ANALYSIS OF THE TRANSMUTATIONAL CAPABILITIES OF A NOVEL MOLTEN SALT REACTOR

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ABSTRACT

Nowadays the molten salt reactor (MSR) concept seems to revive as one of the most promising systems for the realization of transmutation. In the molten salt reactors and subcritical systems the fuel and material to be transmuted circulate dissolved in some molten salt. The main advantage of this reactor type is the possibility of the continuous feed and reprocessing of the fuel.

In the present paper a novel molten salt reactor concept is introduced and its transmutational capabilities are studied. The goal is the development of a transmutational technique along with a device implementing it, which yield higher transmutational efficiencies than that of the known procedures and thus result in radioactive waste whose load on the environment is reduced both in magnitude and time length. The procedure is the multi-step time-scheduled transmutation, in which transformation is done in several consecutive steps of different neutron flux and spectrum. In the new MSR concept, named "multi-region" MSR (MRMSR), the primary circuit is made up of a few separate loops, in which salt-fuel mixtures of different compositions are circulated. The loop sections constituting the core region are only neutronically and thermally coupled. This new concept makes possible the utilization of the spatial dependence of the spectrum as well as the advantageous features of liquid fuel such as the possibility of continuous chemical processing etc.

In order to compare a "conventional" MSR and a proposed MRMSR in terms of efficiency, preliminary calculational results are shown. Further calculations in order to find the optimal implementation of this new concept and to emphasize its other advantageous features are going on.

1. INTRODUCTION

One of the arguments most frequently brought up by the opponents of the utilization of nuclear energy is the requirement that the radioactive waste and the long-lived radioisotopes accumulated in the spent fuel should be isolated from the biosphere for a very long time. In the past 40 years, deep geological repositories have been considered as the primary method of nuclear waste final disposal. However, concerns about this method retarded the application of geological repositories. There is still no consensus whether the nuclear waste processing with conventional methods and final geological disposal are acceptable practice for humanity. A solution, which is fundamentally different from the one mentioned above and new in principle, is the elimination of long-lived actinides (plutonium isotopes and minor actinides) and long-lived fission products by transforming (transmuting) them into short-lived or stable nuclei. In this manner the amount of radioactivity present in the waste can be decreased and the time required for the radioactivity to decay to an acceptable level can be shortened radically. Although this would not make final storage (typically geological) unnecessary, the conditions of storage can be dramatically improved as well as the required isolation period can be reduced by several orders of magnitude. The storage facilities would thus be easier to design and operate and transmutation may help to form a more convincing image for the inhabitants. These issues are dealt with in our earlier work [1], while certain aspects are analyzed in papers [2,3].

The high neutron flux required for transmutation can be realized in nuclear installations. These may be conventional thermal and fast reactors, furthermore dedicated devices, namely thermal and fast reactors and accelerator driven subcritical systems (ADSs), which are specifically designed for this purpose. Some of the most promising systems are the molten salt reactors and subcritical systems, in which the fuel and material to be transmuted circulate, dissolved in some molten salt. In the present paper this transmutational device, as well as recommendations for the improvement are discussed in detail.

2. ANALYSIS OF THE MOLTEN SALT REACTOR CONCEPT IN RESPECT TO TRANSMUTATION

The Molten Salt Reactor (MSR) Program started in the US in 1956. At the end of the 50s and beginning of 60s the interest towards breeder reactors rose and support to the MSR program could be obtained by verifying the breeding capability. The design of the facility called MSR Experiment started in 1960 [4]. The reactor of 8 MW maximum thermal power went critical in 1965 and was operating almost continuously until December 1969, when it was shut down due to the lack of financial support. Later the plans of an installation with 1000 MW_e called MS Breeder Reactor Experiment (MSBR) were completed, but finally, in 1976 the entire molten salt reactor program was stopped due to financial reasons.

The idea of molten salt reactors was brought up again in the 80s, when it was proposed, together with the new concept of accelerator driven molten salt subcritical system, for transmutation purposes. Although the conception of the latter system was worked out at the Los Alamos National Laboratory (LANL), different concepts have become known [5-9] since then.

Considering the above mentioned and other publications, it can be stated that the molten salt reactor has several advantages over the conventional solid fuel heterogeneous systems from the viewpoint of transmutation:

- The continuous removal of fission products from the molten salt by continuous reprocessing reduces the neutron absorption by fission products.
- The molten salt reactor can be designed with a negative temperature coefficient, not depending on the fuel content; therefore overheating of the fuel leads to a loss of fuel from the core by expansion.
- The lack of fuel fabrication makes the fuel cycle more simple and flexible.
- It is much easier to reprocess liquid fuel than solid.
- High burnup is achievable, since in the case of liquid fuel the radiation damage is not a harmful effect.

On the other hand, the known molten salt systems have their disadvantages too:

- Continuous feed of fuel is frequently mentioned as an advantageous feature of these reactors but this is not the case considering transmutation, as the continuous mixing of the fresh and irradiated fuel worsens the transmutational efficiency.

- Since the molten salt containing the isotopes to be transmuted constitutes one single space in the reactor or subcritical system, it is impossible to utilize the advantages due to the spatial distribution of neutron spectrum and neutron flux in order to improve the transmutational efficiency. In a heterogeneous system the above-mentioned advantages can be easily utilized by appropriately arranging the fuel elements.
- A single space molten salt reactor makes it possible to implement only a simple time scheduling of transmutation enabling to modify merely the duration that elapses between the removal of the spent fuel from the nuclear reactor to the start of the transmutation.

As a consequence of the above written facts, neither the molten salt reactors nor the accelerator driven molten salt subcritical systems are capable of achieving as high transmutational efficiency and as effective decrease in radiotoxicity as they would be capable of by the application of an optimal strategy. However, the optimal strategies require modifications to the known solutions of the molten salt systems.

3. THE MULTI-REGION MOLTEN SALT REACTOR AND SUBCRITICAL SYSTEM

The goal is the development of a transmutational technique along with a device implementing it, which yield higher transmutational efficiencies than that of the known procedures and thus results in radioactive waste whose load on the environment is reduced both in magnitude and time length.

The procedure is the multi-step transmutation, in which the transformation is carried out in several consecutive steps of different neutron flux and spectrum. In order to implement this, a multi-region transmutational device, i.e. nuclear reactor or subcritical system is proposed, in which several separate flow-through irradiation rooms are formed with various neutron spectra and fluxes. The characteristics of these rooms may differ significantly in the case of ADSs, particularly if certain regions are filled with some moderating material.

The fission products can typically be transformed by capturing thermal neutrons at adequate efficiency. For this purpose such transmutational devices are necessary, in which (in the total or part of the volume) the thermal neutron flux is high. This can be achieved in an ADS or in a part of it, practically an external region, which is surrounded by a reflector of suitable thickness from the outer surface.

As a result of neutron capture, in most cases actinides transform into isotopes with even longer half-lifes and larger mass numbers, which is an undesired change in the present case. Here the neutron induced fission reactions lead to beneficial changes. Certain actinides (Pu-238, Pu-239, Pu-241, Am-242m, Cm-243 and Cm-245) are fissile upon interaction with thermal neutrons (the cross sections are large), but, unfortunately, their neutron capture cross section is also large in this energy region, which results in the production of a significant amount of heavier actinides. Upon the effect of higher energy neutrons all the actinides are fissile, while the ratio of fission to capture cross section is far greater than in the case of thermal neutron induced nuclear reactions. In view of these facts, those devices are advantageous for actinides, in or in part of which the fast neutron flux is high. Such candidate might be e.g. the innermost region (the region closest to the proton beam or target if any) of an ADS.

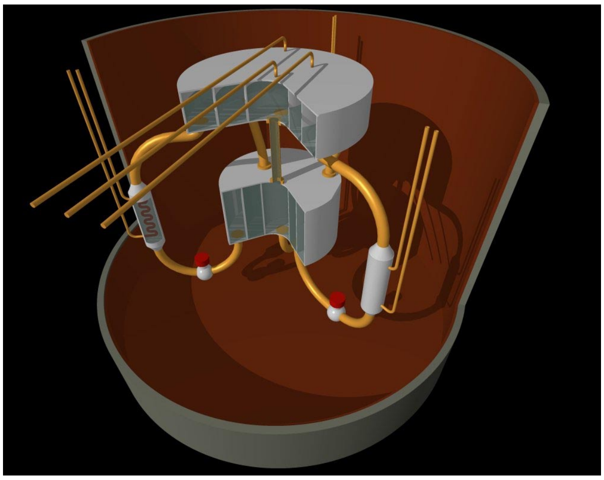

Figure 1. Scheme of a three-region molten salt reactor

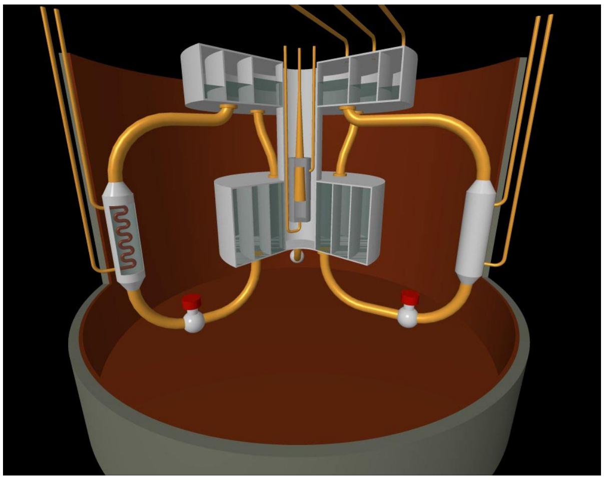

Figure 2. Scheme of a three-region accelerator driven molten salt subcritical system

According to the above considerations, in an optimal transmutational strategy it may be useful to apply irradiation steps of different neutron spectra and combine these steps corresponding to an elaborate schedule, especially in the case of certain isotopes. For this purpose a multi-region transmutational device, the multi-region molten salt reactor (MRMSR) or the multi-region accelerator driven molten salt subcritical system (MRADMS) can be an advantageous candidate[10].

Fig. 1. shows the scheme of a multi-region molten salt reactor, the example being a three-region one. In this example the regions are concentric cylindrical rings. The warmed up molten salt containing the already transmuted and to be transmuted isotopes goes to the upper expansion tank and then via the downcomer to the heat exchanger, where it transfers its heat to the secondary fluid (which may also be some molten salt or e.g. He gas). The resulting cooled down molten salt is pushed back to the first region by the circulating pump via the inlet pipe. The dimensions of the upper expansion tank are such that the level of the molten salt should remain above the prescribed minimal value in the case of the highest potentially occurring density and should not exceed the maximum value when the density is the lowest. The volatile and gaseous fission products getting out of the molten salt are carried off via the gas off-take pipe along with the He gas present in the expansion tank. The three reactor regions are separated using partitioning walls. In the example shown, the upper expansion tanks are also separated using partitioning walls. The entire device outlined is located in a shaft surrounded by a shell. The dimensions of the shaft must be chosen so that, in case the total amount of molten salt present in the system got out, the molten salt accumulated at the bottom of the shaft be subcritical to a prescribed value under any conceivable circumstance.

In the example shown in Fig. 1. the implementation does not contain moderating material in any of the regions and therefore the neutron spectrum is fast in all of them. If a region with thermal spectrum is also necessary, the moderator, which is normally graphite, is usually contained in the outer region. In this case it is practical to surround the reactor with a graphite reflector.

Fig. 2. shows the side view of an accelerator driven three-region molten salt subcritical system. The device is driven by a beam of particles from an accelerator, either with the application of a target core or the target being the molten salt material of the innermost region. The construction is similar to that shown in Fig. 1. with the difference that the innermost region is not cylindrical shaped but rather a cylindrical ring inside which, i.e. around the axis of the system, the beam is supplied to the target. The advantage of this method is that significantly harder neutron spectra can be achieved (especially in the innermost region) than in the same region of the reactor construction shown in Fig. 1.

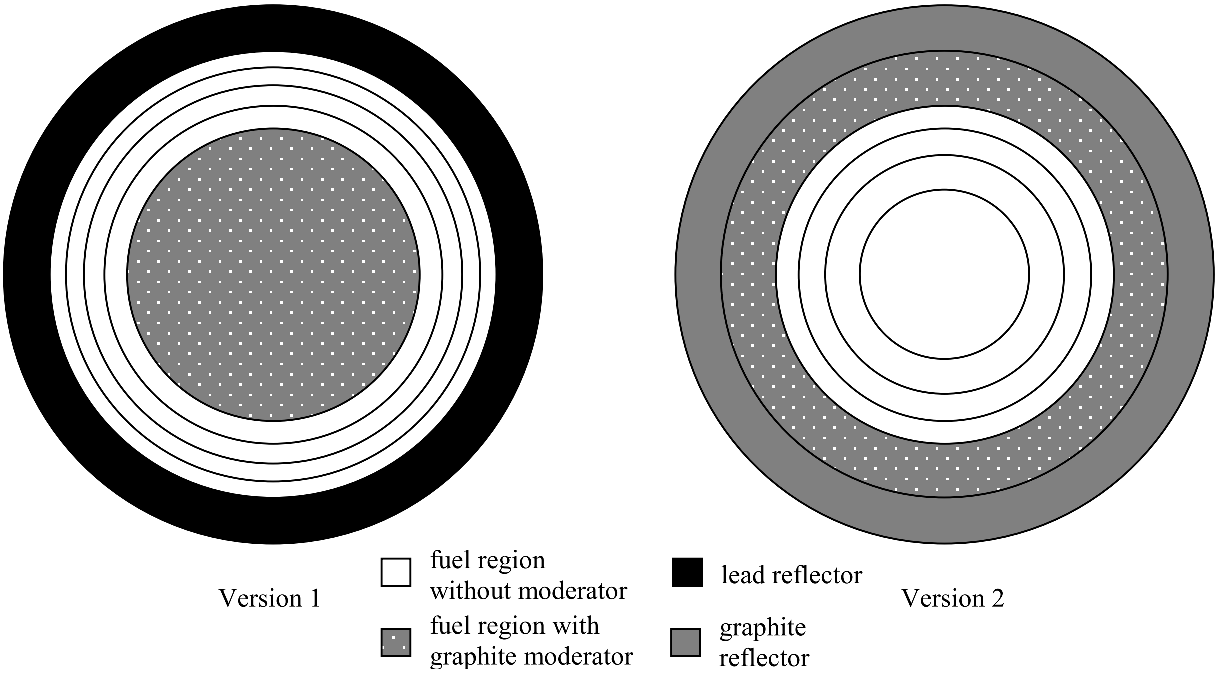

Figure 3. Core maps of two possible version of MRMSR

3. CALCULATIONS

In order to compare a "conventional" MSR and a proposed MRMSR in terms of efficiency, preliminary calcualtional results are shown in this section. The main characteristics of the devices are as follows.

3.1 MOLTEN SALT REACTORS

In the case of the MSR the molten salt is contained in a cylinder, whose diameter and height are both 4 m, surrounded by a steel reflector in order to achieve a harder spectrum. The thermal power of the reactor was supposed to be 2500 MW_{th} (about 1000 MW_e), the average temperature of the salt is 940 K, while that of the reflector is 800 K. The salt composition is 30.04 mol% BeF_2 + 69.62 mol% LiF + 0.34 mol% $(Pu+MA)F_3$. This is far enough from the solubility limit, which is approximately 1% for $(Pu+MA)F_3$. The starting composition of the actinide isotopes corresponds to that found in the fuel of a VVER-440 at 42 MWday/kgU burnup (without U). In order to constantly maintain this salt composition, the burnt out actinides are continuously supplied in the same composition. In this manner the total mass of the actinide isotopes remains constant, only their ratio to each other changes. The evolving fission products are continuously removed using a chemical process[11]. After a given period the whole actinide content of the core is discharged and sent to final disposal. In this preliminary study we supposed 20 years as irradiation time.

3.2 MULTI-REGION MOLTEN SALT REACTORS

In the case of the MRMSR the 4 m high cylindrical core is divided into 5 concentric cylindrical rings of equal volume. The overall volume and the power is the same as in the case of MSR. The initial salt composition is 32.17 mol% BeF_2 + 67.13 mol% LiF + 0.7 mol% $(Pu+MA)F_3$, which still represents a safe distance from the solubility limit. The composition of actinides at loading is the same as with MSR. This goes into the outer region. The 1 year burnt out goes to the 2nd, the 2 year to the 3rd etc. The spent fuel removed from the innermost region is treated as waste and sent to final disposal. There is no continuous feed, but the fission products are removed continuously in this case too. Two different core layout was investigated (Fig. 3.). In the first one the thermal zone is formed in the innermost region. The moderator is graphite and the molten salt circulates cylindrical channels with a diameter of 6.06 cm which form a triangular lattice with a pitch of 10 cm. The volume ratio of moderator to salt is 2:1. In this case the reflector material is lead in order to avoid the termalization in the outer regions. In the other layout the thermal region with the same lattice is formed in the outermost region and the reflector is 45 cm thick graphite, which is also favorable due to the more thermal spectrum of the external region.

3.3 METHODS AND CODES APPLIED

The calculations were performed with the aid of the codes of the SCALE system [12]. The codes BONAMI and NITAWL produce the AMPX working library, which contains resonance self-shielded problem dependent cross sections. This is used by the Monte Carlo code KENO-VI for criticality calculations. The XSDRNPM 1-D discrete ordinates code produces a weighted AMPX library and calculates the flux, spectrum and k_{eff}. The COUPLE code generates ORIGEN working libraries from the data present in the weighted library. The ORIGEN-S point-depletion code is suitable for calculating the burnup of liquid fuel reactors since, besides the nuclear reactions, it is also capable of modeling the continuous feed and blending of different partial flows. In the case of the MSR the burnup was calculated in one-year intervals. Based on the composition data obtained from ORIGEN, the spectrum and cross sections were calculated again at the end of each interval and the burnup of the next interval was calculated using the new data.

In the case of the MRMSR an iterative burnup calculation scheme was applied. At first the salt composition was determined in every region at five points of the one year long cycle using the ORIGEN with the application of an initial library. Then from these compositions the required libraries were created and the burnup calculation was repeated to obtain a better estimation of compositions. This iterative process was applied until the difference between two consecutive steps became sufficiently small.

3.4 RESULTS

According to the calculations, the five regions of the two different MRMSR designs will have the neutron spectra shown in Figs. 4. and 5. (normalized to one fission neutron). One can see from the figures that due to the thermalized regions the spatial dependence of the neutron spectrum is very strong. It can be observed that in the first case (Fig. 4.), when the thermalized region contains the most burned up fuel, the fluxes of the regions are very well balanced and a very high thermal flux can be achieved. In the other case (Fig. 5.), the fresh fuel is charged into the thermalized region, which results in a harder spectrum (due to the high Pu content) and great differences in the power ratio (the power in the outermost region is almost an order of magnitude higher than in the others). In the inner region the very low fissile material content causes an unexpected thermal spectrum due to the presence of Be as moderator. It can be concluded that the division into regions is expected to introduce some improvement. The calculations aimed at the efficiency verified these assumptions (especially regarding radiotoxicity).

Table I. shows the number of VVER-440 reactors which can be “served” by an MSR and an MRMSR of 1000 MWe (with respect to the transmutation of actinides).

Table I. Number of VVER-440 reactors, which can be served by a 1000 MW_e molten salt reactor

Type of transmutational reactor	Initial load	Yearly load
MSR	15.5	5.8
MRMSR	~15.5	6.2

Figs. 6. and 7. show the behavior of radiotoxicity for different cases. OTC (once-through cycle) means the case without transmutation. From Fig. 6. one can draw the conclusion that in terms of the total radiotoxicity of all the actinides the largest and smallest radiotoxicity values are obtained without transmutation and with the application of the MRMSR, respectively. Concerning the relative radiotoxicity of the waste originated in the MRMSR, Fig. 7. shows that this is governed by the Cm isotopes. This is comprehensible since the transmutational devices transform part of the plutonium isotopes into higher actinides, thus raising the amount of actinides. On the other hand, the results reflect the limited transmutational ability of the critical MRMSRs. This is partly due to the comparatively soft neutron spectrum (see Fig. 4. and 5.), where the fission cross section of some of the actinides is low and the σ_f/σ_c ratio is very low for all them. Nevertheless, examining Fig. 6 it can be stated that the MRMSR seems a better transmutational device than the MSR. This is backed up by the numerical figures listed in Table II., in which the initial (corresponding to t=0) relative residual hazard values (which refers to the integrated risk of the whole storage time) [1,2] are summarized for different options.

Table II. Initial relative residual hazard for different cases, %

Transmutational device or method	Relaiv residual hazard
OTC	100.0
MSR (20 years)	15.19
MRMSR 1	6.38
MRMSR 2	7.13

The above shown results obtained using five regions are not the results obtained after optimization. Therefore, the results obtained for the MRMSR cannot be taken as optimal. It is also seen from the results that it may be useful to connect the transmutation going on in the MSR and in the MRMSR as a series. This fact raises the necessity of full-system performance analyses, discussed in more detail in [3].

CONCLUSIONS

The paper analyses possibilities to improve the transmutation aimed molten salt reactor and accelerator driven molten salt subcritical system. It is concluded that the efficiency of transmutation can be increased and waste with lower radiotoxicity can be produced with the application of a multi-step transmutational method, which takes into account the spatial changes in the neutron flux and spectrum of the transmutational device. This method can be implemented in the proposed multi-region molten salt reactor and particularly in the multi-region molten salt subcritical system. Preliminary calculational results for systems, which have not been optimized yet, are presented. This and the transmutation in the multi-region molten salt subcritical system are considered as the next targets of our investigation. The possibility to connect the various transmutational devices into a tandem should also be examined, which requires full-system performance assessment.

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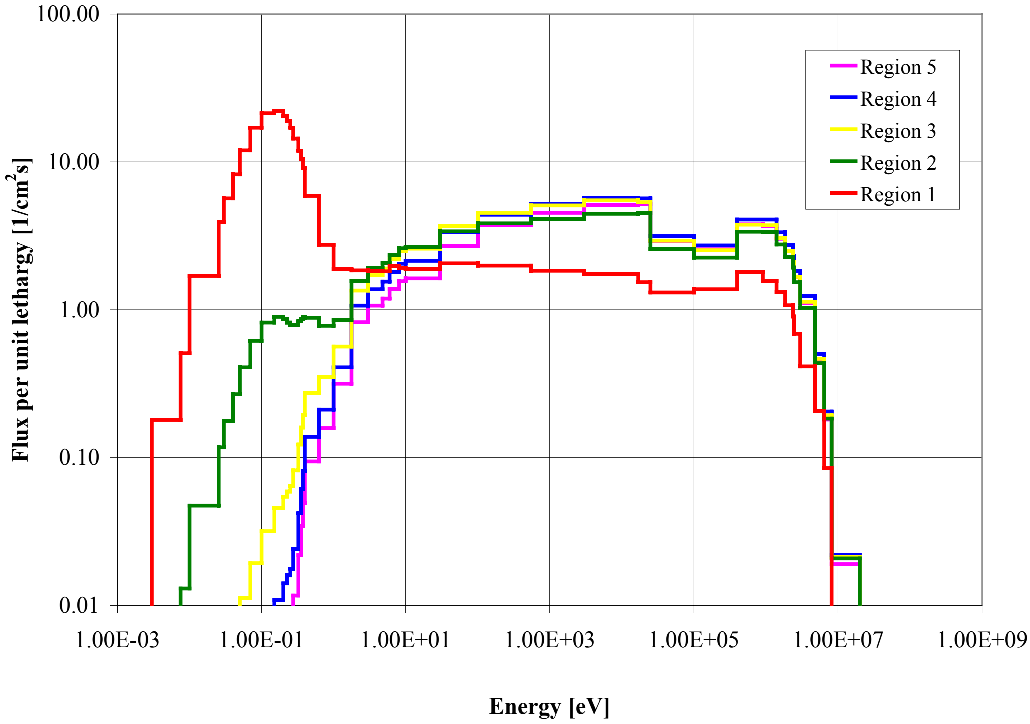

Figure 4. Spectra in the different regions of MRMSR version 1

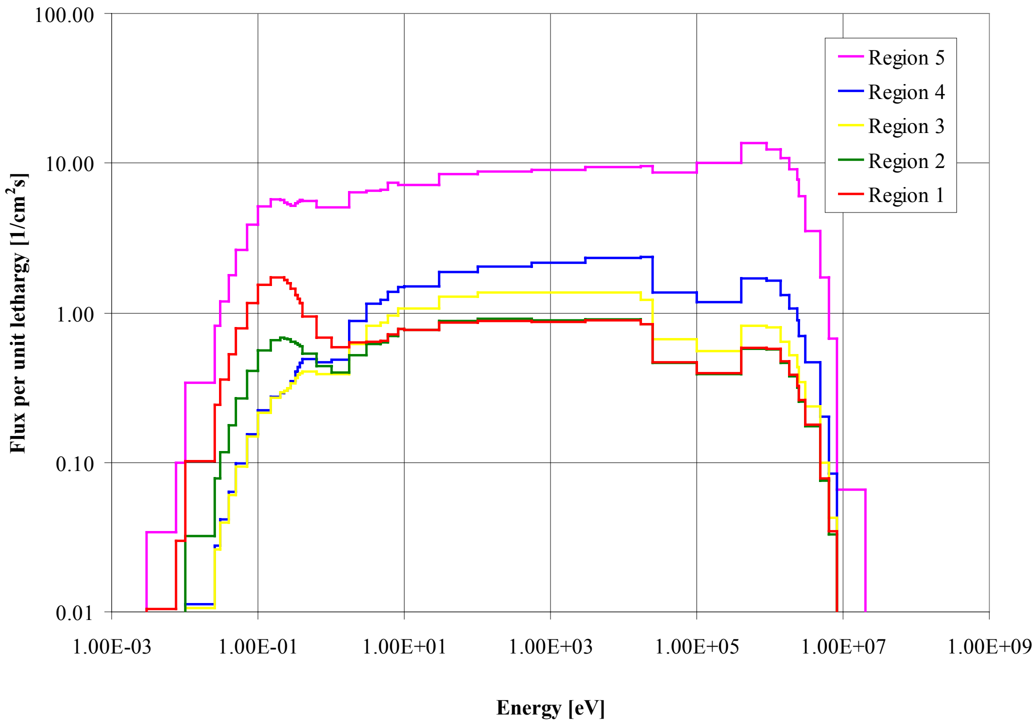

Figure 5. Spectra in the different regions of MRMSR version 2

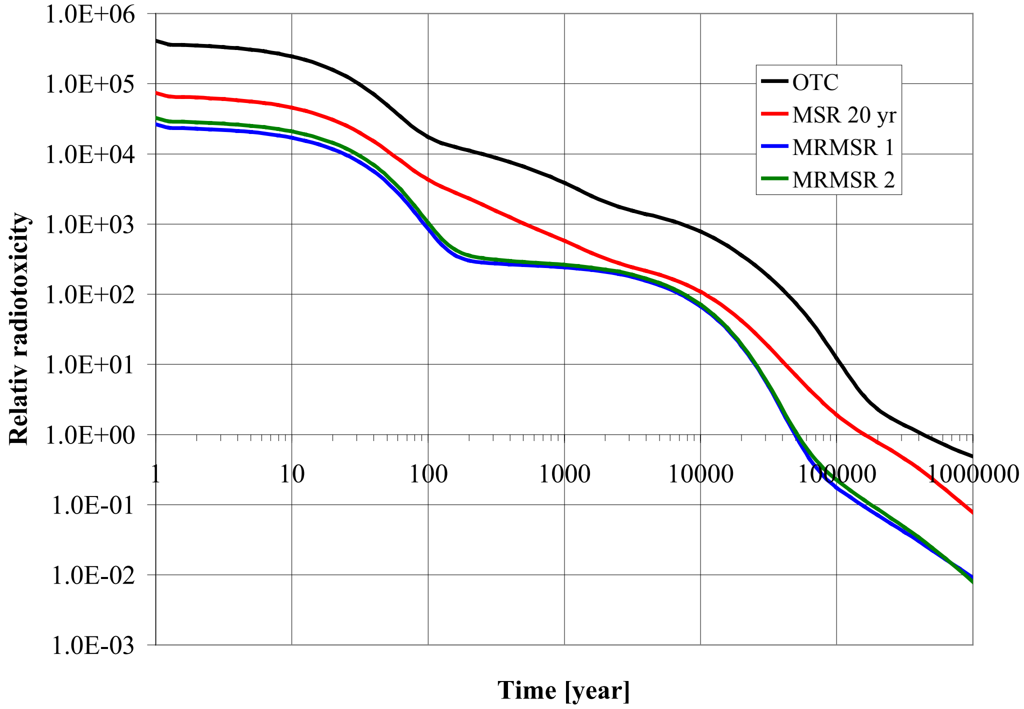

Figure 6. Relative radiotoxicity in the case of different scenarios

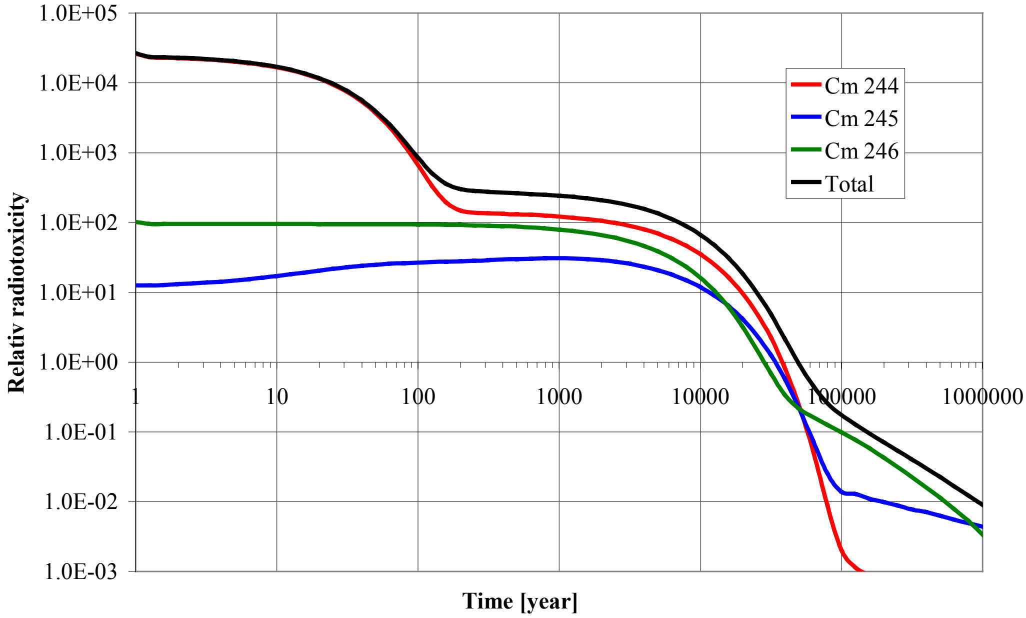

Figure 7. Relative radiotoxicity in the case of MRMSR version 1