A Preliminary Assessment of the Transmutation Potentialities for an ITER-like FW Sector Loaded with MA

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Abstract: The fusion-fission hybrid reactor is a promising technology that is likely to assume an increasingly important role in the global energy scene in the coming years. This kind of reactor can use both the nuclear fusion and fission processes to produce energy: neutrons from fusion reactions are used to sustain the fission of a sub-critical system. This method allows to have an intrinsically safe facility, with higher efficiency than a fusion reactor itself and with a harder neutron energy spectrum than a fission reactor, which could be suitable for nuclear waste transmutation. This paper, in particular, analyzes a type of hybrid reactor for the transmutation of Minor Actinides (MA). Nuclear waste, in the oxide form, is inserted as an element of the First Wall (FW) of an ITER-like fusion reactor. The aim is to demonstrate the feasibility of the transmutation of the MA characterized by higher long term radiotoxicity into shorter lived nuclides. The neutron transport in a detailed 3D geometrical model of the ITER reactor (B-lite) was performed by MCNP6 code, while the transmutation of the MA loaded in a single element of the FW was performed by SERPENT2 code. A pulsed ITER-like irradiation scenario was used. The analysis, which must be considered as a preliminary feasibility study, lead to very promising results, which could be further improved with a longer DEMO-like irradiation scenario and a larger number of MA loaded ("fission waste") elements loaded in the FW.

Keywords: SERPENT, MCNP, MCAM, burnup calculations, minor actinides, hybrid reactors.

1. INTRODUCTION

In nuclear systems, neutron multiplication is produced by the fission reaction, which is induced by the free neutrons created in the previous fission reactions; this behavior leads to the concept of chain reaction, which is self-sustained in a so called critical reactor.

On the contrary, in a subcritical system, the nuclear properties of the nuclear fuel and other components are unable to keep the chain reaction going on, and both the neutron population and the fission reaction rate vanish in a very short time [1].

There are some artificial heavy nuclei and some mixtures of heavy radioactive nuclei with a non-negligible strength of neutron emission. However, more powerful sources are needed to feed a subcritical system if we want to have a power density similar to that of a critical reactor. Generally speaking there mainly are two kind of reactions useful as (potential) independent neutron sources:

- A spallation reaction induced by accelerated charged particles impinging in a target of a heavy element (e.g. lead) [30-33].
- A fusion reaction.

Between these two potential choices, the latter is receiving more and more attention by the international scientific community. This is the basis of the hybrid reactor concept, where neutron generation is not produced just in neutron-induced fissions, but as a "by-product" of the fusion reactions inside the nuclear fusion reactor "core" (i.e. respectively, the void chamber for MCF or the fuel particles for ICF). Although the fusion-fission hybrid concept dates back to the earliest times of the fusion projects (when it was recognized that using fusion neutrons to "support" nuclear fuel cycle could vastly increase the exploitation of the fusion plants), it appears to receive relatively limited attention since the mid-1980s [2]. Notwithstanding, hybrid fusion fission systems have been already studied for some decades, in the most prominent laboratories and a large bibliography was produced [3-29]. Obviously much more papers on this topic have been published in more recent years (just to give some examples up to the end of the previous decade, see [34-48].

2. HYBRID FUSION-FISSION SYSTEMS

As already anticipated, fusion can also be combined with fission in what is referred to as hybrid fusion-fission system, where the blanket surrounding the core represents a "subcritical fission reactor" (i.e. the neutrons are captured, resulting in fission reactions

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E-ISSN: 2409-5818/15

taking place); instead, the fusion reaction acts as a source of neutrons for the surrounding blanket. These fission reactions would also produce more neutrons, thereby assisting further fission reactions in the blanket itself.

The concept of hybrid fusion-fission systems can be compared with an accelerator-driven (subcritical) system (ADS). The blanket of a hybrid fusion system could therefore contain the same fuel as an ADS: for example, the abundant element Th [52] or the long-lived heavy isotopes present in the spent nuclear fuel (SNF) could be used as fuel (see, as examples, [39, 41, 42, 47].

The blanket containing fission fuel in a hybrid fusion system would not require the development of new materials capable of withstanding constant neutron bombardment, whereas such materials would be needed in the blanket of a "conventional" fusion system. A further advantage of a hybrid system is that the fusion process would not need to produce as many neutrons as a "standard" (i.e. non-hybrid) fusion reactor would in order to generate more power than is consumed (so a commercial-scale fusion reactor in a hybrid system does not need to be as large as a fusion-only reactor).

To synthetize, some potential benefits of a fusion-fission hybrid plant could be [66]:

- Inherent safety (when the plant shuts down no runaway fission reactions are possible).
- Proliferation resistance (the plant does not require any enrichment or fuel reprocessing with isotopic separation, so the fuel would not be suitable for use in weapons).
- Reduction of hazardous materials (the plant would transmute most of the long lived isotopes present in SNF).
- Reduction of the need for high-level waste repositories (only about 5% as much repository space would be needed, compared with current OTTO fuel cycle in LWRs).

In the last years, many initiatives on nuclear waste transmutation were proposed in order to reduce the long-term radiotoxicity of the wastes by eliminating a high fraction of the transuranic elements (TRU) from the SNF before their final disposal [49-65]. In this frame, as already anticipated, hybrid fusion-fission systems have an additional degree of freedom because of the independent source: this means that the neutron spectrum can be (reasonably) tailored for the transmutation purposes [1].

Differently from almost all the studies carried out in the past on this topic, in the present study the burnup calculations have been performing under the hypothesis to consider the "real" behavior of a planned fusion plant (namely ITER), i.e. the pulsed steps considered in our burnup calculations have the same duration of those ones planned for the real reactor.

3. CALCULATIONS INSTRUMENTS AND METHODS

Monte Carlo (MC) codes chosen for our calculations are widely known for nuclear applications: namely we used MCNP6 [67] and SERPENT2 [68, 70, 71] (a new version provided to registered nuclear community members already using SERPENT1 [69] version). Both the codes have a self burnup routine but only the SERPENT2 allows to perform burnup calculations with an external (a-priori fixed) neutron sources in subcritical systems: in fact the eigenvalue solution (i.e. the so called kcode mode) could not provide correct solutions for the present case. Moreover MCNP6 does not work in nps mode for burnup calculations, while SERPENT2 has this capability; additionally the latter is relatively faster with respect to other available burnup codes (see, as an example, [55]).

Due to the fact that we have used to simulate the fusion plant (ITER) a very complex input already available for MCNP (namely ITER B-lite V3 [79]), we performed the static calculations on the whole fusion reactor (ITER) by MCNP6, and the burnup calculations on a single "modified" FW "sector" (see par. 5 for further details on this issue) using SERPENT2 code. Particularly, the B-lite V3 model [79] was run in nps mode with 1 million of particles simulated.

Regarding the cross sections, we have performed the calculations on the whole fusion reactor (by MCNP6) using a FENDL2.1 based dataset [72] (more suitable for "pure" fusion calculations) while we used a JEFF-3.1 based dataset [73] (more suitable for fission-based burnup calculations) for the burnup calculations (by SERPENT2 code).

It is important to remark that, in order to extract a proper FW sector from the complex and detailed geometry (described in the MCNP6 input) to be analyzed in the burnup calculations (by SERPENT2 code), the MCAM software [74] was used, too; in detail:

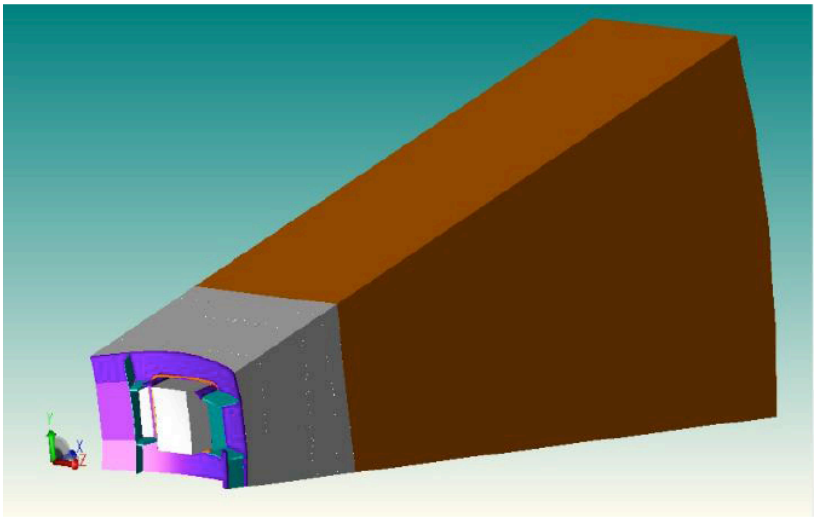

Figure 1: FW and blanket sector analyzed in the burnup calculations, figure created with MCAM.

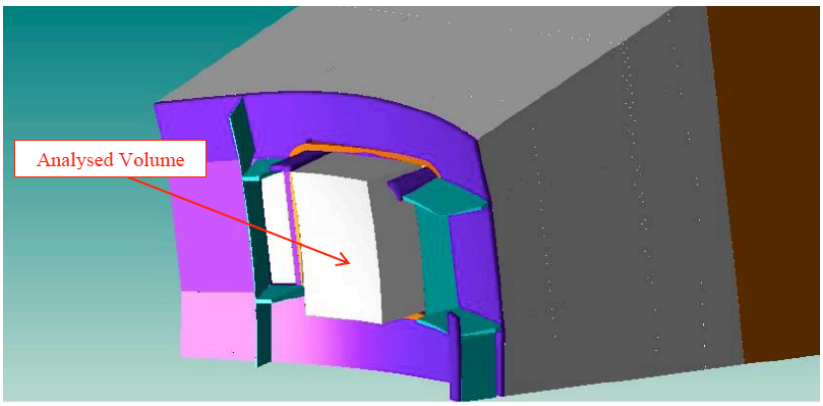

Figure 2: Detail of the FW sector analyzed in the burnup calculations, figure created with MCAM.

- Firstly, we focused on a (relatively) large section of the blanket, focusing step-by-step on a more limited FW sector,
- Then, starting from the input file B-lite V3 [79] has been possible to "extract" through MCAM software [74] (now SuperMC/MCAM), the analyzed FW sector. The underlying Figure **1** and **2** show the volume, in white, considered for the subsequent calculation with SERPENT2.

Once defined the geometric element to be analyzed, the neutron flux through its boundary surfaces (in order to characterize the relative "importance" of neutron inlet flux through each surface) has been calculated. In fact the basic idea for the analysis of this element has been to calculate the flux through each surface of the element so that these data could be used as "external" input data for the burnup calculations by the SERPENT2 code.

From the burning step point of view, the idea was to analyze the behavior of materials during irradiation (i.e. during normal operation of the reactor) and then, during the out-of-pile decay period.

As known, the ITER experimental reactor is still in the construction phase. As a consequence, the ITER operational scenario is in the planning stages. So, in order to perform our burnup calculations we can only suppose in a reasonably way the operating times of switching on/off, breaks and switching timetables. To date, one plausible hypothesis is to ignite the plasma for 450 seconds per pulse, for a total period shorter than 1800 seconds per day [75]. Three pulses (of 450 seconds each) were then hypothesized: the first one at 9 am in the morning, the others at 12 am and 3 pm respectively; this schedule should be repeated for 11 days followed by 3 days of stop, during which some analyses on materials and reactor components are planned. This means that the gap between the begin of a pulse (with a duration of 450 s, corresponding to 0.125 hours) and the begin of the following one should be about 3 hours long; as a result, the gap between the

end of a pulse and the begin of the following one should be about 2.875 hours long. SERPENT2 simulated 35000 neutrons in nps and burnup mode using the schedule reported in Table **1**.

Table 1: Burnup Schedule used in SERPENT2

Step #	Step Length [days]	Type of Step
0	0	initialization
1	0.0052083	depletion step
2	0.11979167	decay step
3	0.0052083	depletion step
4	0.11979167	decay step
5	0.0052083	depletion step
6	0.74479167	decay step
7	0.0052083	depletion step
8	0.11979167	decay step
9	0.0052083	depletion step
10	0.11979167	decay step
11	0.0052083	depletion step
12	0.74479167	decay step
13	0.0052083	depletion step
14	0.11979167	decay step
15	0.0052083	depletion step
16	0.11979167	decay step
17	0.0052083	depletion step
18	0.74479167	decay step
19	0.0052083	depletion step
20	0.11979167	decay step
21	0.0052083	depletion step
22	0.11979167	decay step
23	0.0052083	depletion step
24	0.74479167	decay step
25	0.0052083	depletion step
26	0.11979167	decay step
27	0.0052083	depletion step
28	0.11979167	decay step
29	0.0052083	depletion step
30	0.74479167	decay step
31	0.0052083	depletion step
32	0.11979167	decay step
33	0.0052083	depletion step
34	0.11979167	decay step
35	0.0052083	depletion step
36	0.74479167	decay step
37	0.0052083	depletion step
38	0.11979167	decay step
39	0.0052083	depletion step
40	0.11979167	decay step
41	0.0052083	depletion step
42	0.74479167	decay step
43	0.0052083	depletion step
44	0.11979167	decay step
45	0.0052083	depletion step
46	0.11979167	decay step
47	0.0052083	depletion step
48	0.74479167	decay step
49	0.0052083	depletion step
50	0.11979167	decay step
51	0.0052083	depletion step
52	0.11979167	decay step
53	0.0052083	depletion step
54	0.74479167	decay step
55	0.0052083	depletion step
56	0.11979167	decay step
57	0.0052083	depletion step
58	0.11979167	decay step
59	0.0052083	depletion step
60	0.74479167	decay step
61	0.0052083	depletion step
62	0.11979167	decay step
63	0.0052083	depletion step
64	0.11979167	decay step
65	0.0052083	depletion step
66	1.74479167	decay step
67	1.0	decay step
68	1.0	decay step

The material composition of the “fission waste” element to be “burned” used for the burnup calculations has been assumed on the basis of a previous study performed on the reactor ALLEGRO for burnup evaluation purposes [76]. Particularly we chose to use a fuel contained only americium and curium (with a relative isotopic composition similar to that used in the GoFastR project [63-65, 76]) because we wanted to assess the transmutation potentiality of an ITER-like reactor for those specific materials (such as some MA isotopes) heavily transmuted in fission reactors. On the contrary, in ITER, thanks to the presence of high energy neutrons (~14MeV), MA could be transmuted (almost) completely and more easily.

Looking at the MA chemical matrix, we assumed that the materials have been inserted in the “fission waste” element as oxides; as a result, the final density of the materials containing the MA is that typical of the oxides (10.579g/cm^3). The use of this chemical compound instead of more performing carbides (more

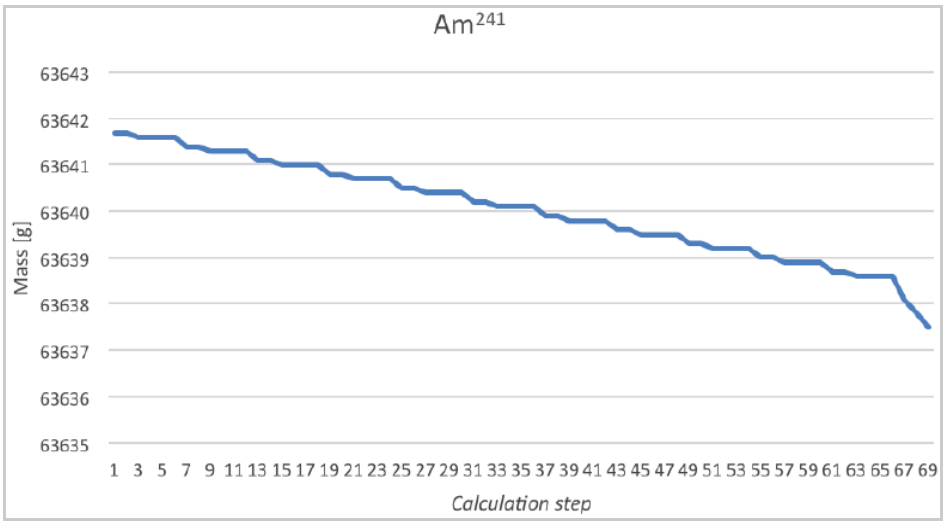

Figure 3: Am^{241} mass vs. calculation step.

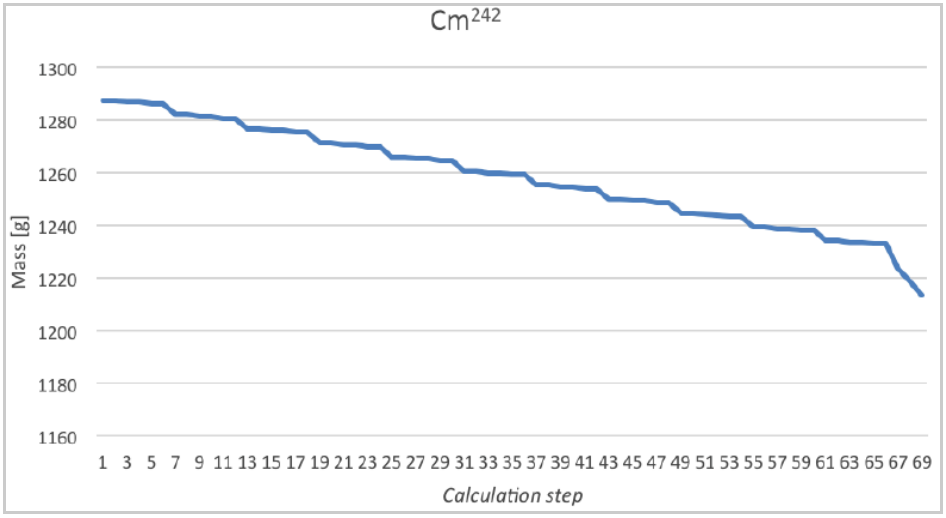

Figure 4: Cm^{242} mass vs. calculation step.

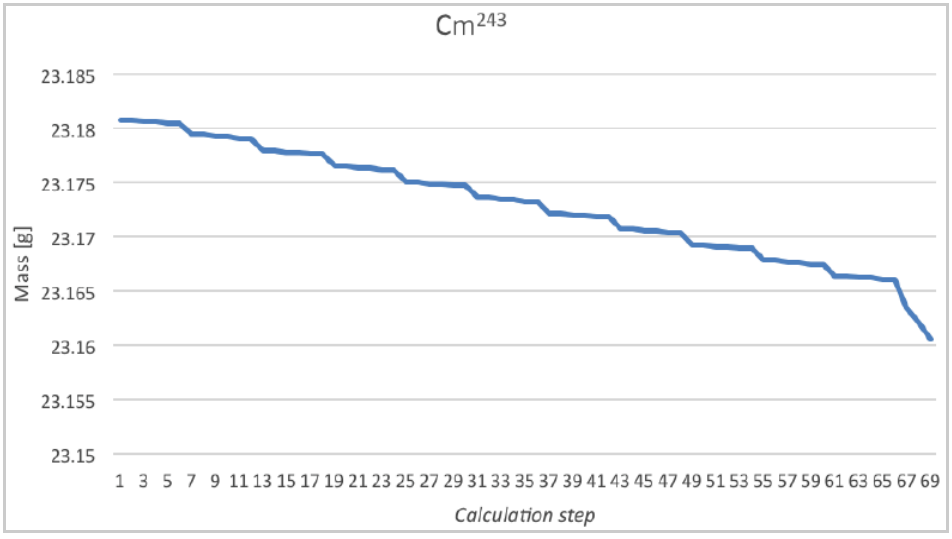

Figure 5: Cm^{243} mass vs. calculation step.

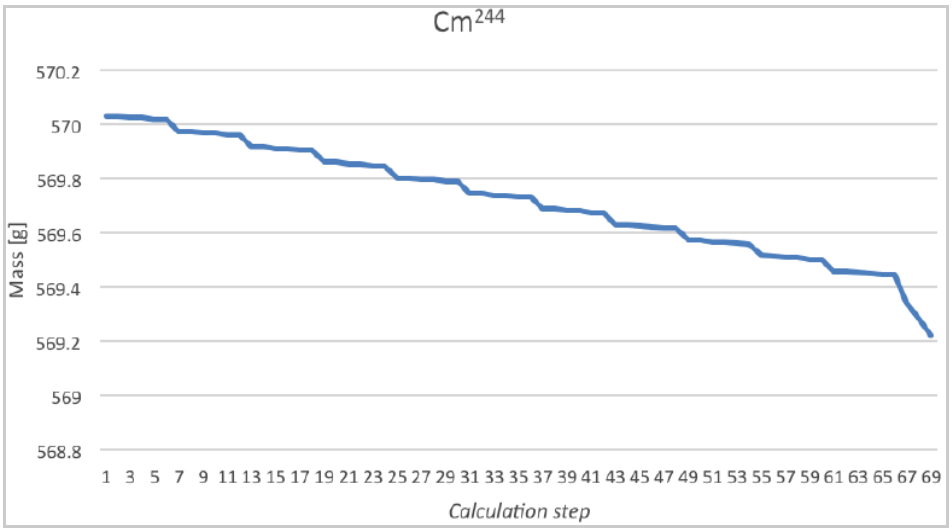

Figure 6: Cm^{244} mass vs. calculation step.

resistant to the involved high temperatures) is mainly due to the coolant used for blanket and/or FW: although different coolants have been proposed (e.g. helium), in this case (and, maybe, in the real one) it was assumed the use of pressurized (~4MPa) water (at an average temperature of ~70°C). If the carbides was used, as known, a loss of water from the coolant pipes/systems could cause a highly exothermic reaction with the same carbides. Clearly, it is not possible to accept a potential scenario like this, so we opted for the less performing but more conservative (at least from the safety point of view) oxides. The burned volume had a volume of about $1.7 \cdot 10^6$ cm^3 and a shape similar to that considered for MCNP and extracted by MCAM. As already anticipated, the "external" neutron source was obtained by mean of fluxes data through 6 surface sources.

4. RESULTS

The results obtained at the end of the burnup calculations (by SERPENT2 code) are shown in the following figures. Particularly we have simulated 14 days of burnup (11 days of "normal" operation plus 3 days of reactor stop, as better detailed in the previous paragraphs) by 69 time steps calculations; the behavior of the main Am and Cm isotopes is reported in the Figure **3-6** and summarized in Table **2**.

Looking at Figure **3-6** and at Table **2**, the Am and Cm variations due to transmutation (and not to pure decay) seem very limited, but two fundamental aspects have to be considered:

I. The timing of each pulse is extremely reduced - The daily duration of the pulses is 1350 seconds equal to only 0.015625 days. Multiplying this quantity for 11 days of reactor operation, we obtain 0.171875 days. Therefore, the irradiation time is short and this is reflected clearly in the MA transmutation. In future, however, once the fusion technology will be more assessed, it should be reasonable to conjecture a quite longer irradiation time.

Table 2: Main MA Isotopes Trend after the Transmutation Process

Elements	Initial value	Trend	Variation	Variation [%]
Am^{241}	~ 63642 g	decreasing	~ 4 g	~ - 0.01%
Am^{243}	~ 10000 g	~constant	negligible	negligible
Cm^{242}	~ 1280 g	decreasing	~ 60 g	~ - 4.7%
Cm^{243}	~ 23.18 g	decreasing	~ 0.02 g	~ - 0.086%
Cm^{244}	~ 570 g	decreasing	~ 0.6 g	~ - 0.1%
Cm^{245}	~ 11.1062 g	increasing	~ 0.0006 g	~ 0.01%
Cm^{246}	~ 0.10849 g	increasing	~ 0.000003 g	negligible

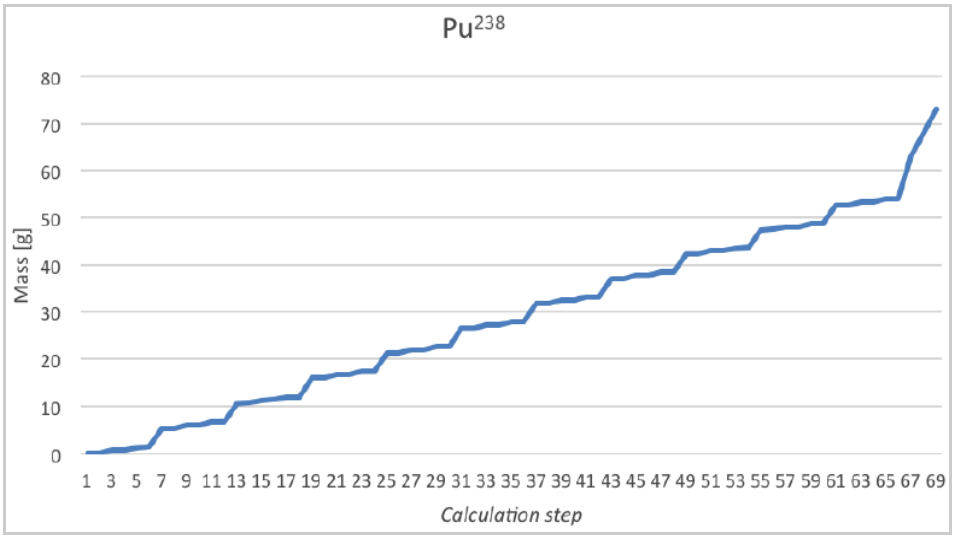

Figure 7: Pu^{238} mass vs. calculation step.

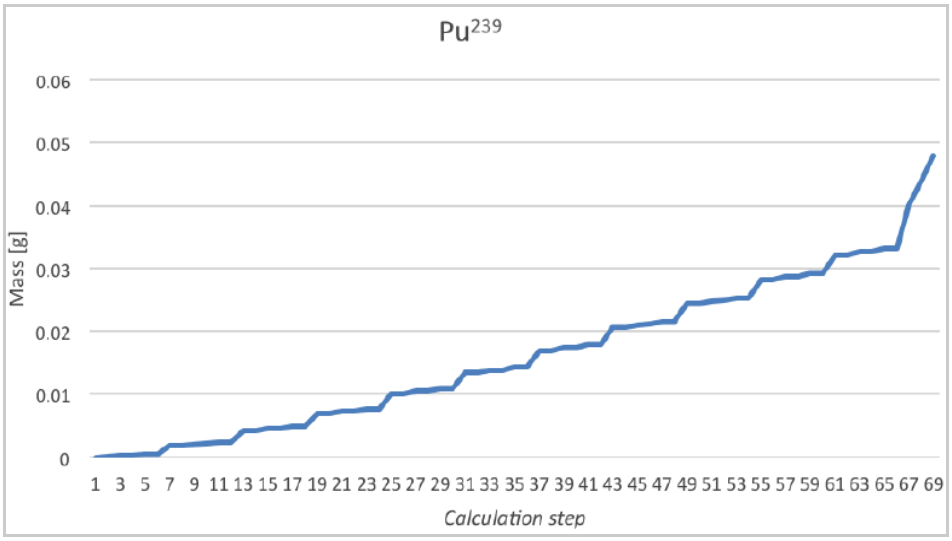

Figure 8: Pu^{239} mass vs. calculation step.

II. The single element inserted into the reactor blanket - The goal of the study is in fact, to demonstrate the *feasibility* of this process, so we consider only one element inside the blanket. Obviously as a future development, when a more mature and proven technology will be available, it could be possible to load many "modified" sectors/modules simultaneously, so increasing the MA transmutation rate.

Another important issue to be considered is the production of uranium and plutonium, absent at the beginning of the irradiation period: as obvious, the isotopes of these element increase; here, as examples, only three isotopes of plutonium are shown in Figure **7**, **8** and **9**. The amount of uranium that is created during the process is summarized in the following Table **3**, while the plutonium production is reported in Table **4**. While the production of uranium is very modest, the plutonium increases more substantially: the final amounts of the Pu isotopes are not negligible (as, instead, it is in the case of uranium). The Pu^{238} build-up is mainly due to the large amount of Am^{241} [75]: as known, following a neutron capture, this latter is transmuted into Am^{242}, which is a short life isotope (half-life of 16.2 hours); the 82.7% of Am^{242} decays[1] into Cm^{242}, while the remaining 17.3% transmutes (by an electron capture) into Pu^{242}; finally Cm^{242} (characterized by a half-life of about 160 days),

[1] β-decay

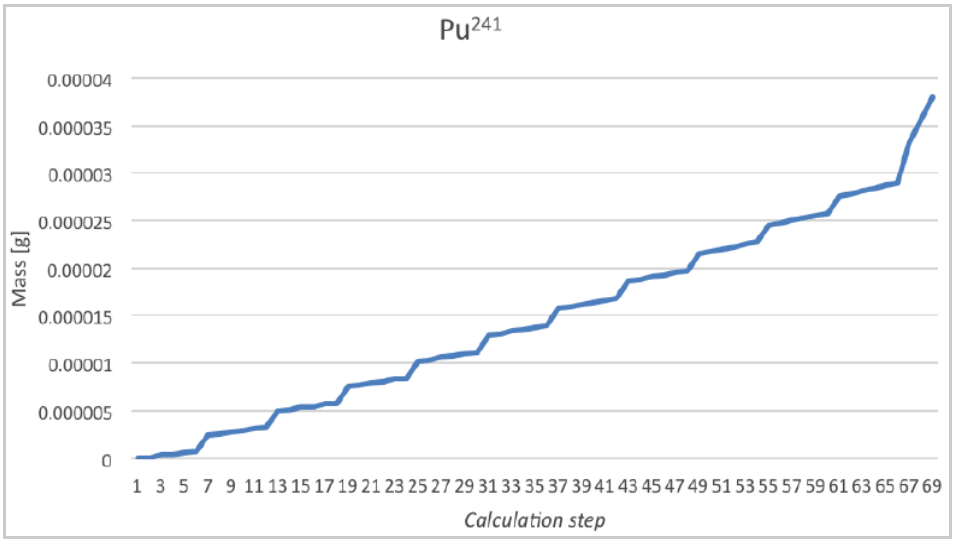

Figure 9: Pu^{241} mass vs. calculation step.

decays[2] into Pu^{238}. So, in the end, it is true that the amount of plutonium increases, but the isotope which largely increases, is characterized by a significant radiotoxicity for a time interval shorter than that typical of other MA [75]. This is a key issue, because it is precisely one of the aim of the present study: indeed the goal is not to completely "destroy" the initial radiotoxicity inventory, but rather to transform the initial long-lived radionuclides in others, characterized by a shorter half-life.

Table 3: U Isotopes Build-up after Irradiation

U^{232}	~ 3E-11 g
U^{233}	~ 2.5E-9 g
U^{234}	~ 0,012 g
U^{235}	~ 2.5E-8 g
U^{236}	~ 0.0000016 g
U^{238}	~ 7E-10 g

Table 4: Pu Isotopes Build-up after Irradiation

Pu^{238}	~ 70 g
Pu^{239}	~ 0.05 g
Pu^{240}	~ 0.08 g
Pu^{241}	~ 0.00004 g
Pu^{242}	~ 0.035 g
Pu^{243}	~ 0 g
Pu^{244}	~ 0.000012 g

[2] α-decay

Finally Figure **10** shows the 69-groups spectra at begin of cycle: it is important to note that there is a peak between 100eV and 100keV; this finding is very interesting, because in this range it is possible to have a very good MA transmutation. In fact, as known, some MA cross sections (e.g. those related to Am^{241} and Cm^{244}) are typically higher for that neutrons energy range.

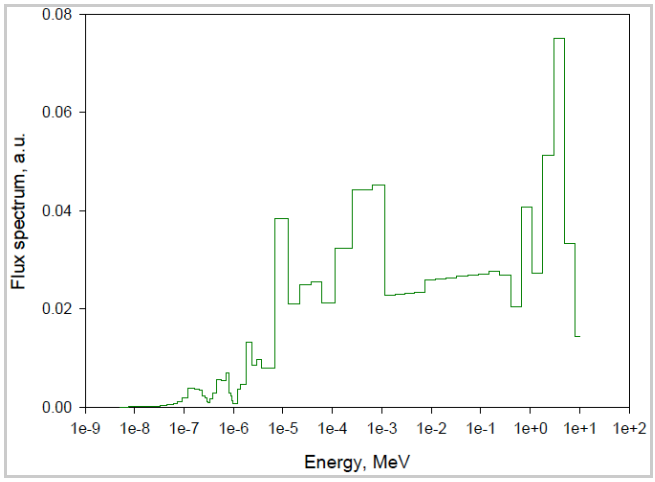

Figure 10: 69-groups spectrum at begin of cycle.

In general, we can conclude that the MA mass inventory trend is coherent with the spectra: starting from a 14MeV neutrons source from the fusion plasma (which is a typical value for a thermonuclear fusion reactor), it is possible to obtain neutrons fluxes suitable for the MA transmutation.

However, it is interesting to carry out some additional considerations related to the reactor operating time: it has to be recalled that ITER will be an

experimental plant and, as previously mentioned, will work in a pulsed mode; the (pre-)"commercial" nuclear fusion reactors (DEMO and later) obviously will work for much longer periods of time, possibly in a continuous mode (in order to, among the others, increase the energy production rate and the compatibility with the electric grids). Starting from this considerations, it could be possible to assess (although at a very preliminary level) the hybrid reactors transmutation potentialities in a more realistic way.

As already anticipated, the daily duration of the considered pulses is only 1350 seconds (i.e. around 0.015625 days); multiplying this quantity for 11 days of the reactor operation (plus 3 days of subsequent decay), we obtained 0.171875 days of total irradiation. On the basis of the previous considerations, if we consider a (pre-)"commercial" (DEMO-like) nuclear fusion reactor, and assume a linear transmutation behavior[3], we have to multiply 14 days (corresponding to 0.171875 days of "burning pulses") by a factor of 26, in order to have a reactor operation equal to a (reasonable) time (i.e. about 1 years); under this hypothesis, as a first rough approximation, also the MA final quantities have to be multiplied by a factor around 26. This would provide (for a single "fission waste" loaded element), for example, the transmutation of more than 100 g of Am^{241}. Similarly, if you multiply by a factor of 650 (that is to have an equivalent reactor operating time in a pulsed regime equal to about 25 years), it is possible to obtain (for a single "fission waste" loaded element) a decrease of about 2.6 kg of the Am mass initially present. Therefore, if we consider those assumptions, the quantity of "burned" actinides would be much higher, as better detailed in Table **5**[4].

Table 5: MA Transmutation Performances vs. Equivalent Reactor Operating Time

Nuclide	Reference Case (14 days)	~1 Years	~25 Years
Am^{241}	~ 4 g	~ 104 g	~ 2600 g
Cm^{242}	~ 60 g	~ 1560 g	~ 39000 g
Cm^{243}	~ 0.02 g	~ 0.52 g	~ 13 g
Cm^{244}	~ 0.6 g	~ 15.6 g	~ 390 g
Cm^{245}	~ 0.0006 g	~ 0.0156 g	~ 0.39 g
Cm^{246}	~ 0.000003 g	~ 0.000078 g	~ 0.00195 g

[3] Obviously this preliminary rough assumption has to be further refined on the basis of the future developments of the present research, taking also into account separately the decay and transmutation contributions.

[4] Again, this preliminary rough assumption has to be further refined on the basis of the future developments of the present research, taking also into account separately the decay and transmutation contributions.

However it is obvious that these extensions in reactor operating time would increase the build-up of unwanted materials (Table **6**): to give an example to obtain the amount of Pu^{238} after 1 year the original amount should be multiplied by 26 thus obtaining a quantity equal to around 1.8kg. Nevertheless, as already highlighted in the previous paragraphs, Pu^{238}, in addition to being usable in various fields (including the aerospace industry), is not characterized by a long term high radiotoxicity level, so its storage would certainly be safer and less expensive (if compared with other MA).

Table 6: Pu Transmutation Performances vs. Equivalent Reactor Operating Time

Nuclide	Reference Case (14 days)	~1 year	~ 25 years
Pu^{238}	~ 70 g	~ 1820 g	~ 45500 g
Pu^{239}	~ 0.05 g	~ 1.3 g	~ 32.5 g
Pu^{240}	~ 0,08 g	~ 2.08 g	~ 52 g
Pu^{241}	~ 0.00004 g	~ 0.00104 g	~ 0.026 g
Pu^{242}	~ 0.035 g	~ 0.91 g	~ 22.75 g
Pu^{244}	~ 0.000012 g	~ 0.000312 g	~ 0.078 g

Looking at the other Pu isotopes, although they constitute a potential long term problem, they could be (relatively) easily transmuted also in fast (fission) reactors (e.g. [50, 51, 53, 54, 58, 63].

5. CONCLUSIONS

As already found in previous publications, also our preliminary calculations and results confirm the feasibility and the potentiality of transmutation in hybrid fusion-fission reactors. In fact, it was found a decrease (although relatively limited) in many actinides masses and the build-up of nuclides characterized by reduced levels (more than an order of magnitude) in long term potential hazard (in term of radiotoxicity, etc.), if compared to the starting materials. In this way, the long term storage of nuclear waste would be cheaper and, above all, safer (since such wastes could be potentially dangerous only for a time interval lower than in the case in which there had been no transmutation).

The obtained results are interesting, but it should also be remarked that this paper has to be considered only as a first preliminary step, useful mainly to demonstrate the feasibility of the process in "realistic" fusion plants. In fact, only when the nuclear fusion

technology (including materials, blanket configuration, etc.) will be fully developed and the amount of high-level radioactive materials from fission reactors fleet to be burned inside fusion reactors will be precisely defined, we could be able to develop in detail a realistic hybrid fission-fusion "fuel cycle".

Nevertheless if we should consider a (pre-) "commercial" (DEMO-like) nuclear fusion reactor with a longer irradiation time (even remaining in a pulsed regime) and the presence of many (instead of a single one) "fission waste" elements loaded, we could assume, although as a first (very) rough approximation, to have a substantial decrease in MA quantities: the quantity of actinides "burned" would indeed be, under these hypotheses, much higher.

ACKNOWLEDGEMENTS

This work was carried out using an adaptation of the B-lite MCNP model which was developed as a collaborative effort between the FDS Team of ASIPP China, University of Wisconsin-Madison, ENEA Frascati, CCFE UK, JAEA Naka, and the ITER Organization. We want to thank all these organizations.

We want also to thank the FDS Team members (China) for their precious collaboration in MCAM installation, configuration and application.

Our thank goes to Dr. J. Leppänen of VTT for his continuous support in SERPENT configuration and documentation supply.

Finally, special thanks go to Ing. W. Borreani and Ing. A. Muzzioli of GeNERG-DIME/TEC (University of Genova) for their help in computational issues, Dr. Ing. R. Marotta of Nucleco S.p.A. and Ing. M. Dellabiancia of RC-Řež for their suggestions.

LIST OF ACRONYMS

ADS	Accelerator-Driven System
ALLEGRO	Gas-cooled Fast Reactor Demonstrator
ASIPPC	Institute of Plasma Physics Chinese Academy of Science
CCFE	Culham Centre for Fusion Energy
DEMO	Demonstration fusion power plant
ENEA	Italian National Agency for New Technologies, Energy and Sustainable Economic Development
FDS	Advanced Nuclear Energy Research team (China)
FENDL	Fusion Evaluated Nuclear Library
FW	First Wall
ICF	Inertial Confinement Fusion
ITER	International Thermonuclear Experimental Reactor
JAEA	Japan Atomic Energy Agency
JEFF	Joint Evaluated Fission and Fusion File
LWR	Low Water Reactor
MA	Minor Actinide
MC	Monte Carlo
MCAM	Monte Carlo Automatic Modeling
MCF	Magnetic Confinement Fusion
MCNP	Monte Carlo N-Particle
OTTO	Once Through Then Out
SNF	Spent Nuclear Fuel
TRU	Transuranic element

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Received on 26-11-2015 Accepted on 28-12-2015 Published on 31-12-2015

DOI: http://dx.doi.org/10.15377/2409-5818.2015.02.02.4