TRANSURANICS ELIMINATION IN AN OPTIMISED PEBBLE-BED SUB-CRITICAL REACTOR

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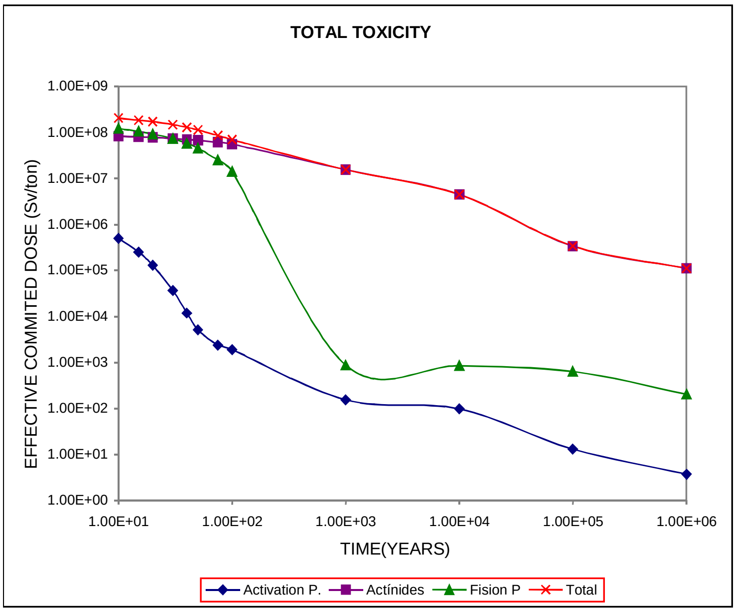
Abstract

In a nuclear energy economy the nuclear waste is a big burden to its further development and deployment. The possibility of eliminating the long-term part of the waste presents an appealing opportunity to the sustainability and acceptance of a better and cleaner source of energy. It is shown that the proposed pebble-bed transmutator has suitable characteristics to transmute most of the isotopes that contribute to the long-term radioactivity. This proposed reactor presents also inherent safety characteristics, which is a necessary element in a new reactor design to be accepted by the society. Throughout this paper, we will characterise the new reactor concept, and present some of the neutronics and safety characteristics of an accelerator driven pebble-bed reactor, (ADS) for transuranics elimination.

1. Introduction and background

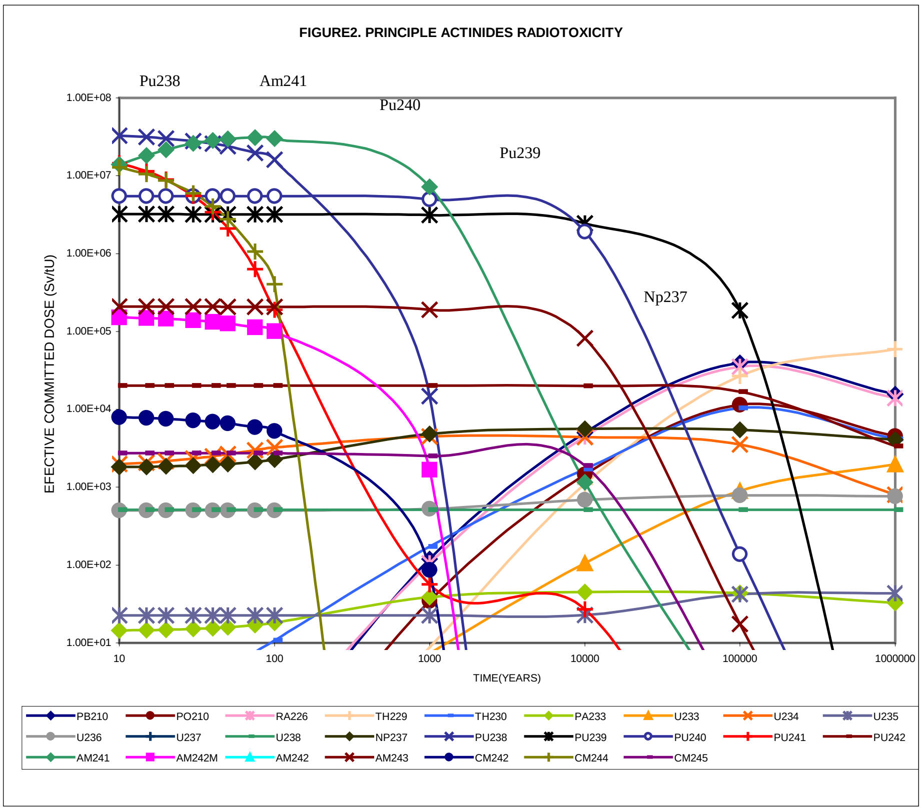
The sustainability of nuclear energy depends on the capability to decrease the long-term radiotoxicity of nuclear waste [1], as the present situation in which toxical waste has a lifetime of 10^5 years and more, is totally unacceptable. The main contributors to the radiotoxicity of the spent nuclear fuel of a light water reactor (LWR.), which is the most common design in the world nuclear park, are depicted in Figure 1.

Figure 1. **Total toxicity of the spent nuclear fuel from LWR**



As can be seen the activation products have the smallest contribution, and will decay in a period of less than 50 years. (At this time they reach a radiotoxicity similar to natural uranium). Fission products are the main short-term (less than 100 years) contributors, they reach the radiotoxicity of the uranium ore in a period of less than 400 years, which is an acceptable period of time that our technology can control. As we can see the actinides have different characteristics, it takes an important contributor in the short term, but the only contributors in the long term. It takes approximately one million years for the radiotoxicity of MA to reach the uranium ore level, so that the main radiotoxic heritage problem are these isotopes. To identify exactly which actinides are the most dangerous in the nuclear waste, Figure 2 presents the LWR waste isotope composition as a function of time after removal from the reactor.

Figure 2. **Main actinides radiotoxicity**



In this figure, we can see that the Pu isotopes (in particular ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu via their disintegration to ^{241}Am and this to ^{237}Np) are the main contributors. The disposal of the nuclear fuel without any treatment (open cycle) in a deep storage facility (DSF) has several problems: we must consider nuclear proliferation, the criticality during the isotopic evolution of the fuel, the heat released by decay of these isotopes and the uncertainties associated to the enormous period of time (for example, the geometry in the DSF might change) [2-6]. The elimination of the actinides, in particular the Pu isotopes and the minor actinides (specially the Am and Cm isotopes) will reduce the burden of DSF in a substantial way.

2. Pebble-bed transmutator

Recently a new concept of sub-critical nuclear reactors is being considered, namely the accelerator driven systems (ADS) [6-12]. The principal characteristics of this kind of reactors are:

Improved safety characteristics. In a sub-critical reactor, the shut-down of the external neutron source (something easily achievable), results in the instant shut-down of the reactor power. If we can guarantee the sub-criticality of the reactor under any situation, the risk of reactivity accidents, as happened in Chernobyl, are inherently null.

Flexibility in the reactor burn-up. Since the main goal of a transmutator is to get rid of all the fuel (mainly fissile) isotopes, the k_{eff} will decrease very rapidly during the burn-up. In a normal critical reactor, this is an insurmountable problem. In a sub-critical reactor, we have the additional freedom of the external neutron source. With a suitable recycling strategy, and a somewhat variable external neutron source, the reactor can maintain the neutron flux level and power during the life cycle of the fuel, and an almost constant k_{eff}.

Flexibility of the isotopic composition of the fuel. In the case of a critical reactor, the necessity to obtain a k_{eff} equal to one during the burn-up of the fuel is a constrain to the fuel composition. In the case of the proposed open cycle PB reactor, the isotopic composition is fixed by the spent fuel discharged from the nuclear park in each country. The advantage of the ADS is that the external neutron source can maintain the neutron flux at the desired level, so that the composition of the fuel is less restricted than in a conventional critical reactor.

The Pebble-bed transmutator [23] studied in this paper is an ADS, with an external spallation neutron source, created by interactions of high energy protons with a heavy metal, such as Pb. The fuel surrounding the target is contained inside of 3 cm of radius graphite spheres (the pebbles), having two regions [13-15]. The inner region contains carbide fuel kernels surrounded by a porous carbonaceous layer, called buffer. The buffer function is to absorb the gaseous fission products. It is coated with two layers of high density pyrolitic graphite and a layer of silicon carbide in between (TRISO), and the external part of the pebble is made of high purity graphite. The TRISO micro-spheres have a diameter of 0.9 mm [16-19].

In the neutronic calculations carried out in the present study, a homogeneous mixture of carbon and fuel atoms was assumed in the fuel region. We can do this [24] because the mean free path of the neutrons is larger than the size of the TRISO coated fuel particles, so for a neutron it is an homogeneous zone. The external radius of the pebble is 3 cm, and is a fixed parameter in these neutronic studies. An important characteristic of pebble-bed reactors is that it is possible to change the neutron spectra in the reactor by varying the radius of the inner region of the pebble assuming a constant mass of fuel. In the calculations below, 2 g per fuel sphere were assumed.

The isotopic composition of the pebbles is given in Table 1. This isotopic composition corresponds to the actinides discharged from a LWR after an average burn-up of 30 MWd/kg-Metal. Only isotopes with a higher contribution to the radiotoxicity of the spent fuel have been chosen. The reactor is cooled by CO_2, and the average temperature of the gas inside the core is 550 K, and the nominal outlet temperature is 800 K. This value is well below the disassociation threshold of CO_2. The main characteristics of the PBT prototype studied are described in Table 2.

The pebble-bed high temperature gas cooled reactors (HTGCR) has been designed already in the 60s and advanced concepts in the 70s. Some analysis on fast gas-cooled critical reactors [25] pointed out that decompression accidents (loss of coolant) would imply reactivity insertions leading to slightly supercritical states in a very short time (shorter than the estimated scram time, and without a significant Doppler effect in a core loaded with minor actinides). From the point of view of nuclear safety, such a type of critical transmutator would be impossible to license. However, in the case of an ADS, with an adequate margin of reactivity a transmutator of this type could be possible. The reactor will have continues refuelling and discharge of pebbles. It is important to notice that there is no intention of reprocessing the pebbles. That is, a once through transmutation scenario is imposed.

Table 1. **Fuel composition**

Isotope	Discharged mass LWR (gr/ton U)	Isotopic composition	Isotope	Discharged mass LWR (g/t U)	Isotopic composition
^{236}**Np**	5.312E-04	4.575E-08	^{242m}**Am**	2.452E+00	2.112E-04
^{237}**Np**	6.514E+02	5.610E-02	^{243}**Am**	1.446E+02	1.245E-02
^{238}**Pu**	2.277E+02	1.961E-02	^{242}**Cm**	5.933E-03	5.110E-07
^{239}**Pu**	5.912E+03	5.092E-01	^{243}**Cm**	4.326E-01	3.726E-05
^{240}**Pu**	2.593E+03	2.233E-01	^{244}**Cm**	3.090E+01	2.661E-03
^{241}**Pu**	6.823E+02	5.867E-02	^{245}**Cm**	2.339E+00	2.014E-04
^{242}**Pu**	5.983E+02	5.153E-02	^{246}**Cm**	3.165E-01	2.726E-05
^{244}**Pu**	4.176E-02	3.597E-06	^{247}**Cm**	3.656E-03	3.149E-07
^{241}**Am**	7.651E+02	6.590E-02	^{248}**Cm**	2.440E-04	2.101E-08

Table 2. **Main specifications of a cylindrical prototype core**

Pebble-bed sub-critical prototype data	
Thermal power: 100 MW	Porosity: 0.396
Fuel sphere diameter: 0.06 m	Total gas volume: 0.835 m^3
Inner fuel zone diameter = 0.03 m	Mean radius of the neutron source channel = 0.29 m
Number of pebbles: 11 200	Active core height = 1.2 m
Thermal power per pebble = 9 000 W	Reactor core radius = 0.8 m
Pebble outer surface: 113 cm^2	Reactor cross-section area = 1.74 m^2
Total outer surface of pebbles:125 m^2	Average gas flow cross-section: 0.6975 m^2 (for an active height of 1.2 m)
Average heat flux: 8×10^5 W/m^2	Graphite reflector inner radius = 0.8 m
Total pebble volume: 1.255 m^3	Reactor vessel inner radius = 1.00 m (20 cm thick graphite reflector)
Total reactor volume: 2.09 m^3	Reactor vessel outer radius = 1.07 m

3. Effective cross-sections and transmutation rates

The calculation model was made up from an infinite array of hexagonal channels. Inside each channel the pebble-bed spheres are stacked from bottom to top, in an infinite array. Each sphere contained an inner fuel region of variable diameter containing a mixture of graphite and TRISO coated fuel kernels. For the calculations, the fuel zone was assumed to be homogeneous. MCNPX code was used for all the neutronic calculations using ENDF-B/VI libraries. The average effective microscopic cross-sections obtained are given in Table 3.

We can observe significant differences for different fuel radii. The average cross-section depends on the neutron spectra shown in Figure 3, which depends on the mass ratio between carbon and fuel atoms. For fuel region Rf = 2.5 cm, so we are going to have a high build up of ^{241}Pu due to the higher value of capture microscopic cross-section of ^{240}Pu than the absorption of ^{241}Pu, with the appearance of ^{241}Am and ^{237}Np as result of the decay, which have a high radiotoxicity and are more difficult to destroy. On the other hand, with Rf = 0.5 cm, all the cross-sections are small and it is not possible to achieve the desired burn-up, in particular for ^{242}Pu. The neutron spectrum in this case is hard. The best transmutation results are obtained with fuel regions having a radius of 1.5-2 cm and therefore the 1.5 cm was chosen as the basis for the present design. The transmutation of Pu isotopes for 1.5 cm is shown in Figure 4.

Table 3. Average microscopic cross-section for an infinite array of pebbles. Case of 2 grams of fuel per pebble

	Rfuel = 2.5 cm		**Rfuel = 2 cm**		**Rfuel = 1.5 cm**		**Rfuel = 1 cm**		**Rfuel = 0.5 cm**	
	Capture	**Fission**	**Capture**	**Fission**	**Capture**	**Fission**	**Capture**	**Fission**	**Capture**	**Fission**
^{237}Np	43.4594907	0.2431753	44.6410243	0.2495093	45.4299176	0.2677779	44.0585301	0.3206016	32.5835109	0.5329437
^{238}Pu	23.0414352	2.0672338	26.1688234	2.1537858	31.1980560	2.2972491	38.1362471	2.5198135	40.8787764	2.7071440
^{239}Pu	37.0417824	64.5387731	39.4894265	70.2675055	41.269663	76.2579395	40.2015133	79.5149652	31.3709412	69.7655902
^{240}Pu	184.515625	0.39656019	153.332879	0.39417989	114.638993	0.39956794	76.9422796	0.43448781	40.9676545	0.6160064
^{241}Pu	25.2185185	75.9947917	27.6275723	83.1830142	30.7858063	92.9785454	33.8962673	103.39741	31.805035	98.4604738
^{242}Pu	61.6064815	0.2074265	59.0063594	0.21262737	53.442918	0.22794492	41.5126133	0.27269487	17.7652483	0.45292418
^{241}Am	99.0798611	0.83390046	102.948982	0.85475025	105.002587	0.87309424	100.267171	0.88392484	74.4904723	0.94933279
^{243}Am	91.3512731	0.28546412	89.1338323	0.28893663	84.5810905	0.30078927	73.9322169	0.33711723	44.2873282	0.49500373
^{242}Cm	6.66435185	0.25365278	6.64541172	0.27884109	6.59329833	0.324211	6.993233	0.40709087	5.11853893	0.5936618
^{244}Cm	31.1480903	0.87228009	30.2226087	0.86749186	28.4144073	0.86336317	24.5326305	0.87094279	13.8792891	0.95424777
Cnat	0.00025757	0	0.00028724	0	0.00033754	0	0.00043462	0	0.000654	0

Figure 3. **Neutron flux spectra for different fuel radii**

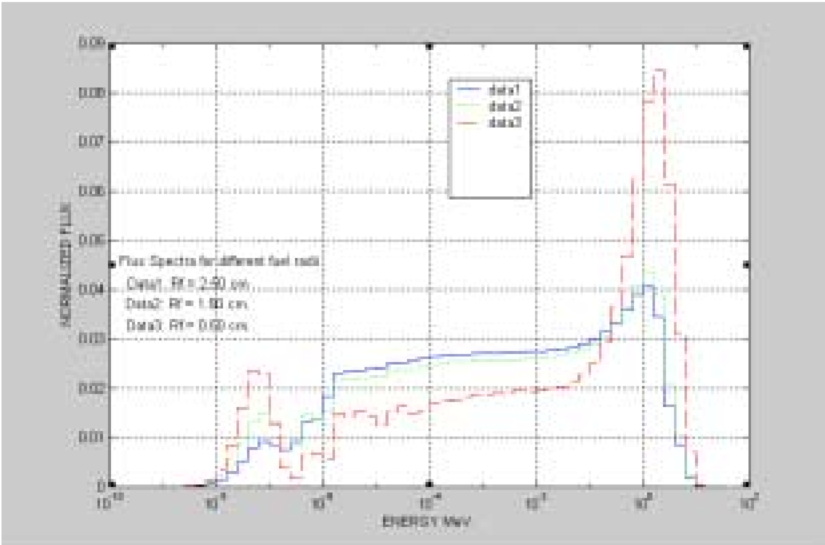
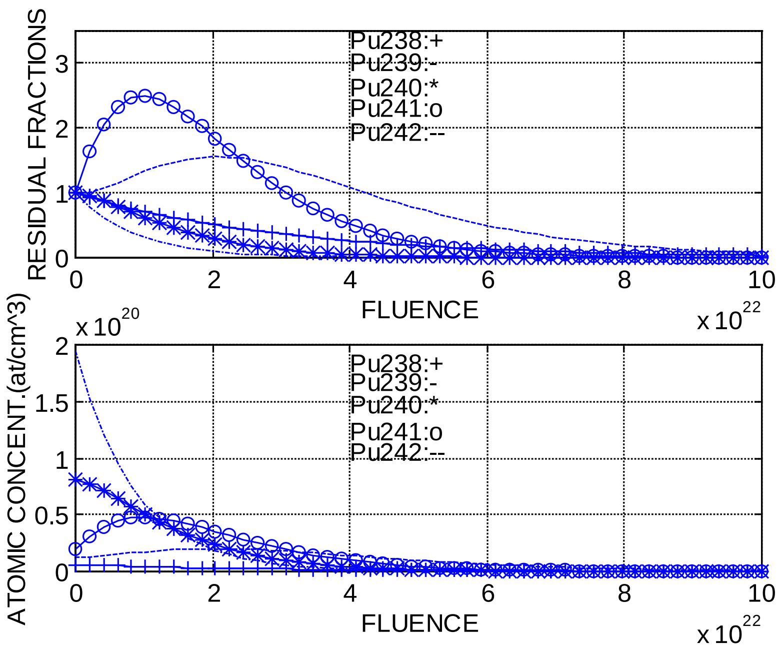


Figure 4. **Transmutation of Plutonium isotopes for Rf = 1.5 cm**



Assuming that the residual Pu fraction should be 0.001, then the other Pu isotopic concentration will be as shown in Table 4, where the fluence needed to achieve this degree of burn up is also shown. As can be seen from the table the chosen design of 1.5 cm results in the best transmutation. There is still more than 20% of ^{242}Pu left, however this Pu isotope has a low radiotoxicity.

Table 4. **Residual fractions of the fuel isotopes when the residual fraction of ^{239}Pu isotope is 0.001. Infinite array cell assumed.**

Fuel radius.	^{238}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Fluence
Rf = 2.5 cm	0.0985	0.0006	0.0119	0.0677	$9.23 \cdot 10^{22}$
Rf = 2 cm	0.0914	0.0009	0.0146	0.1131	$8.45 \cdot 10^{22}$
Rf = 1.5 cm	0.0767	0.0022	0.0254	0.2277	$7.67 \cdot 10^{22}$
Rf = 1 cm	0.0545	0.0143	0.0692	0.5361	$7.16 \cdot 10^{22}$
Rf = 0.5 cm	0.0313	0.0862	0.1658	1.4177	$7.95 \cdot 10^{22}$

The desired source intensity to achieve the transmutation levels in Table 4 as well as other core parameters are shown in Table 5.

Table 5. **Source intensity and proton beam requirements for a 10 MWt prototype, as a function of the radius of the fuel region, for two values of the proton energies**

Rf	k_{eff}	Average flux ($n/cm^2 \cdot s$)	Source (n/s)	Proton beam intensity (mA)	
				E = 0.45 GeV Yield = 10	E = 1 GeV Yield = 30
2.5 cm	0.6643	1.39E+14	4.10E+17	6.60	2.18
2 cm	0.71093	1.31E+14	3.20E+17	5.15	1.70
1.5 cm	0.76362	1.26E+14	2.43E+17	3.90	1.28
1 cm	0.79526	1.29E+14	1.98E+17	3.17	1.06
0.5 cm	0.69828	1.77E+14	3.36E+17	5.38	1.80

In this table we can see the relation of the k_{eff} value to the source needed to obtain a specific thermal power. The neutron population in a sub-critical reactor is given by equation:

$$\mathbf{N} = \frac{\mathbf{S} \cdot \mathbf{l}}{\mathbf{1 - k_{eff}}}$$

Where "N" is the neutron population inside the reactor, "S" is the neutron source, and "l" is the averaged neutron lifetime. The closer the k_{eff} is to unity, the smaller is the external neutron source needed. However safety characteristics must also be considered in choosing the level of sub-criticality. An optimisation of the k_{eff} value is needed for a final reactor design.

4. Problems with burn-up

An important aspect during the burn-up of the fuel is the decrease of k_{eff}. In the fuel loaded, there is only a small amount of fertile ^{240}Pu, consequently the k_∞ of the pebble decreases significantly with burn-up, as can be seen in Figure 5. In order to maintain a constant power (and k_{eff}) in the core, an appropriate recycling strategy has to be developed.

A possible way to maintain the k_{eff} with the burn-up is the increase of the fuel content in a pebble. However, an increase of the mass concentration can give a lower k_∞, as we have seen in the case of Rf = 0.5cm. We can see this results in Figure 6, which shows the needed increase of the fuel in the pebble to obtain higher values of k_∞.

Figure 5. **Variation on the k_∞ with burn-up**

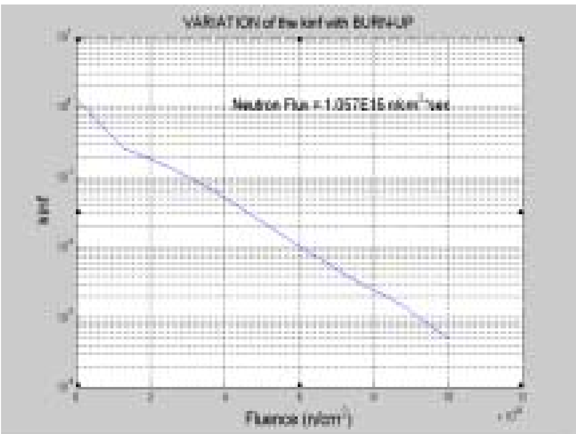
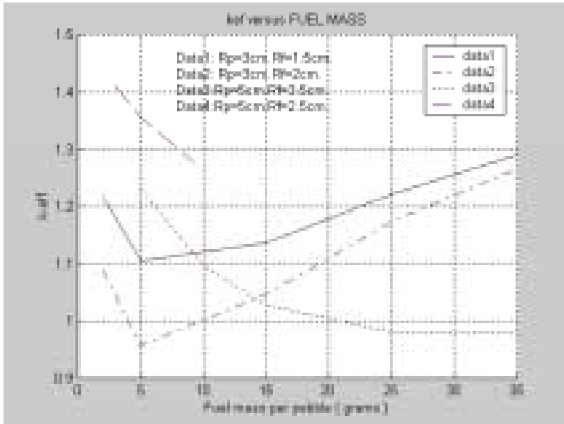


Figure 6. **Variations on the k_∞ with fuel mass per pebble**



The effective microscopic cross-sections changes significantly with the mass charged per pebble. If the mass is very high, the neutron spectra become hard, and the effective microscopic cross-sections are low. That means that we can not vary the mass loaded per pebble with Rp = 3 cm, it must remain at 2 g. Another possibility is to change the pebble size and increasing the fuel sphere radius to 5 cm (with the internal fuel region 2.5 cm) and 9.26 g of fuel per pebble. A much higher k_{eff} of the core is obtained as shown in Figure 6. The total quantity of fuel in the core is the same as with 3 cm pebbles, and higher big burn-up can be achieved. The possibility of employing larger fuel spheres permits more flexibility in designing the core, the k_{eff} and the refuelling strategy. However, one should note that until today there is no experience with pebbles larger than 6 cm.

These results open additional options. Maintain the pebbles radius in Rp = 3 cm and try to obtain a suitable recycling strategy to achieve a nearly constant k_{eff} during the burn-up of the fuel, or change the original design, to permit variations in the fuel contents per pebble, so as to increase the k_{eff}, with a suited recycling strategy. Further studies to optimise the pebble size and its content will be performed.

Another key point is the power distribution in the reactor during the burn-up. The pebbles with a higher burn-up will produce less power, so the axial power distribution inside the reactor will vary significantly from top to bottom. Another objective of the recycling strategy is to try to obtain a power distribution as uniform as possible. In the case of ADS, we have and additional parameter design, which is the location of the external neutron source inside the reactor, which can be used to improve the system performance.

To achieve a high destruction rate of TRU the fuel sphere has to withstand high burn-up and fluence. From the previous work on pebble-bed reactor at FZA (Schenk) it was concluded that 600 MWd/kg was easily endured by the pebbles. In order to understand it and to explain how such high burn-ups are achievable, note that in a 6 cm diameter pebble there will be more than 240 g of carbon and two grams of TRU. In terms of nuclei, it means $1.2 \cdot 10^{25}$ of C and $5 \cdot 10^{21}$ of TRU, i.e. the number of C nuclei is near 2 400 times larger than the number of TRU nuclei. Taking into account that in the neutron cycle 3 neutrons are born per each TRU nuclei eliminated by fission, including source neutrons. Some of them escape in the moderation process, but an average of 80 elastic collisions with carbon are suffered by a neutron. The lethargy gain per collision is 0.1577 in this case and a slowing down from 3 MeV to 1 eV, 14.9 lethargy units must be passed. Totally there will be about 240 collisions in C for each fission. If it is taken into account that the number of C nuclei is about 2 400 times larger than the number of TRU nuclei, the number of C nuclei affected in a neutron cycle is one tenth of the number of TRU affected nuclei.

A total estimate of collisions per C atom along a fluence can be obtained including the very important burn-up effect. Let σ_f be the average microscopic fission cross-section. The neutron fluence to achieve a residual fraction r, is given by:

$$\mathbf{r} = \frac{\mathbf{Na(t)}}{\mathbf{Na(0)}} = \mathbf{e}^{(-\sigma_f \cdot \Phi \cdot t)}$$

where Na is the number of actinide (TRU) nuclei. On the other hand, the number of fission neutrons per carbon atom is:

$$\mathbf{Tc} = \frac{\sigma_s \cdot \mathbf{Nc}}{\mathbf{Nc}} \cdot \Phi \cdot \mathbf{t} = \sigma_s \cdot \left(-\frac{\ln \mathbf{r}}{\sigma_f} \right)$$

For instance, for σ_s = 4.5 b (scattering microscopic cross-section), an effective σ_f = 70 b, for r = 0.05 (95% of burnt-up TRU) 0.19 collisions per C atom are obtained, which seems to be a moderate value that could be withstood by the graphite matrix.

5. Safety characteristics

Calculations have been performed to asses the safety characteristics of the proposed design. The accidents studied included: water ingress, changes in reactor pressure and changes in the void fraction. In all cases the reactor remains sub-critical, except during water ingress. In a sub-moderated reactor, as the PBT presented in this paper, the ingress of water is accompanied by a large increase in reactivity. The good moderation properties of water lead to higher k_{eff} values. Consequently, in the proposed design, water ingression must be excluded. As the fuel loaded has a small amount of fertile material, the Doppler effect will have little importance. At the beginning of cycle, the presence of ^{240}Pu with a very high capture cross-section, and a broad resonance region at high energies leads to a small Doppler effect with a reactivity coefficient of $\rho = -0.52 \cdot 10^{-5}$ ΔK/K. During the burn-up of the fuel this coefficient will change.

6. Conclusions and future work

A preliminary conclusion drawn from the present analysis is that it is theoretically feasible to eliminate a large fraction of transuranics by means of an ADS pebble-bed transmutator, without the need of chemical reprocessing. Such reprocessing would be almost impossible to do, because of the resistance of the graphite matrix. In fact, the objective is to make TRU-fuelled pebbles from the spent LWR fuel, and to burn them in successive burn-up cycles by unloading and reloading the pebbles in an appropriate mixture with fresh fuel. Further work must be done in the recycling studies and in the optimisation of the size and TRU load of the pebbles. The objective is to reach an elimination fraction higher than 99% of ^{239}Pu, and higher than 95% in the rest of the offending nuclei. An overall goal is to eliminate about 95% of the TRU.

The final answer to the question of the capability of the pebble to withstand the high neutron fluence has to be of an experimental nature, but it is important to asses that the sought value of fuel burn-up does not result in an unbearable number of collisions per C atom. This point supports the feasibility of massive TRU elimination by an ADS pebble-bed transmutator.

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