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REPROCESSING TECHNIQUES OF LWR SPENT FUEL FOR REUTILIZATION IN HYBRID SYSTEMS AND IV GENERATION REACTORS

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ABSTRACT

Since the era of nuclear technology begins, nuclear reactors have been produced spent fuel. This spent fuel contains material that could be recycle and reprocessed by different processes. All these processes aim to reduce the contribution to the final repository through the re-utilization of the nuclear material. Therefore, some new reprocessing options with non-proliferation characteristics have been proposed and the goal is to compare the different techniques used to maximize the effectiveness of the spent fuel utilization and to reduce the volume and long-term radiotoxicity of high-level waste by irradiation with neutron with high energy such as the ones created in hybrid reactors. In order to compare different recovery methods, the cross sections of fuels are calculated with de MCNP code, the first set consists of thorium-232 spiked with the reprocessed material and the second set in depleted uranium that containing 4.5% of U-235 spiked with the reprocessed material; These sets in turn are compared with the cross section of the UO_2 in order to evaluate the efficiency of the reprocessed fuel as nuclear fuel.

1. INTRODUCTION

The large-scale development of the nuclear industry and the proliferation of nuclear weapons have created a state of worldwide concern about the consequences of the introduction of radioactive nuclides into the atmosphere, hydrosphere and lithosphere. The radioactivity of the transuranic nuclides and the relatively long half-lives of many of these radionuclides provide ample reasons for this concern; therefore, establishing radiochemical techniques for the treatment and recovery of nuclear fuel, with simplification of processes and minimization in the evolution of radioactive waste, will contribute to preserve our environment.

Over the years, many reprocessing methods have been developed for the recovery of specific actinides (plutonium, uranium and some minor actinides), such as PUREX, UREX, TRUEX, DIAMEX, etc. In 2005, more than 75,000 MTHM spent nuclear fuel had been processed worldwide [1]. In general, technologies for separating spent nuclear fuel are divided into two processes, either aqueous or non-aqueous, as shown in Table 1.

Table 1: Some processing methods

Aqueous methods	Non-aqueous methods
PUREX	LiCl-KCl
UREX	NaCl-KCl
TRUEX	Flouride Volatility
DIAMEX	FLUOREX
SANEX	PYRO-A
UNEX+	PYRO-B

Due to the large number of recovery methods [2] , it is necessary to determine a suitable path for nuclear fuel reprocessing. However, each proposed method presents a different set of attributes with respect to: complexity, safety, residues and proliferation risks [3].

The methodology presented consists of calculating the total cross section of two sets of fuels, the purpose of this calculation is to compare the cross section between the different recovery methods and with the fuel UO_2, this comparison will allow to see the efficiency of the reprocessed fuel as a nuclear fuel, and will allow to see the differences that exist between the cross section with the presence of transuranic element. As mentioned, we study two sets of fuels, the first set consists of thorium spiked with the reprocessed fuel from the different processes described in section 2 and the second set consists of a depleted uranium (0.2% of 235-U) spiked with the nuclear fuel reprocessed under the different processes provided in section 2.

To obtain the cross sections they were processed using NJOY99.364 and library was ENDF/B-VII.1, the methodology consist of generated the cross section of each nuclide and use them to plot the material cross section considering the weighted mean of each nuclide. The plot generated using the Vised from the MCNP.

2. REPROCESSING METHODS

Over the past 70 years, various methods of reprocessing used nuclear fuel and other radioactive materials have been proposed [3] . For the purposes of this study, only those that are applicable to the oxide fuels are discussed. The processes considered in this work are explained below.

2.1 PUREX process

PUREX is an acronym from Plutonium and Uranium Recovery by Extraction. The PUREX process is an extraction of the liquid-liquid method used to reprocess spent nuclear fuel, in order to obtain uranium and plutonium, independently, from the fission products.
The process is based on the selective affinity of tributyl phosphate (TBP) for uranium and plutonium. There are three basic functions of the process:

1. Extraction, where uranium and plutonium are separated from the fission products.
2. Separation, where plutonium is extracted from uranium;
3. Stripping, plutonium undergoes further decontamination, with an average decontamination factor greater than 10^6 [4], [5].

After the fuel is chopped and declad, the fuel is dissolved in nitric acid to create a feed solution. This solution is introduced into a solvent extraction process with TBP diluted in n-dodecane ($C_{12}H_{26}$) [6] . Using this reprocessing method, the recovery percentages according [4] are 99.9% for uranium, plutonium recovery is 99.8% and neptunium recovery is 95%.

2.2 GANEX process

The GANEX process (the grouped actinide extraction), developed by the CEA for the reprocessing of nuclear fuels and the homogeneous recycling of actinides, is composed of two extraction cycles after the dissolution of the spent fuel in the nitric acid. The first cycle consists where the uranium bulk is removed from the fuel dissolution liquor and the second cycle where the transuranic elements as well as the residual uranium are extracted together as a group [7]. In this work the results of two types of reprocessing based on the GANEX method will be used; the first is to use the second cycle of the GANEX method [7], the second is to adapt the DIAMEX-SANEX process to manage the separation of neptunium and plutonium along with americium and curium in the second cycle of the GANEX process [10]. For the first case, the recovery percentages are 93.5% of U, 99.8% of Np, 99.6% of Pu, 99.8% of Am and 99.8% of Cm. For the second case, the recovery percentages for U, Np, Pu, Am, Cm are 0.1%, 95%, 99.5%,99.5%, 99.5 % respectively, and 5% for Nd, Sm and Eu.

2.3 TRUEX and SREX process

The TRUEX and SREX processes use centrifugal contactors to mix radioactive tank waste with a solvent containing an extractant. In the TRUEX process, an extractant [octyl(phenyl)-N, Ndiisobutylcarbamoylmethylphosphine oxide (CMPO)] is dissolved in organic solvent. The transuranic elements are extracted into the organic phase, and the other waste components remain in the aqueous phase. The transuranic elements are subsequently stripped from the organic phase, which can then be recycled and reused in the process. Similarly, the SREX process uses a crown ether [4',4'(5')-di-(tertbutyldicyclohexo)-18-crown-6 (DtBuCH18C6)] in an organic solvent to remove strontium from a tank waste solution [8]. According to [13] the recovery percentages for the TRUEX process of U, Pu, Am are 99.9%, 99.97%, 99.8% respectively, and the recovery percentages for the SREX process of U, Pu, Am are 99.6%, 99.97%, 0.2% respectively.

2.4 UREX+ process

The UREX + (URanium EXtration) process basically involves five steps of solvent extraction [9]. In the first step, the Tc and U are recovered together and, in a subsequent step, are separated. The Cs and Sr are removed from the aqueous stream in a second step. During the third step the Pu and Np are separated from the previous phase and recycled to produce the MOX together with the uranium from the first phase.

As in the co-processing process, UREX + provides the possibility of separation of the actinides. In this technique, the Am, Cm, Ru, Zr, Nd and Ce are extracted in the final stage of the process. According to [10] the recovery percentages of U, Np, Pu, Am and Cm are 99.95%, 71%, 99.5%, 98% and 79% respectively.

2.5 DIAMEX process

The DIAMEX process (Diamide Extraction) is a French innovation using diamides as extraction agents. This process has the advantage of avoiding the formation of organic waste containing different elements of carbon, hydrogen, nitrogen and oxygen. Also of greater affinity of extraction towards the minor actinides and lanthanides. Also less tendency to form a third phase and a design that gives shorter degradation products, which simplifies the organic phase cleaning [11]. According to [11] the recovery percentages of U, Np, Pu, Am and Cm are 35%, 49.4%, 26.4%, 99.7% and 99.94% respectively.

2.6 Phosphine Oxide Process (POR)

Alkylated phosphine oxides (POR) are typically excellent extractants for TRUs in the +3, +4, or +6 valence states from acidic waste solutions. Attachment o different alkyl or aryl substituents to the phosphonate grou results in different compounds with variations in selectivity, extraction efficiency, solubility, and radiolytic and hydrolytic stability. A phosphine oxide solvent extraction process consists of an alkylated phosphine oxide in a hydrocarbon diluent [12].

Laboratory batch contact, countercurrent pilot-scale testing with simulated and actual waste solutions has been performed. POR flow sheet has been demonstrated using actual waste solution in a centrifugal contactor pilot plant. According to [13] the recovery percentages of U, Np, Pu, Am and Cm are 35%, 49.4%, 26.4%, 99.7% and 99.94% respectively.

3. METHODOLOGY

In order to evaluate the different recovery methods provided in section 2, this work calculates the cross sections with the MCNP code for two sets of fuels.

The first set consists of thorium spiked with the reprocessed fuel from the different processes described in section 2 and the second set consists of a depleted uranium (0.2% of 235-U) spiked with the material reprocessed under the different processes provided in section 2. The reprocessed fuels were obtained from the spent fuel discharged from the PWR reactor [14]. These reprocessed fuels must have 4.5% fissile isotopes (233-U, 235-U, Pu 239 and 241-Pu) for the purpose of this work. In order to obtain that fissile percentage, the reprocessed fuel will be spiked with thorium and the other will be spiked with depleted uranium.

After obtaining the composition of the two reprocessed fuel assemblies, the total cross section is calculated for the two sets and then the total cross section of the UO_2 is calculated in order to compare the cross section of the UO_2 with the fuels reprocessed.

3.1 Obtaining the new fuel mixture

It will be used the discharge fuel composition provided in Table 2 for a representative PWR assembly of 4.5-wt% 235U initial enrichment and 50-GWd/MTU burn-up [14] .

Table 2: Discharge fuel composition (4.5 initial wt% U-235, 50-GWd/MTU) for calculating time-dependent spent fuel composition [14].

Isotope	Atom density (atom/barn.cm)	Isotope	Atom density (atom/barn.cm)	Isotope	Atom density (atom/barn.cm)
C-14	1.85E-09	Cs-133	6.99E-05	Pa-231	1.87E-11
Cl[c]-36	1.00E-06	Cs-135	3.46E-05	U-232	2.31E-11
Ca[c]-41	1.00E-06	Cs-137	7.51E-05	U-233	8.71E-11
Ni[c]-59	1.00E-06	Nd-143	4.66E-05	U-234	4.57E-06
Se-79	5.06E-07	Nd-145	3.80E-05	U-235	2.50E-04
Zr-93	6.36E-05	Sm-147	4.71E-06	U-236	1.50E-04
Sr-90	4.86E-05	Sm-149	1.38E-07	U-238	2.19E-02
Nb-93	6.63E-11	Sm-150	1.63E-05	Np-237	1.99E-05
Nb-94	6.21E-11	Sm-151	9.71E-07	Pu-238	9.35E-06
Mo-93	1.15E-14	Sm-152	6.32E-06	Pu-239	1.83E-04
Mo-95	6.08E-05	Eu-151	1.56E-09	Pu-240	7.29E-05
Tc-99	6.67E-05	Eu-153	6.62E-06	Pu-241	4.80E-05
Ru-101	6.56E-05	Gd-155	5.46E-09	Pu-242	1.90E-05
Rh-103	3.47E-05	Pb-210	3.89E-18	Am-241	2.23E-06
Pd-107	1.85E-05	Ra-226	1.44E-15	Am-242M	5.26E-08
Ag-109	6.07E-06	Ra-228	9.40E-22	Am-243	5.61E-06
Sn-126	1.29E-06	Ac-227	3.26E-16	Cm-245	1.40E-07
Sb-126	2.38E-10	Th-229	5.97E-14	Cm-246	1.39E-08
Sb-126	3.18E-13	Th-230	5.51E-11	-	-
I-129	1.03E-05	Th-232	1.27E-11	-	-

The values presented in Table 2 are affected by the recovery factors of the different recovery processes, all values of recovery factors described in section two are summarized in Table 3.

Table 3: Recovery factors for the processes described in section 2.

	GANEX [10]	**GANEX** [7]	**UREX+**	**TRUEX**	**SREX**	**POR**	**DIAMEX**	**PUREX**
U	0.001	0.935	0.9995	0.999	0.996	-	0.035	0.999
Np	0.95	0.998	0.71	-	-	-	0.494	0.95
Pu	0.995	0.996	0.995	0.9997	0.9995	0.9997	0.264	0.998
Am	0.995	0.998	0.98	0.998	0.002	0.564	0.997	-
Cm	0.995	0.998	0.79	-	-	-	0.9994	-
Nd	0.05	-	-	-	-	-	-	-
Sm	0.05	-	-	-	-	-	-	-
Eu	0.05	-	-	-	-	-	-	-

The percentage composition of the reprocessed fuel is presented in Table 4, as well as the percentage of fissile material present, for all the processes considered in this work.

Table 4: Percentage composition of the material processed for the different recovery processes and the percentage of fissile isotopes.

	GANEX	**GANEX**	**UREX+**	**TRUEX**	**SREX**	**POR**	**DIAMEX**	**PUREX**
Np-237	5.170	5.467	3.957	0.000	0.000	0.000	9.196	5.328
Pu-238	2.557	2.576	2.618	2.726	2.791	2.754	2.320	2.643
Pu-239	50.365	50.744	51.580	53.704	54.992	54.260	45.712	52.064
Pu-240	20.089	20.240	20.573	21.421	21.934	21.642	18.233	20.767
Pu-241	13.288	13.388	13.608	14.169	14.508	14.315	12.060	13.736
Pu-242	5.284	5.323	5.411	5.634	5.769	5.692	4.795	5.462
Am-241	0.618	0.624	0.623	0.659	0.001	0.375	2.117	-
Am-242	0.015	0.015	0.015	0.016	0.000	0.009	0.050	-
Am-243	1.566	1.581	1.579	1.671	0.003	0.952	5.367	-
Cm-245	0.039	0.040	0.032	0.000	0.000	0.000	0.135	-
Cm-246	0.004	0.004	0.003	0.000	0.000	0.000	0.013	-
Nd-143	0.384	-	-	-	-	-	-	-
Nd-145	0.318	-	-	-	-	-	-	-
Sm-147	0.040	-	-	-	-	-	-	-
Sm-149	0.001	-	-	-	-	-	-	-
Sm-150	0.141	-	-	-	-	-	-	-
Sm-151	0.008	-	-	-	-	-	-	-
Sm-152	0.055	-	-	-	-	-	-	-
Eu-151	0.000	-	-	-	-	-	-	-
Eu-153	0.058	-	-	-	-	-	-	-
Fissile material	63.653	64.132	65.188	67.873	69.5	68.575	57.772	65.800

It is important to note that although many of the recovery processes manage to extract uranium from the burned fuel, in this work, the reprocessed uranium is not considered in the reprocessed fuel, since the percentage of U-238 is much higher than the percentage of fissile material that wants to obtain (4.5%).

In order to obtain the new fuel mixture ($(Th{-}TRU)O_2$ and $(U-TRU)O_2$), the reprocessed material (Table 4), for each of the recovery processes, is spiked with Thorium 232 in the first case, and, for the second case, the reprocessed material is spiked with depleted uranium (0.2% of 235-U).

As can be seen in Table 4, the amount of fissile material in almost all processes exceeds 60%, and the new fuel to be obtained must contain 4.5% fissile material. To normalize the values of the new mixture to a percentage of fissile material of 4.5% and 95.5% of non-fissile materials, the following equation is used.

$$NF_R = \frac{f - 4.5\%}{f - f_R} \quad ; \quad NF = \frac{4.5\% - f_R}{f - f_R} \tag{1}$$

Where:

- NF_R is the normalization factor for the reprocessed material.
- NF is the normalization factor for the material with which the reprocessed material is spiked (Th-232 or depleted uranium).
- f is the percentage of fissile material present in the material with which the reprocessed material is spiked (Th-232 or depleted uranium).
- f_R is the percentage of fissile material present in the reprocessed material.

The equation (1) is used to normalize the values of the composition of new fuel, for the two sets of fuels. In the case of reprocessed fuel spiked with thorium $f = 0$ because only Th-232 is used, and in the case of the reprocessed fuel spiked with depleted uranium $f = 0.2\%$.

3.2 Procedures for calculating the cross section

Considering that the actinides were processed at 1200 K, to calculate the material cross section it should be know the mass fraction contribution of each nuclide to the material. Therefore, the cross section of each nuclide is weighted mean to produce following the percentage contribution to the material. The fuel cross section was plotted using the Vised from the MCNP. To sum up the material cross section have a contribution of each nuclide with their respectively percentage.

Then to represent the differences between the fuels: UO2, the reprocessed fuel spiked with depleted uranium and the one spiked with thorium. They were plotted together to appreciate the differences between them with fissile material about 4.5%. The differences between each fuel cross section must be due to the different amount of minor actinides in the fuel or the absence of them [15].

4. RESULTS

4.1 Fuel processed with thorium and depleted uranium

The reprocessed material with thorium obtained by the processes described in section 2 are shown in Table 5. Obviously, the mass fractions of each isotope are different because the recovery factor of each process is different. All the fuels presented in Table 5 are composed of 4,5 % fissile material.

Table 5: Fuel composition (perceptual) for the reprocessed fuels spiked with thorium

	GANEX	GANEX	UREX+	TRUEX	SREX	POR	DIAMEX	PUREX
Th-232	81.689	81.740	81.839	82.079	82.214	82.138	81.064	81.895
Np-237	0.321	0.337	0.240	0.000	0.000	0.000	0.630	0.320
Pu-238	0.159	0.159	0.159	0.159	0.159	0.159	0.159	0.159
Pu-239	3.130	3.130	3.130	3.130	3.130	3.130	3.130	3.130
Pu-240	1.248	1.248	1.248	1.248	1.248	1.248	1.249	1.248
Pu-241	0.826	0.826	0.826	0.826	0.826	0.826	0.826	0.826
Pu-242	0.328	0.328	0.328	0.328	0.328	0.328	0.328	0.328
Am-241	0.038	0.038	0.038	0.038	-	0.022	0.145	-
Am-242	0.001	0.001	0.001	0.001	-	0.001	0.003	-
Am-243	0.097	0.098	0.096	0.097	-	0.055	0.368	-
Cm-245	0.002	0.002	0.002	0.000	-	-	0.009	-

Cm-246	0.000	-	-	-	-	-	0.001	-
Nd-143	0.024	-	-	-	-	-	-	-
Nd-145	0.020	-	-	-	-	-	-	-
Sm-147	0.002	-	-	-	-	-	-	-
Sm-149	0.000	-	-	-	-	-	-	-
Sm-150	0.009	-	-	-	-	-	-	-
Sm-151	0.001	-	-	-	-	-	-	-
Sm-152	0.003	-	-	-	-	-	-	-
Eu-151	0.000	-	-	-	-	-	-	-
Eu-153	0.004	-	-	-	-	-	-	-
O-16	12.096	12.092	12.092	12.093	12.094	12.094	12.088	12.093

In the case of reprocessed material with depleted uranium, the recovery methods presented in section 2 were also used. The mass fractions obtained are presented in Table 6. The fraction of fissile material in these fuels is 4,5%.

Table 5: Fuel composition (perceptual) for the reprocessed fuels spiked with depleted uranium.

	GANEX	**GANEX**	**UREX+**	**TRUEX**	**SREX**	**POR**	**DIAMEX**	**PUREX**
U-235	0.164	0.164	0.165	0.165	0.165	0.165	0.163	0.165
U-236	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
U-238	82.015	82.064	82.160	82.392	82.522	82.449	81.411	82.214
Np-237	0.309	0.324	0.231	0.000	0.000	0.000	0.606	0.308
Pu-238	0.153	0.153	0.153	0.153	0.153	0.153	0.153	0.153
Pu-239	3.009	3.009	3.009	3.008	3.008	3.008	3.010	3.009
Pu-240	1.200	1.200	1.200	1.200	1.200	1.200	1.201	1.200
Pu-241	0.794	0.794	0.794	0.794	0.794	0.794	0.794	0.794
Pu-242	0.316	0.316	0.316	0.316	0.316	0.316	0.316	0.316
Am-241	0.037	0.037	0.036	0.037	-	0.021	0.139	-
Am-242	0.001	0.001	0.001	0.001	-	0.000	0.003	-
Am-243	0.094	0.094	0.092	0.093	-	0.053	0.353	-
Cm-245	0.002	0.002	0.002	0.000	-	0.000	0.009	-
Cm-246	0.000	-	-	-	-	-	0.001	-
Nd-143	0.023	-	-	-	-	-	-	-
Nd-145	0.019	-	-	-	-	-	-	-
Sm-147	0.002	-	-	-	-	-	-	-
Sm-149	0.000	-	-	-	-	-	-	-
Sm-150	0.008	-	-	-	-	-	-	-
Sm-151	0.001	-	-	-	-	-	-	-
Sm-152	0.003	-	-	-	-	-	-	-
Eu-151	0.000	-	-	-	-	-	-	-
Eu-153	0.003	-	-	-	-	-	-	-
O-16	11.846	11.842	11.842	11.842	11.842	11.842	11.841	11.842

4.1 Cross-section of fuels

Figures 1 to 7 show the total absorption cross sections for the different fuels obtained, which are thorium 232 spiked with the reprocessed fuel and depleted uranium spiked with the reprocessed fuel for each of the recovery processes. In addition, each figure includes the total cross section of UO_2 enriched with 4.5%, in order to compare the cross sections.

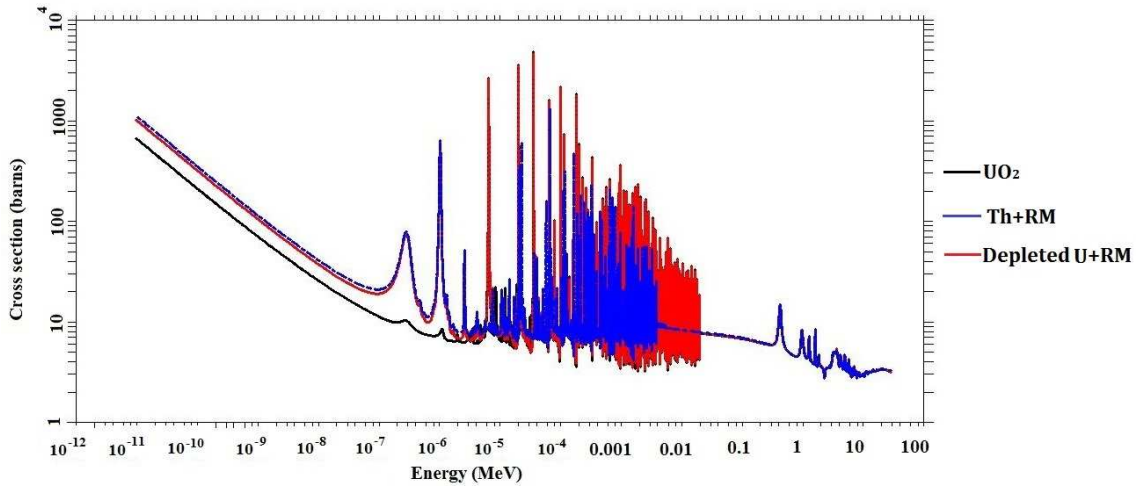

Figure 1: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the GANEX process.

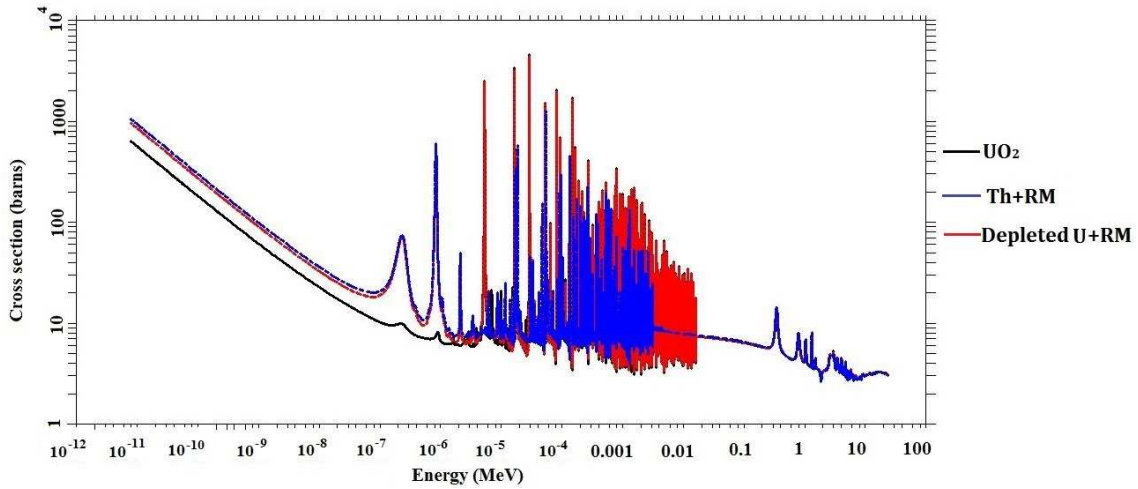

Figure 2: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the UREX+ process.

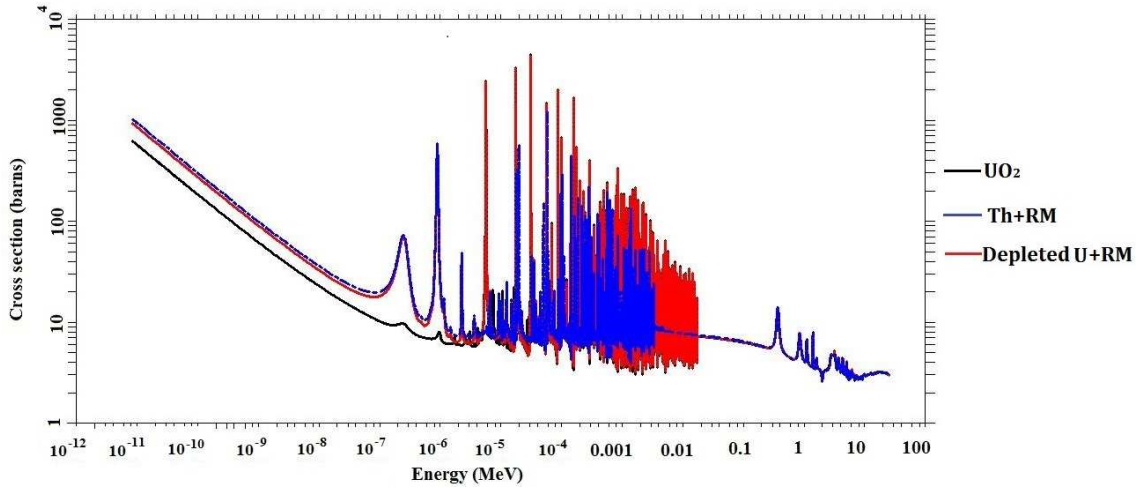

Figure 3: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the TRUEX process.

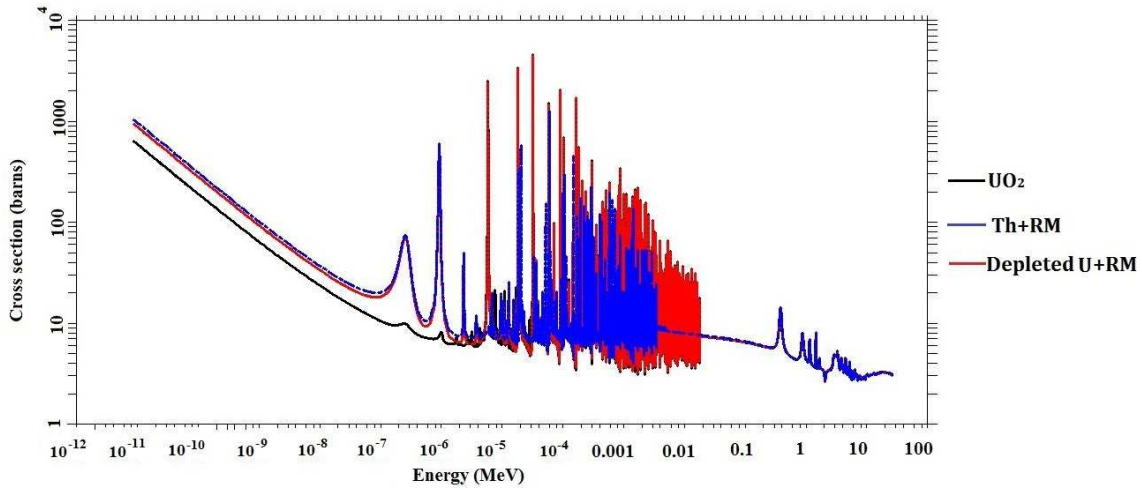

Figure 4: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the SREX process.

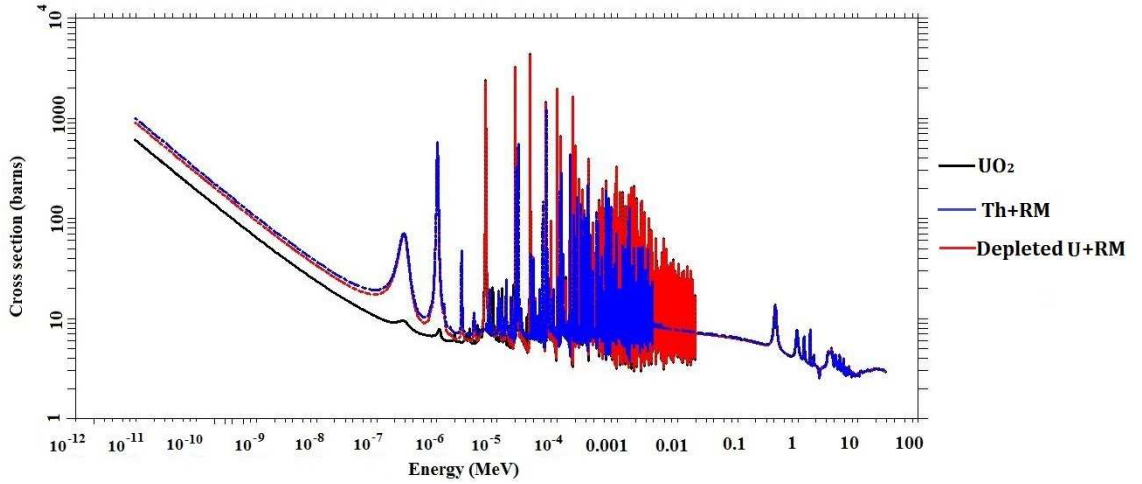

Figure 5: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the POR process.

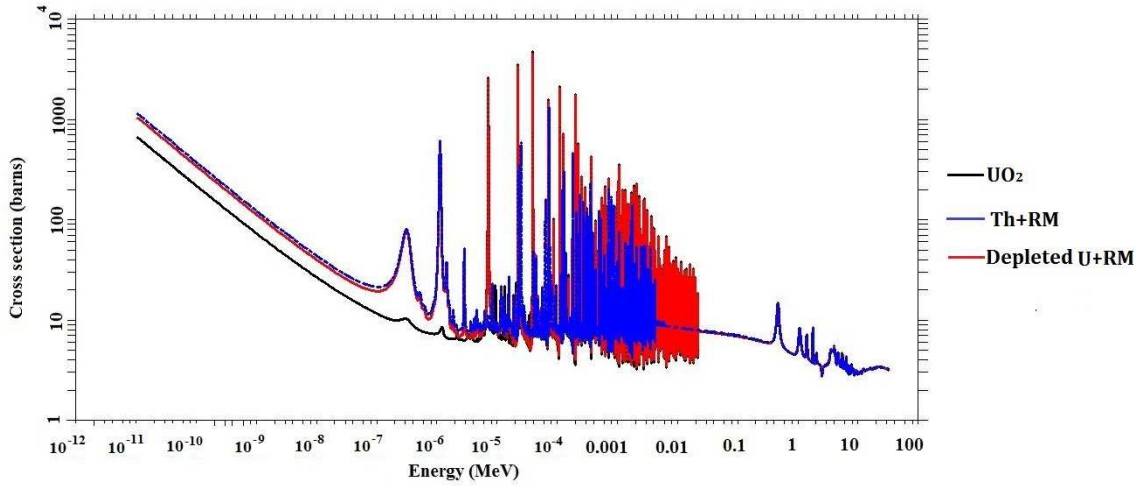

Figure 6: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the DIAMEX process.

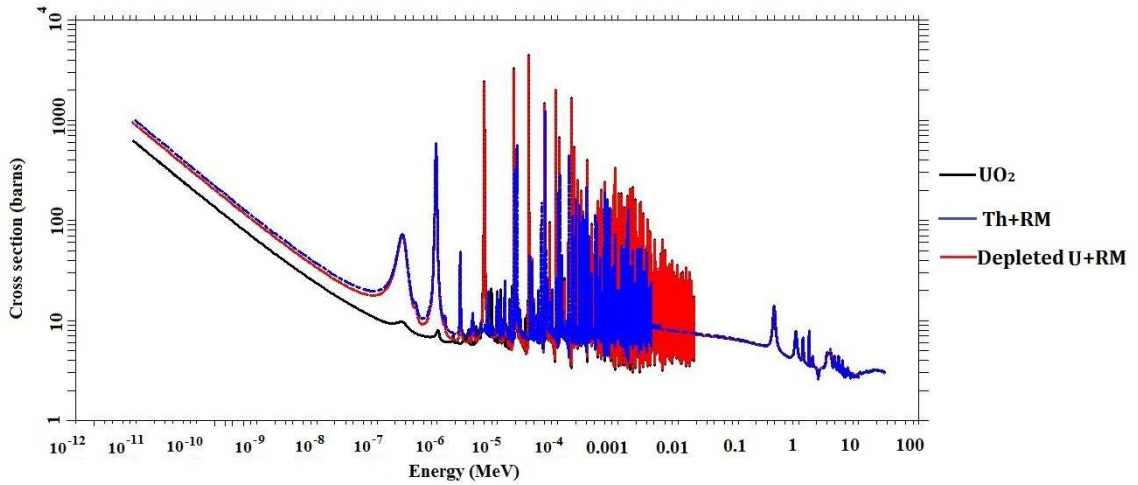

Figure 7: Total cross section of the UO_2, reprocessed material spiked with thorium and reprocessed material spiked with depleted uranium for the PUREX process.

It can be observed that there is no notable difference between the sections of the different recovery processes. However, a difference can be seen by comparing the cross sections of the reprocessed material spiked with thorium and the spiked with depleted uranium, that is approximately in the range 0.01 MeV.

5. CONCLUSION

The different recovery processes presented in this work are used for the reprocessing of the burned fuel of a PWR reactor. Two types of fuel were produced with the material recovered, thorium spiked with the reprocessed material and another consisting of reprocessed material spiked with depleted uranium. The cross sections of these fuels are very similar for the different reprocessing methods. Therefore, the insertion of these reprocessed fuels does not modify to a great extent the behavior of the cross sections. Then it can be concluded that the recovery processes do not modify the cross sections, so that each recovery process is efficient in relation to the actinides it can recover. For the particular case presented in this work, the processes that most actinides are recovered are GANEX, DIANEX and UREX+, therefore these processes are more efficient for the elimination of actinides. If the cross section of the UO_2 is compared to the reprocessed material spiked with depleted uranium, the total cross sections do not differ much as expected. However, if the cross section of UO_2 is compared with thorium fuel spiked with the reprocessed fuel, it can be seen that there is a remarkable difference in the range of energy 0.01 MeV.

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