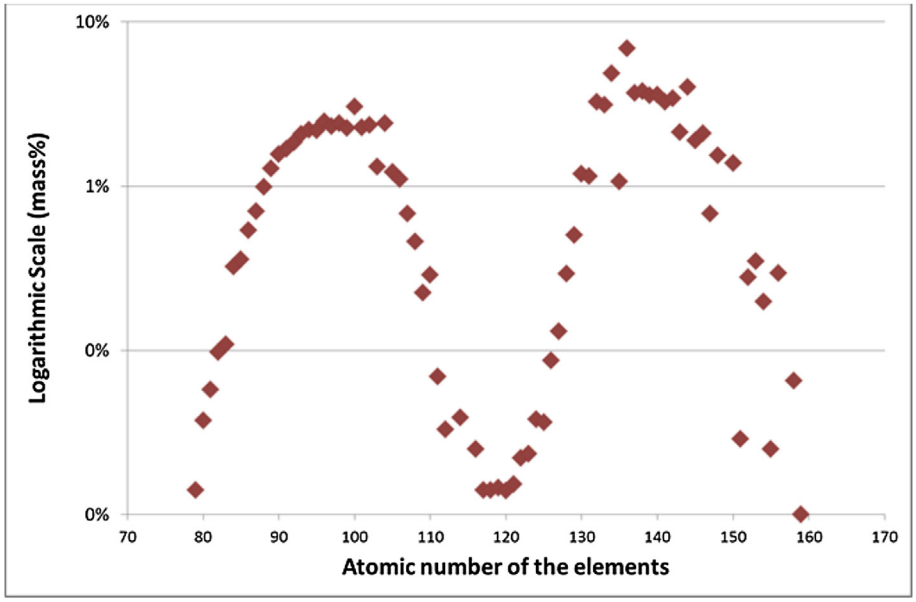

Fig. 1. Distribution of the fission products in the UOX spent fuel after 4 years of irradiation at 47.5GWd/t. The composition was calculated by using the CEA DARWIN 2.4.0 code (Marimbeau, 1998; Tsilanizara, 1999; San Felice et al., 2013) with the JEFF3.1.1 database (Santamarina et al., 2009).

life ranging from a few microseconds to millions of years. Most of them have several isotopes with different lifetimes that could hardly be separated. Therefore, many fission products are a cocktail of several isotopes. These isotopes decay down to a stable isotope in several steps in which different isotopes/elements with different half-lives are involved. As long as a given element is not separated from the initial material, only the parent isotope will effectively decay. The intermediary isotopes stay in equilibrium with their parents and daughters. This is not true anymore when the elements are separated from the initial material. Once separated, each isotope starts to decay, according to its radioactive period, giving daughters that can also decay until equilibrium is reached. Such a complexity needs to be accounted when addressing the long-term radioactivity of the elements of interest.

In order to assess the overall radioactivity of the elements of interest, activities were calculated as a function of time starting from 10 years after irradiation as explained before. Decay and activity calculations have been performed with the online software "Universal Decay Calculator, Version B" with the Oak Ridge database developed in the field of the WISE Uranium (Wise Uranium Project). Calculations were made by taking into account all the isotopes identified in the DARWIN code.

Very short-lived isotopes are very radioactive but they decay very fast. They do not contribute to the activity of an element on the long-term, except if they have a radioactive daughter with a relatively long half-life. At the opposite, the activity of long-lived isotopes will not decay significantly at the scale of a potential industrial use. These long lived isotopes will generate a stable "radioactive baseline". The case of short or medium-lived isotopes is much more hindering because these isotopes will generate high but decaying radioactivity for tens of years. Three examples are given below to illustrate the different cases.

- In the case of cerium, 0.0016% of the total Ce inventory is ^{144}Ce with a half-life of 284 days. It dominates the total cerium radioactivity and yields to an activity of 2TBq/g, 10 years after irradiation when it could potentially be recovered from spent nuclear fuel. However, this activity would drop to 70Bq/g after only 20 years of interim storage after separation and to 2.10^{-10}Bq/g after 50 years. If needed, cerium could hence reasonably be recycled after a decay interim storage.
- In the case of samarium, 1.13% of the total Sm inventory is ^{151}Sm with a half-life of 88 years which dominates the total activity of samarium for centuries (well above 1TBq/g), requiring irrelevant centuries of interim storage before any potential industrial use.
- In the case of neodymium, 34% of the total Nd inventory is ^{144}Nd with a half-life of $2.29.10^{15}$ years which determines the total activity of Nd around 14mBq/g for millions of years. Nd could be recycled if an exemption threshold would be defined.

For this study, only aggregated results will be presented for each element of interests by summing the contributions of the different isotopes.

2.3. Assessment of a potential decay interim storage time

As evidenced in the previous example, any recycling of critical raw materials from spent nuclear fuel would be possible if a preliminary decay interim storage is implemented in order to allow the radioactivity level to be low enough for any potential reuse. This issue can be easily addressed by comparing the residual radioactivity of the separated critical elements as a function of time to a threshold that would allow declassifying the material as non-nuclear. Indeed, the reuse of any materials coming from nuclear industry is only possible if a clearance level is defined and implemented by the safety authority, *i.e.* a threshold below which a material coming from the nuclear industry can be considered as non-harmful for the general public and reused without any specific precaution. The situation clearly differs from one country to another one.

- In France, such a clearance or exemption level does not currently exist whatever the radioactivity of the material. The French Code of Public Health (Code de Santé Publique) forbids the use out of the nuclear industry of any material coming from buildings, materials, or waste coming from any nuclear facility when these products are contaminated or potentially contaminated (art R-1333-3). Dispensations can be given if the advantage to use these products is higher than the potential health risk (art R-1333-4). These dispensations are delivered by the Health Ministry on advice of the Nuclear Safety Authority (ASN) and of the High Council for Public Health. Under these restricted conditions, a limited recycling may be possible and it has been implemented for instance for lead which has been recycled out of the nuclear industry with a clearance level of 0.5Bq/g.
- In Germany, such clearance levels exist for the recycling of some materials coming from the nuclear industry, based on European or international (IAEA) guidelines.

In front of such a diversity of situations, we based our analysis on the international agreed standard, namely the clearance levels proposed by IAEA in 1998 (International Atomic, 1998). Thresholds proposed by IAEA range between 10^4Bq/g as for Er and Pm, and 10^2Bq/g as for some transition metals such as Mo and Tc. In order to conservatively assess the potential interest of such critical elements, we consider in our calculations a general threshold of 10^2Bq/g.

3. Results of the potential critical elements that are of interest within spent nuclear fuels

Table 4 gathers data for the elements from the European critical list published in 2014 that are present in the spent fuel at more than 1 ppm and for which the amount that could be recovered annually from the French reprocessed spent fuel is over 0.01% of the world production. We found in this list the REE from cerium to gadolinium, the PGM (Ru, Rh and Pd). We compare in this table the 2014 annual world production (United States Geological Survey) with the potential French production from the reprocessed spent fuel, with the potential production if all the spent fuel discharged annually in the world would be reprocessed and with the potential reserve stored in all the spent fuel storage facilities in the world since the beginning of the nuclear energy production (290,000 tons – here an average burn-up of 33GWday/t and a cooling time of 30 years were considered).

Following sections discusses the potential interest for each family of critical elements.

3.1. Rare earth elements (REE)

With a content between 1000 and 5000 ppm for light REE and around 200 ppm for Eu and Gd, REE are in significant amount in spent fuel, at a higher grade than in most of the ores. However this inventory is very low compared to the world annual production. Only Sm could present an interest but, as mentioned above, its residual radioactivity is very high (above1TBq/g) and forbids any industrial use. However, the total world reserve in spent nuclear fuel represents several tens of years and a global approach could present an interest.

Table 4
Potential production of key critical elements coming from the processing of either French or worldwide spent nuclear fuel compared to the current world production, reserve and lifespan (reference year 2014).

Materials	2014 prod (t) (United States Geological Survey)	g/t in spent fuel	Potential French Annual prod (kg/year)	% world production	potential world production (kg/year)[a]	% world production	Potential world reserve (tons)[b]	production lifespan based on 2014 reference (years)
Cerium	39,850	3373	3879	0.010%	26,051	0.065%	710,462	18
Praseodymium	6075	1584	1821	0.030%	12,230	0.201%	333,546	55
Neodymium	18,925	5757	6621	0.035%	44,463	0.235%	1,212,661	64
Samarium	730	1156	1329	**0.182%**	8929	1.223%	246,547	338
Europium	330	190	218	0.066%	1467	0.444%	36,995	112
Gadolinium	1360	212	244	0.018%	1639	0.121%	48,094	35
Ruthenium	17	3222	3705	**21.796%**	24,885	146.381%	678,639	3,9920
Rhodium	21	641	737	**3.511%**	4952	23.581%	135,053	6431
Palladium	203	2125	2444	**1.204%**	16,412	8.085%	447,638	2205

Bold signifies % of the world production over 0.1%.
[a] Potential world production if all the discharged spent fuel would be reprocessed annually.
[b] Based on an average burn-up of 33GWd/t and a cooling time of 30 years.

3.2. Platinum group metals

Only about 2.4 tons of palladium, 3.7 tons of ruthenium and 0.7 ton of rhodium could be recovered annually from the French spent fuel, with a content of 2,100, 3200 and 640 ppm respectively. However, this represents a significant amount compared to the world production, particularly for ruthenium (22%). The treatment of the spent fuel discharged annually in the world could fulfil all the needs in ruthenium, one fourth of the needs in rhodium and about 10% of the needs in palladium. The amount available in the stored spent fuel at the world level represents a real mine for these PGMs.

4. Assessment of the possibility of declassing recycled SNF critical elements for subsequent use in the industry

Numerous isotopes of each element are produced during fission reactions. For instance, up to 53 isotopes have been identified for Indium. Most of these isotopes are radioactive, with half-life ranging from tenth of second to several millions of years. Very often, they are in negligible amount in mass, but they are responsible for most of the radioactive of a given element (Table 5).

Results for the selected elements are given in Table 6 and Fig. 2. We can notice that only Ce, Pr, Nd and Gd for REE and Ru and Rh for PGM reach an activity equal to or lower than 100Bq/g.

Table 5
Isotope abundances of the elements and associated contribution to the activity. Only isotopes representing more than 0.1% of the mass or 1% of the activity are mentioned.

	Weight %	Activity[a] %		Weight %	Activity[a] %		Weight %	Activity[a] %
Cerium			**Europium**			**Gadolinium**		
Ce140	51.738%		Eu151	0.6%		Gd153	1.8E-8%	96%
Ce142	48.260%		Eu152	4E-3%		Gd154	12.4%	
Ce144	2E-3%	100%	Eu153	89.2%		Gd155	4.6%	
Praseodymium			Eu154	8.8%	77%	Gd156	66.7%	
Pr141	100.0%		Eu155	1.4%	23%	Gd157	0.1%	
Pr144F	1.4E-7%	99%	**Neodymium**			Gd158	14.7%	
Pr144M	7.9E-10%	1%	Nd142	0.6%Gd159		Gd159	1.4E-12%	2%
Samarium			Nd143	18.6%		Gd160	1.4%	
			Nd144	33.5%		**Dysprosium**		
			Nd145	16.3%				
Sm147	28.7%		Nd146	17.6%		Dy160	12.3%	
Sm148	18.5%		Nd147	1.7E-11%	36%	Dy161	28.4%	
Sm149	0.3%		Nd148	9.2%		Dy162	29.7%	
Sm150	35.2%		Nd149	1e-13%	33%	Dy163	22.7%	
Sm151	1.2%	100%	Nd150	4.3%		Dy164	6.8%	
Sm152	11.7%		Nd151	5.3E-15%	14%	Dy165	3.9E-13%	51%
Sm154	4.5%		Nd152	3.3E-15%	9%	Dy165M	9.3E-17%	1%
Nd155	26.8%		Nd153	9.6E-17%	6%	Dy166	7.3E-12%	27%
Palladium			Nd154	3.0E-17%	2%	Dy167	4.5E-15%	13%
						Dy168	2.7E-15%	6%
Pd104	17.3%		**Ruthenium**			Dy169	5.7E-17%	2%
Pd105	28.3%					**Rhodium**		
Pd106	25.0%		Ru100	4.8%				
Pd107	15.4%	100%	Ru101	35.0%		Rh103	100.0%	
Pd108	10.3%		Ru102	35.7%		Rh106	3.1E-8%	100%
Pd110	3.6%		Ru104	24.4%				
			Ru106	6.7E-3%	100%			

[a] Calculations made using the WISE online calculation tool: http://www.wise-uranium.org/rccb.html(Wise Uranium Project).

Table 6
Activities[a] of selected elements after interim storage period following their separation from the spent fuel, in Bq/g of metal.

Interim storage period	Ru	Rh	Pd	Ce	Pr	Nd	Sm	Eu	Gd
0 y.	9.2E+09	4.6E+10	3.0E+06	2.0E+09	4.4E+09	1.9E+03	1.1E+10	1.2E+12	1.8E+04
10 y.	1.9E+07	1.6E+06	2.0E+06	5.6E+05	1.0E−03	5.5E−01	1.0E+10	4.7E+11	4.8E−01
20 y.	2.0E+04	1.4E+05	2.0E+06	7.6E+01	7.2E−05	3.9E−01	9.4E+09	2.0E+11	1.4E−05
30 y.	2.0E+01	1.3E+04	2.0E+06	1.0E−02	5.1E−06	2.8E−03	8.7E+09	8.7E+10	3.4E−10
40 y.	2.1E−02	1.2E+03	2.0E+06	1.4E−06	3.7E−08	2.5E−05	8.0E+09	3.9E+10	1.1E−14
50 y.	2.2E−05	1.1E+02	2.0E+06	1.9E−10	2.6E−08	6.0E−05	7.5E+09	1.8E+10	3.2E−19

[a] Calculations made using the WISE online calculation tool: http://www.wise-uranium.org/rccb.html (Wise Uranium Project).

When decaying, these radioactive isotopes produce new elements which will have to be separated from the target element after the decay period of time. Thankfully, less than 0.01% of Ru is radioactive (the decay product being Pd) and less than 1E-7% of Rh (giving also Pd).

Fig. 2 shows the activity decay as a function of time for the REE and PGM of interests, compared to the thresholds proposed by IAEA.

When considering a threshold at 100Bq/g, Nd, Pr and Gd could be recycled in non-nuclear industry after about 10 years of interim storage, Ce after 20 years, Ru after 30 years and Rh after 50 years.

Eu, Sm and Pd keep a too high activity to be re-used, whatever the interim storage time.

5. Assessment of the possibilities of designing dedicated and efficient separation processes

Thanks to studies carried out at CEA over the last decades on the recycling of minor actinides, several processes, mature enough for industrialization, have been developed. They allow a recovery of the REE from spent fuel dissolution liquors, after a PUREX process as implemented at AREVA La Hague Plant. The DIAMEX-SANEX processes, developed at CEA in the frame of the French Acts of 1991 and 2006 on nuclear waste management, allow the separation and the recovery of REE from PUREX raffinate.

Nevertheless, if these REE need to be separated one from each other, additional step would be required, based on existing processes, such the ones implemented today at the Solvay's Plant at La Rochelle (France), for example.

The recovery of the PGM is more complex because of their specific physic-chemical form in the spent fuel. Actually, they are embedded in metallic precipitates very difficult to dissolve with the current separation processes. Specific processes have to be considered if a quantitative recovery is targeted. Several options exist:

- For elements embedded in the precipitates, pyrochemical processes could be applied. However, their performances would not allow a sufficient decontamination factor. Additional developments are required.
- For PGM in solution, industrial processes already exist but should be adapted to the specific media. Selective extractants such as organophosphines (Cyanex process), quaternary ammonium salts or resins could be used (Hazelton et al., 1986).
- For ruthenium, the existence of the volatile Ru(VII) species could be the basis for an efficient separation process based on the selective oxidation/volatilization of Ru followed by a condensation at lower temperature. Available experimental suggest that very high decontamination factors would be achievable (Mousset, 2004).

According to the very high decontamination factors (DF) required (DF > 10^8), hydrometallurgical processes seem to be the only type of processes that would be suitable for the recovery of

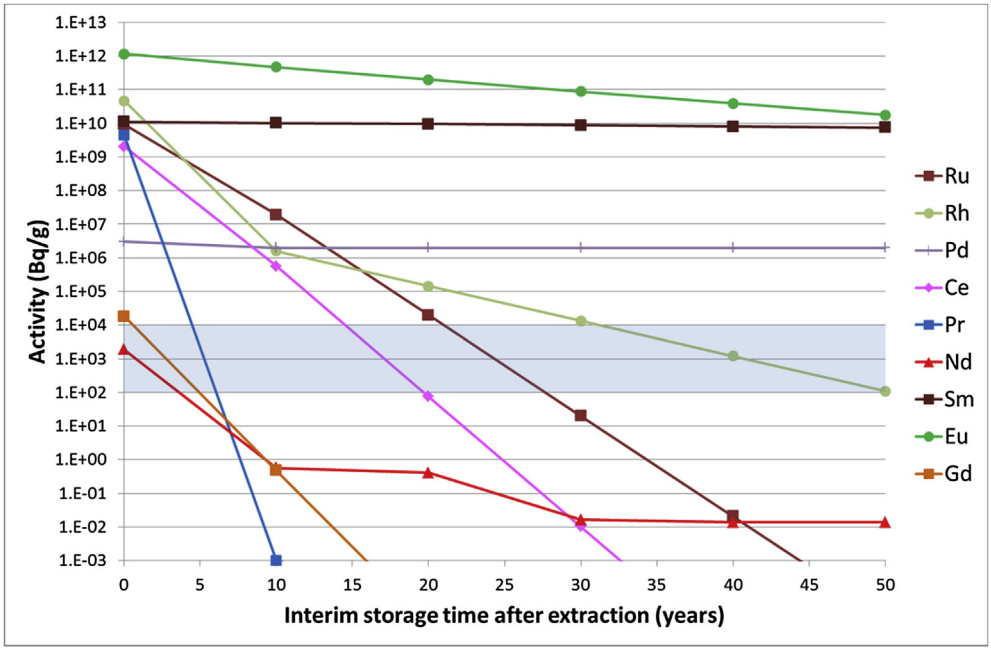

Fig. 2. Time dependence of specific activity of critical elements after their recovery from the spent fuel.

these elements.

6. Conclusions

Critical raw materials, although used at trace levels, are of paramount importance for the development and production of new technologies and therefore for sustaining the economy and wealth creation. However, their world stockpile is limited and their production is often dominated by a very limited number of countries, hence creating a strong dependence for non-producing countries such as the European countries for instance. An intensive effort has therefore been initiated either to find new potential sources of production (new mines, byproducts, waste recycling) or to substitute them by non-critical elements. In this context, production of electricity in nuclear reactors yields to the continuous production within the spent nuclear fuel of significant amount of newly formed fission products, some of them being part of the critical raw materials. Among them, six were considered of potential interest for other industrial applications and were more carefully studied in this paper by considering (i) their inventory compared to the global need, (ii) their residual radioactivity: cerium, neodymium, praseodymium, gadolinium, ruthenium and rhodium. The assessment developed in this paper demonstrates that:

- The inventory of the REE that would be recovered from spent nuclear fuel is too low compared to the annual world production to be economically viable. The recovery price from the spent nuclear fuel would be prohibitive compared to the market price (25–150$/Kg) which is limited.
- Annual rhodium production from French reprocessed spent fuel could cover 3.5% of the 2014 world production. Moreover, the market price is quite high (40–300$/g). However, a 50 year interim storage would be needed before any use. A complex and expensive separation process is to be developed and there is not enough data to make a relevant estimation at the time.
- Ruthenium is the most interesting element. It could be possible to recover about 3.7 tons annually in France. This would represent about 20% of the world production. Its market price is very fluctuating but remains high (3–30$/g). It could be re-used after 10 years of spent fuel cooling before recovery plus 25 years of interim storage (35 years in total). As for Rh, specific separation processes must be developed, complex and expensive to implement in a nuclear environment.

The previous economics balance would still be much more interesting if it would be possible to treat a large part of the spent nuclear fuel discharged annually in the world (up to a factor of 10 by comparison to the French potential production). In addition, the global inventory of spent nuclear fuel stored since the beginning of the industrial use of the nuclear energy represents a real mine for these elements, stockpiling up to several tens to several thousands of some critical raw material annual production (on the 2014 production basis). However, recovering these elements would only be worth to be implemented if uranium and plutonium are also recycled for nuclear electricity production in order to share the treatment cost. It is hence mainly relevant for countries which already recycle or plan to recycle nuclear materials, such as France, UK, Japan, and in the future, China.

Finally, apart from the technical motivations and criteria, among which the residual radioactivity and dose rate are key issues, recycling materials from spent nuclear fuel for non-nuclear applications is also clearly a strong political and societal question which relates to the approach chosen for preventing any risk of contamination of the end-users. Either supported by technical regulations or laws, this choice hence includes a part of irrational which drives the way the society globally accepts and manages the residual technological risk, from very cautious approaches based on the precaution principle to more pragmatic approaches based on the actual risk assessment.

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