PEBBLE BED REACTORS FOR ONCE THROUGH NUCLEAR TRANSMUTATION.

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Abstract

Pebble bed reactors fuel elements are specially suited for withstanding very high burn-up values. This is one of the main characteristics of a transmutation fuel. The higher the burn-up achieved in the fuel, the smaller the number of reprocessing steps needed for obtaining a high transmutation rate. In this paper, a new transmutation cycle option will be introduced, based in a single pass of the fuel through the reactor, with no reprocessing steps in the cycle. With a maximum burn-up of 700 MWth, the fuel transmuted is nearly 75% of the actinides originally charged in the reactor. For a minimisation of the radiotoxicity in the fuel element, the 25% of the mass of actinides remnant in the fuel after transmutation have to be low radiotoxic isotopes. For example, ^{242}Pu, which has a long half life, and by α disintegration, it converts to ^{238}U, beginning the Uranium cycle.

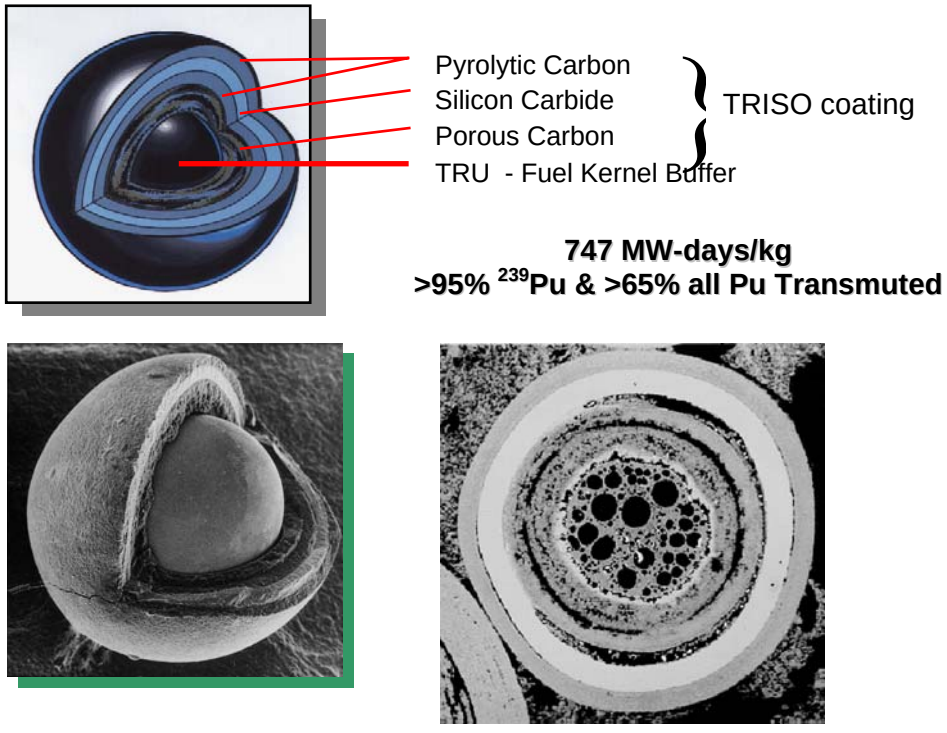
In the paper, the optimum spectrum for minimising the final radiotoxicity is studied, and the possibility of initially drive the reactor as a critical reactor, with a last subcritical step, is analysed. In the calculations, it is shown that a good reduction in radiotoxicity of the fuel can be achieved by this strategy, with no reprocessing steps during the transmutation phase. The final disposal of the fuel elements, which have very good confinement characteristics, is also an additional factor for the use of Pebble Beds as Transmutation reactors.

Once Through Transmutation Scenario

The Once Through Transmutation scenario permits the minimisation of the reprocessing steps needed for transmutation of spent nuclear fuel. The spent fuel from commercial reactors, after a given burn-up, and a cooling period, has to be reprocessed to separate the Minor Actinides, Plutonium, rest of Actinides (impoverished Uranium) and Fission Products. The goal of the Once Through Scenario is to minimise the number of reprocessing steps, with one reprocessing of the spent fuel prior transmutation (this step is necessary in a non-transmutation scenario, for a final disposition of the spent fuel in a Deep Storage Facility), and a reprocessing step after transmutation. The minimisation of the number of reprocessing steps implies a decrease in the cost of transmutation, and a decrease in the risks associated to operating with high radiotoxic elements.

In the case a Once Through scenario is chosen, the burn-up of the fuel have to reach very high values. In the literature, one of the fuel elements that can withstand higher burn-up values is the TRISO Coated Fuel Particles. A 747 MWd/kg BUP has been reported, for a PuO_2 based fuel. During the Burn up, more than 95% of the Pu^{239}, and more than 65% of the total mass of Pu isotopes were transmuted in a thermal spectrum. From the figure, it can be seen that the TRISO particle is capable of confine perfectly the fission products generated during burn-up. In this analysis, a maximum burn-up of 700 MWd/kg have been taken.

Figure 1. **Maximum Burn-up for a Triso Coated Fuel Particle**



Description of the Isotopic Composition of the Spent Fuel

The isotopic compositions of the spent fuel after 40 MWd/kg BUP, in a commercial LWR, and after 15 years cooling, are given in Figure 2.

Figure 2. **Isotopic composition of the Spent Fuel, from LWR, after 40 MWd/kg and 15 years cooling**

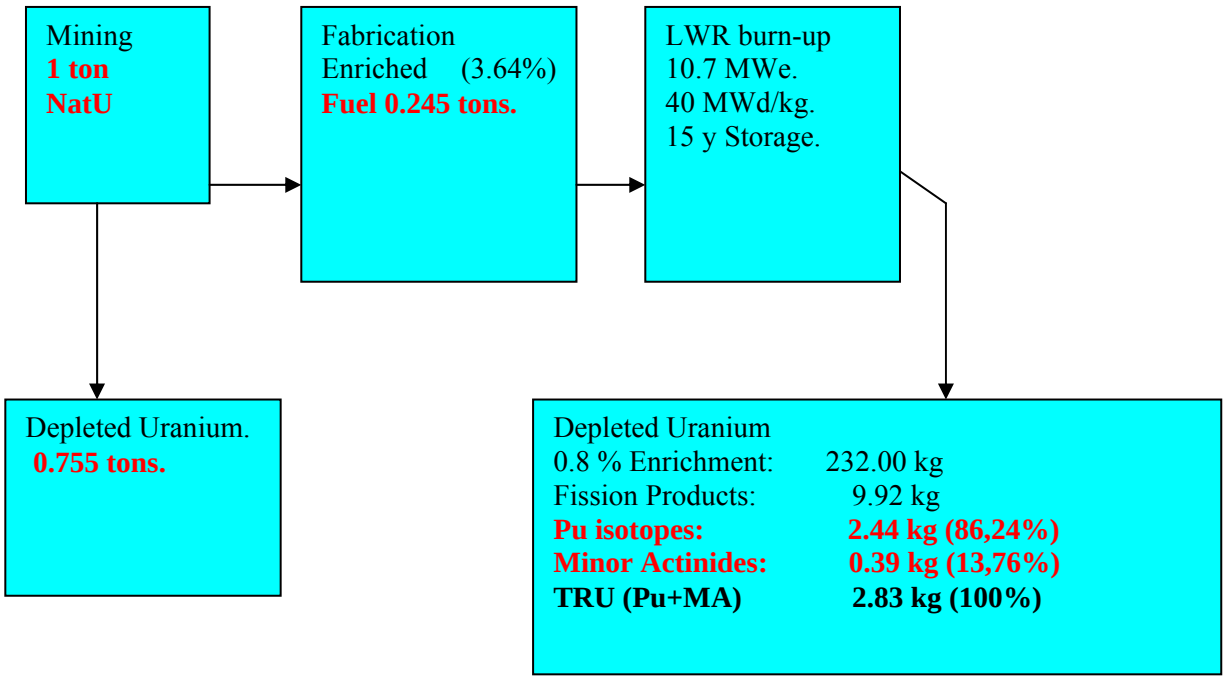
Element	Mass %
Np	5.61%
Pu	86.24%
Am	7.86%
Cm	0.29%

Pu Isotopes	Mass %
Pu238	2,27%
Pu239	59,04%
Pu240	25,90%
Pu241	6,81%
Pu242	5,98%
Pu244	0,00%

From the above figure, it can be seen that the composition of the fuel is mainly Pu (86.24%), specially Pu^{239} and Pu^{240}. Pu^{242} initially have a lower contribution in the final mass after BUP. But the important characteristic for evaluation the potential risk of the spent fuel is not the mass of the actinides; it is their radiotoxicity (in ingestion effective committed dose.) Of course, in the calculation of the final Deep Storage Facility, it is important also the evolution with time of the isotopes heat deposition, and their migration capabilities in the biosphere (for example, Np^{237}.)

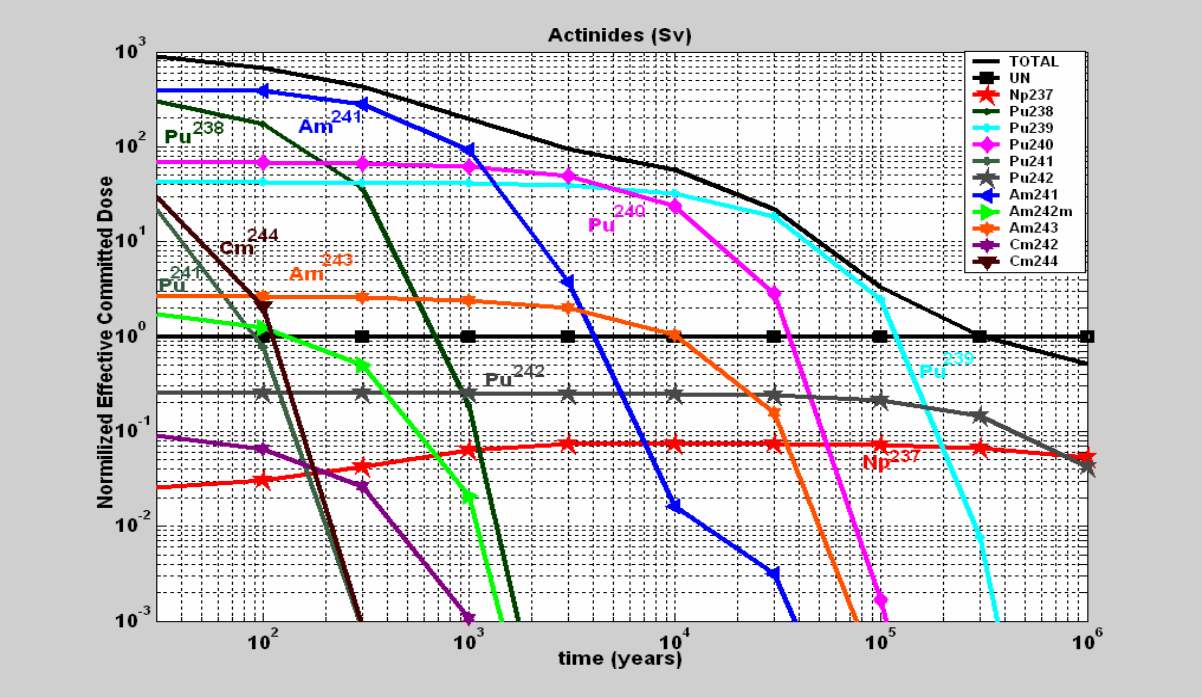
Initially, a calculation of the ingestion effective committed dose of the actinides of the spent fuel (Figure 2) was done. To understand the result, it is important to know which is the fuel cycle taken as reference. In our case, the reference is an open fuel cycle, where Pu and the rest of Minor Actinides are taken as waste. The fuel cycle is defined in Figure 3.

Figure 3. **Nuclear Fuel Cycle for Analysis**



From the chart, per Natural Uranium tone, 2.83 kg of Pu and Minor Actinides are generated. The Natural Uranium (NatU) Dose has been chosen as reference for Dose calculations, taking into account the production of actinides per ton of NU. From ICRP 68, the dose of NU is 19.7 Sv/kg. The doses of the rest of the isotopes have been evaluated with ICRP 68. ICRP 72 gives a higher dose value for NU (30.8 Sv/kg), due to the increment of radiotoxicity of Po^{210}. In this last case, the reference level is higher. Also, the same calculation has been done for a different unit of the dose, m^3. The calculations in this case have been obtained with ORIGEN code. The results in the time the actinides without transmutation reach the Natural Uranium reference value are quite different (300.000 years for Sv, 20.00 years for m^3.) Figure 4 represents the results of the dose in Sv for the spent fuel without transmutation. It can be seen that Pu isotopes and daughters (Am^{241} from Pu^{241}) are the principle contributors to the effective committed dose.

Figure 4. **Ingestion Effective Committed Dose in Sv**



In a thermal reactor, the behaviour of the Pu isotopes during transmutation is fission by Pu^{239} and Pu^{241}, and the rest of the isotopes capture neutrons and continue the Pu isotopic chain (from Pu^{238} to Pu^{242}.) For Pu^{242}, a neutron capture gives Pu^{243}, with a half life less than 5 hours, that decays by β^- to Am^{243}, starting the production of Am and Cm isotopes during transmutation. These isotopes, as it will be seen later, have a high radiotoxicity inventory.

Once Through Strategy

If the 700 MWd/kg is taken as BUP value for the once through strategy, a certain mass of actinides from the spent fuel is not transmuted. We take into account the following fairly accurate formula that connects the BUP with the final fraction mass of actinides not transmuted

$$\mathbf{BUP\ (MWd/kg) = 975.9 \cdot (1\text{-}RF(mass))}$$

where RF is the final residual fraction of the actinides not transmuted. In the case of a 700 MWd/kg BUP, this fraction is 0.2827. This means that 28.27% of the actinides mass initially charged are not transmuted. The isotopic composition of this mass is very important for the final ingestion dose stores in the Deep Storage Facility. Cm and Am isotopes have a very high dose per mass (Sv/kg). Cm isotopes (Cm^{242} to Cm^{248}) decay by 'α' disintegration to Pu isotopes, so the final radiotoxicity evolution with time is high (especially for Cm^{244}.) Am isotopes (Am^{243} to Am^{241}) behave differently than Cm isotopes. Am^{243} and Am^{241} decay by 'α' to Np. Np^{239} decays to Pu^{239} by 'β-', and Np^{237} decays by 'α'. Am^{242} decays primarily (83%) to Cm^{242} by 'β-' disintegration, and the rest to Pu^{242}.

If 28.27% of the actinides after transmutation were Am and Cm isotopes, the final radiotoxicity would be very high, and the dose evolution would be very similar as the one without transmutation. A way to minimise the dose is to stop the Am and Cm production by minimizing the captures of Pu^{242}. Pu^{242} has a half life of $T_{1/2}$ = 3.7E5 seconds, and by 'α' disintegration, it decays to U^{238}, the isotope that starts the fuel cycle. So, if the final composition of the remnant actinides is mainly Pu^{242}, the radiotoxicity is going to be low, and the final isotope after decay is NU.

To quantify the advantages of this strategy, a theoretical analysis have been carried out. In the analysis, Am and Cm from the spent fuel are not transmuted (since Cm isotopes are very difficult to transmute in a thermal spectrum), and the rest of isotopes (Np and Pu) are transmuted to a certain degree, and after transmutation, it converts to fission products, and the rest of actinides not transmuted are Pu^{242}. Figure 5 represents the theoretical analysis. Results are depicted in Figure 6, in Sv.

From the results, it can be seen that with this strategy, an important reduction in the dose of actinides not transmuted can be reached. In the case of a 700 MWd/kg BUP, the actinides mass remnant in the fuel is near 30%, with the rest (70%) transmuted to fission products. This means that the final radiotoxicity level is quite similar as the Natural Uranium ore. If the BUP achieved by transmutation is higher, then the final radiotoxicity value can be below the NU reference level.

To obtain a final accumulation of Pu^{242} isotopes after transmutation, the capture cross section of this isotopes have to be minimised. In the case of a thermal spectrum, the neutron flux level, with the isotopic composition given in Figure 2, is defined by the fission cross section of Pu^{239}, the concentration of the isotope and the power density. So, the capture cross section of Pu^{242} has to be low *as compared to* the fission cross section of Pu^{239}.

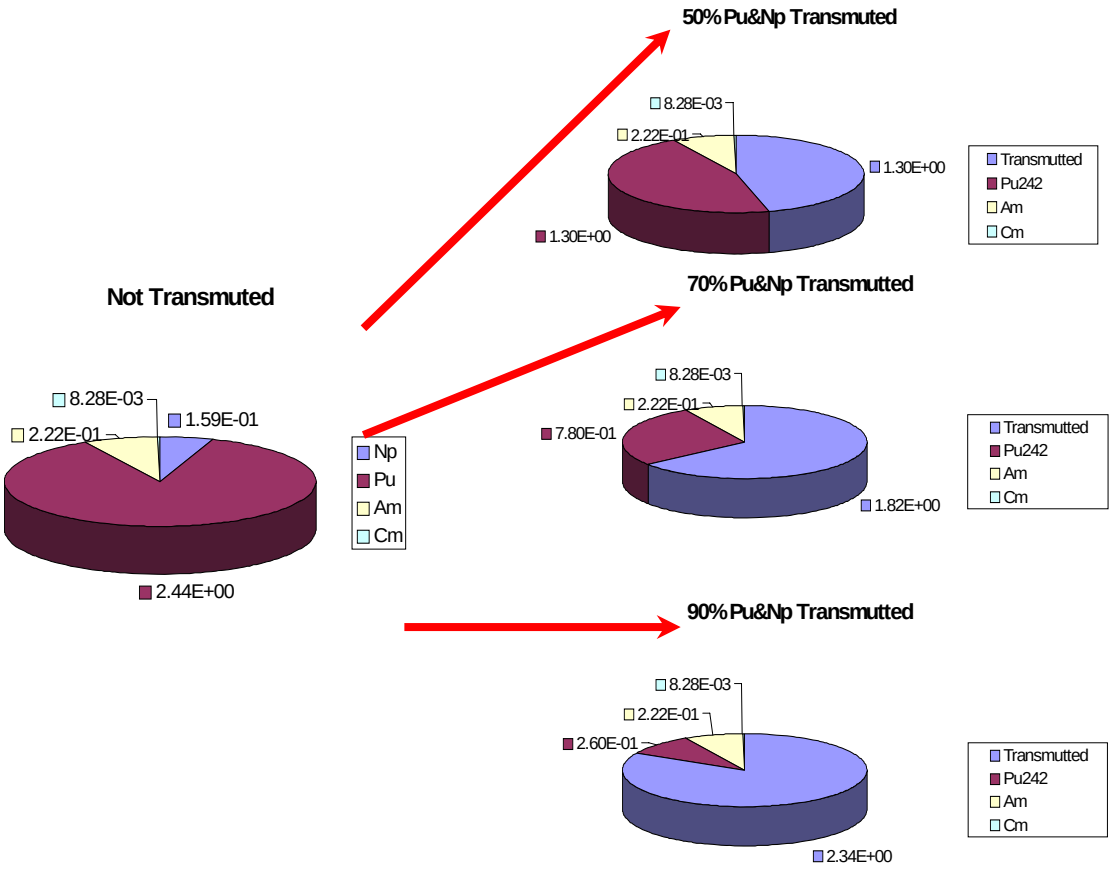
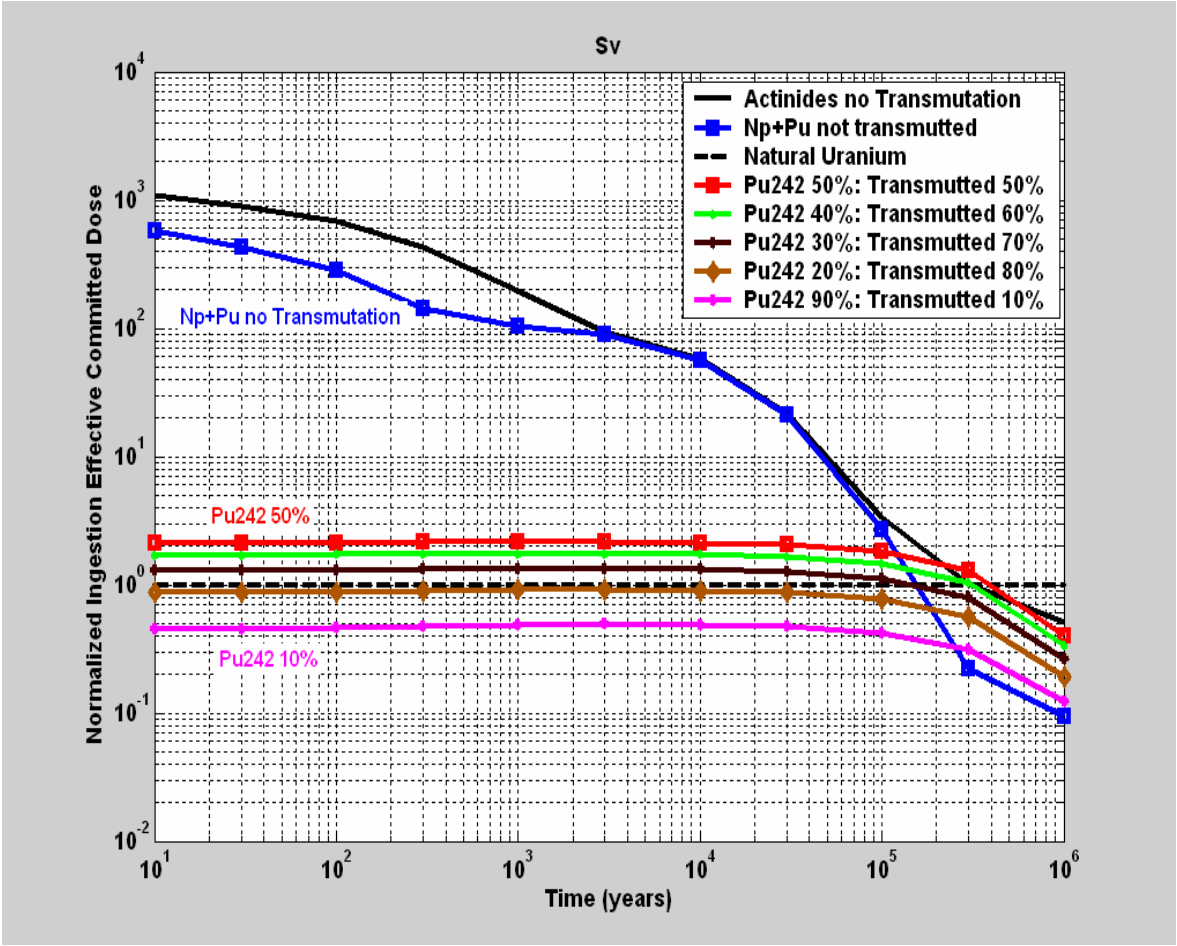


Figure 5. **Theoretical analysis of Pu^{242} accumulation during transmutation phase**

Figure 6. **Theoretical analysis of Pu^{242} accumulation during transmutation phase:dose**

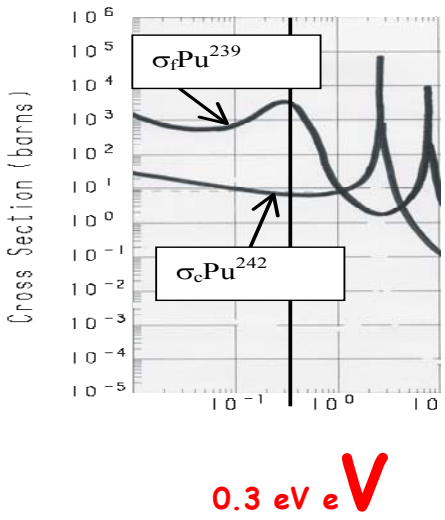


This defines the following spectral index:

$$i = \frac{\sigma_c^{Pu^{242}}}{\sigma_f^{Pu^{239}}}$$

The smaller the spectral index, the higher the accumulation of Pu^{242} after transmutation. To obtain an optimum spectrum, Figure 7 defines the cross sections of Pu^{239} (fission) and Pu^{242} (capture.) From the figure, it can be seen that the optimum spectrum is for a neutron energy of 0.3eV.

Figure 7. **Cross section of fission Pu^{239} and capture Pu^{242}**

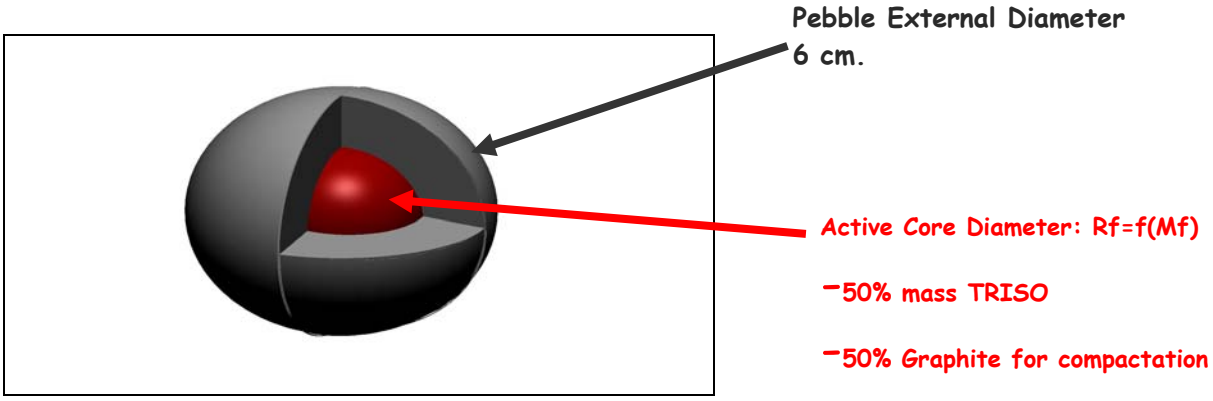


After the theoretical analysis, a physical model has to be developed to obtain the optimum spectrum to minimise Pu^{242} neutron captures. The physical model chosen is a Pebble Bed Reactor.

Pebble Bed Transmutator

The advantage of pebble bed reactors fuel for transmutation is that it can withstand very high BUP, and that the fuel can be designed to obtain different neutron spectra. It is easy then to tailor a neutron spectrum that suits with minimizing Pu^{242} captures. In the analysis, the following fuel model has been studied (Figure 8.) The external pebble radius is the standard 3 cm, and the active core has a different radius as a function of the fuel mass charged per pebble. The active radius is going to be composed of TRISO coated fuel particles (50% of the active zone mass), and graphite for compact the active zone (50% rest of the mass.) If, for example, the fuel charged per pebble is 1gr, the number of TRISO coated fuel particles is fixed, and the mass of the TRISO is also fixed. Once the masses are known, the volumes are evaluated by the different densities of TRISO and graphite.

Figure 8. **Fuel Model for calculations**



Small active zones give a more thermal neutron spectrum. To optimise the spectrum for a minimum spectral index defined above, the following calculations have been performed (see Figure 9.)

Figure 9. **Optimisation of the fuel design for a minimisation of Pu^{242} neutron capture**

Mass charged per Pebble	Active zone Radious	$i = \frac{\sigma_c^{Pu\ 242}}{\sigma_f^{Pu\ 239}}$	$\sigma_f Pu^{239}$
Mf = 0.25 gr	0.340 cm.	1.63E-01	2.33E+02
Mf = 0.5 gr	0.428 cm.	4.61E-02	1.74E+02
Mf = 1 gr	0.539 cm.	1.19E-01	9.37E+01

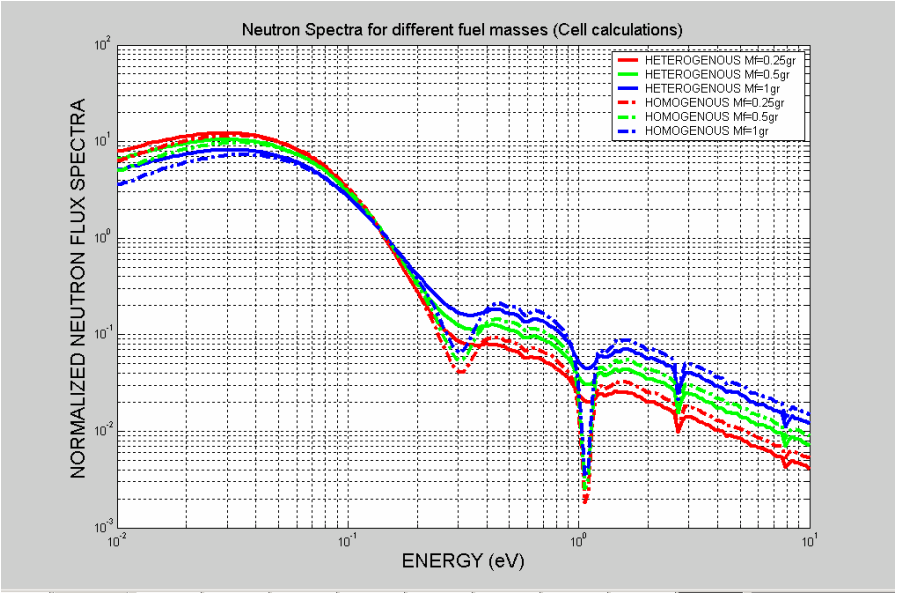
From the above figure, the optimum fuel mass is 0.5gr, for a 0.428 cm active zone radius. This calculation has been performed by an infinite array of pebbles. An important study is the analysis of the definition of the active zone for calculations. The homogeneous calculations take into account a perfect mix between fuel and graphite, while the heterogeneous calculation, much more intensive in computing time, take into account the perfect definition of the TRISO coated fuel particles, well differentiated from the graphite used for compacting. Figure 11 shows the importance of defining the pebble as a heterogeneous system, since the neutron spectrum for the homogeneous and heterogeneous systems are quite different. The differences in neutron spectrum give differences in the cross sections. For example, the fission cross section of Pu^{239} is 185.24 in the homogeneous calculation and 233.71 in the heterogeneous one. The calculations have to be performed with a heterogeneous calculation of the active zone.

After the initial neutronic analysis, some thermo-hydraulics analyses have been carried out. The reactor parameters are given in Figure 10.

Figure 10. **Reactor analysis for thermal-hydraulic calculations**

Reactor Parameters	
Total Power	20 MW
Number of Fuel Elements (Pebbles.)	193,750 Pebbles
Mass Charged per Pebble	0,25 gr
Pebble Active Zone Radius.	0,34 cm.
Power per Pebble	103,22 W/Pebble
Power Density	626,96 W/cm^3

Figure 11. **Neutron spectrum for homogeneous or heterogeneous active zone**



The radius of 0.34 cm, corresponding to 0.25gr charge per pebble, corresponds to the critical case in the refrigeration scheme. The axial and radial power distribution of the reactor is given in the following figure.

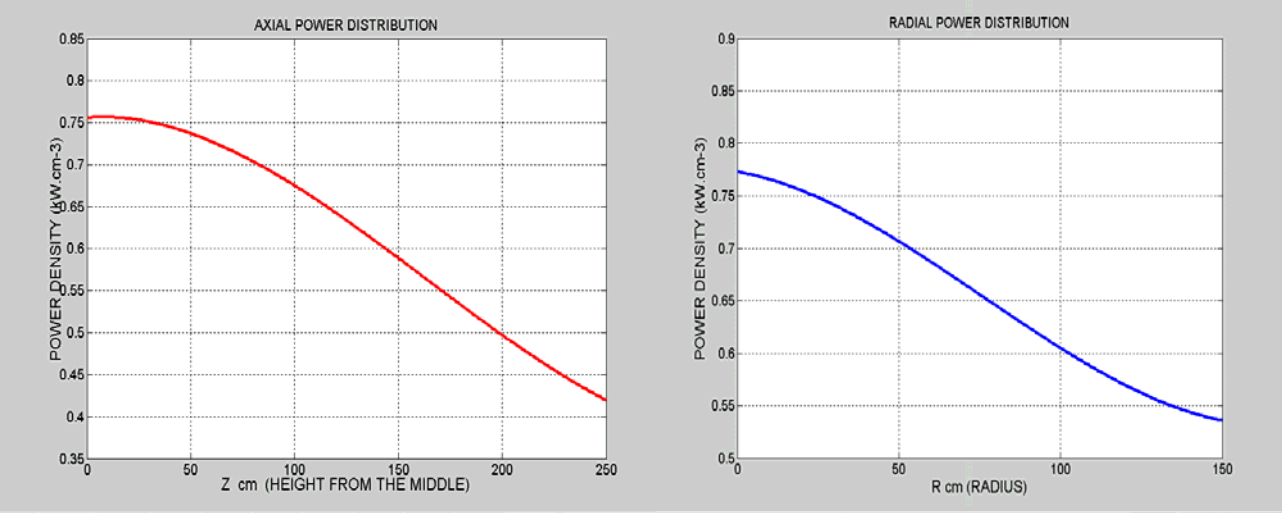
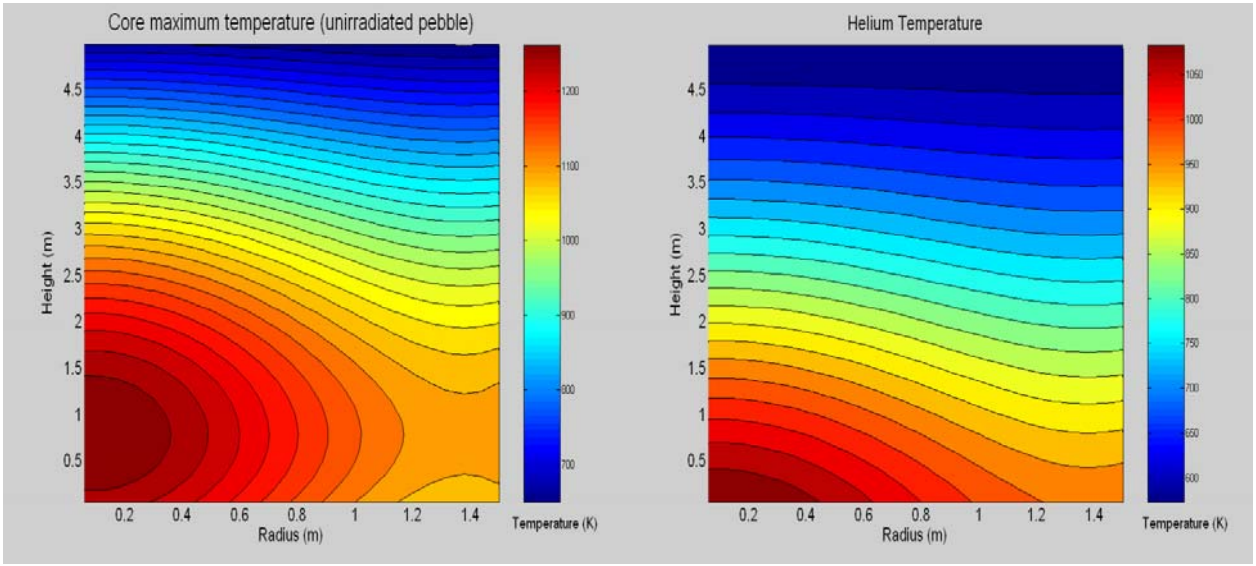


Figure 12. **Axial and radial distribution for PBR transmutator**

With these core parameters, and with the following axial and radial power density distribution, the core maximum temperatures, for He gas cooling, are defined in Figure 13. The maximum temperature is below 1,300 K, near 950 ºC. This maximum core temperature leads to the next figure, which represents the He temperature of the reactor. The coolant flows downwards, and the maximum outlet temperature is bellow 1,100 K. An optimisation of the system can lead to H_2 production, if the outlet temperatures of the He are high enough.

Figure 13. **Maximum fuel temperatures and He temperatures for the PBT**



Conclusions

Pebble bed reactors present excellent characteristics to be used in a "Once Through Strategy" transmutation scenario. The high BUPs achieved, and the possibility of tailoring different neutron spectrum, are among them. The Pu^{242} accumulation in the 28.27% of mass not transmuted after 700 MWd/kg BUP minimise the final radiotoxicity for this strategy. From the analysis, heterogeneous calculations have to be performed in the active zone, defining TRISO coated fuel particles geometry and material in the MCNPX model. An additional advantage of the PBT scenario is the minimisation of Pu^{239} after irradiation, following a non-proliferation strategy. Finally, a high temperature operation of the reactor can lead to H_2 production.

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