THE MULTIREGION MOLTEN-SALT REACTOR CONCEPT

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Abstract

The molten-salt reactor (MSR) concept is one of the most promising systems for the realisation of transmutation. The objective is the development of a transmutational technique along with a device implementing it, which yield higher transmutational efficiencies than that of the known procedures. 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 sub-critical system is proposed, in which several separate flow-through irradiation rooms are formed with various neutron spectra and fluxes. The paper presents calculations that were performed for a special 5-region version of the multi-region molten-salt reactor.

Introduction

In the molten-salt reactors and sub-critical 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.

The molten-salt reactors for transmutation purposes, along with the concept of accelerator-driven molten-salt sub-critical systems were proposed in the 80s. Although the conception of the latter system was worked out at the Los Alamos National Laboratory (LANL), different concepts have become known [1-5] since then.

The molten-salt reactor has several advantages over the conventional solid fuel heterogeneous systems with respect to transmutation:

- The continuous removal of fission products from the molten-salt by continuous reprocessing reduces the neutron absorption by fission products.
- A 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 simpler and more flexible.
- Liquid fuel is far easier to reprocess than solid.
- High burn-up is achievable, since liquid fuel is significantly less sensitive to radiation damage.

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 sub-critical system, it is impossible to utilise 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 utilised by appropriately arranging the fuel elements.
- In a single space molten-salt reactor only a rather simple “time scheduling” of transmutation is possible: the duration that elapses between the removal of the spent fuel from the nuclear reactor to the start of the transmutation may be modified.

In view of the above facts, neither the molten-salt reactors nor the accelerator-driven molten-salt sub-critical 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 concepts of the molten-salt systems.

The multi-region molten-salt reactor and sub-critical system

The present work is aimed at 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 proposed 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 sub-critical system is proposed, in which several separate flow-through irradiation rooms are formed with various neutron spectra and fluxes. [6]

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

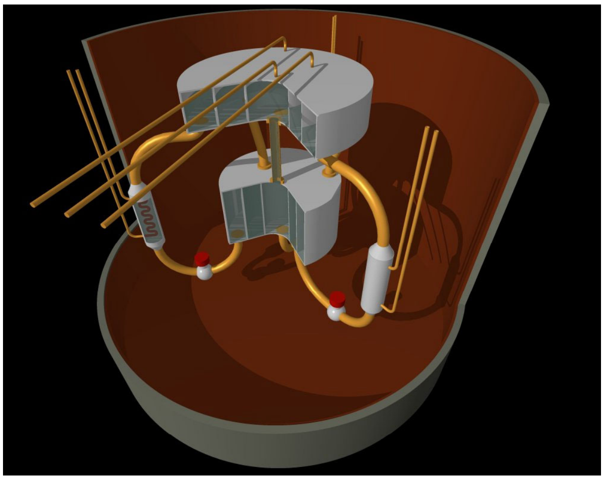


Figure 2. **Scheme of a three-region accelerator-driven molten-salt sub-critical system**

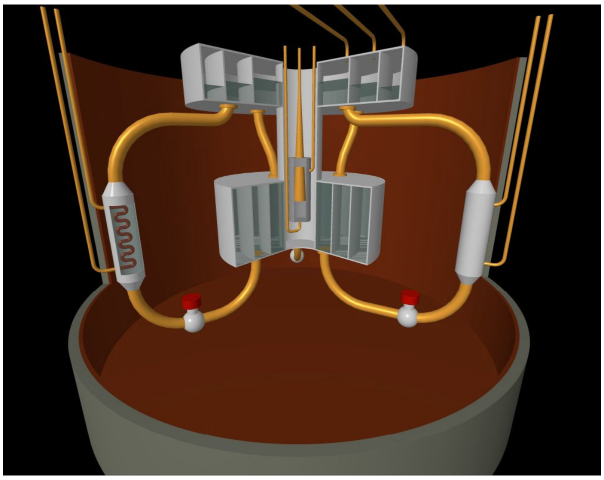


Figure 1 shows the scheme of a multi-region molten-salt reactor (MRMSR), the example being a three-region one. In this example the regions are concentric cylindrical rings. The warmed up molten-salt containing the already transmuted and to be transmuted isotopes goes to the upper expansion tank and then via the down-comer 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 sub-critical to a prescribed value under any conceivable circumstance.

In the example shown in Figure 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.

Figure 2 shows the side view of an accelerator-driven three-region molten-salt sub-critical system (MRADMS). 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 Figure 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 Figure 1.

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 (^{238}Pu, ^{239}Pu, ^{241}Pu, ^{242m}Am, ^{243}Cm and ^{245}Cm) are fissile upon interaction with thermal neutrons (the cross-sections are large). However, 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-sections is far greater than in the case of thermal neutron induced nuclear reactions. In order to illustrate this, those incident neutron energies, above which the σ_f/σ_c ratio exceeds 1, 10 and 100 are listed in Table 1. In view of these facts, those devices are advantageous for actinides, in or in part of which the flux is as high as possible above 1 MeV neutron energies. Such candidate might be e.g. the innermost region (the region closest to the proton beam or target if any) of an ADS.

Table 1. **Neutron energies above which the σ_f/σ_c ratio exceeds the given value, MeV**

σ_f/σ_c	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{243}Am	^{244}Cm	^{245}Cm
1	0,462	0.015	0[(a)]	0.353	0[(a)]	0.400	0.753	0.390	0[(a)]
10	1.04	0.505	0.361	0.752	2.42	0.836	1.39	0.706	0.152
100	-[(b)]	4.49	2.53	3.36	4.29	3.49	2.96	2.92	2.1

(a) fissile at thermal energies
(b) 78.8 at 5.01 MeV (maximum)

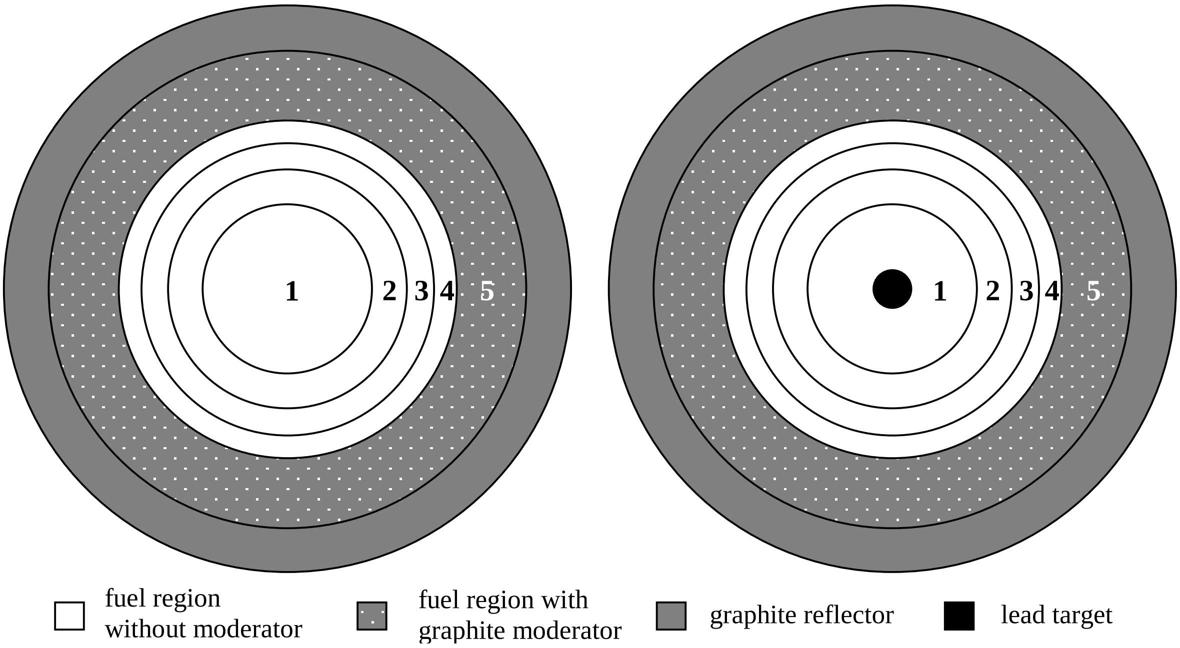
Calculations

Detailed calculations were performed for a special 5-region version of the multi-region molten-salt reactor. The core map of the proposed MRMSR can be seen in Figure 3. The 4 m high cylindrical core is divided into five coaxial regions. The four inner regions have equal volumes, while the outermost is four times larger, since this one contains graphite as moderator and the salt circulates in cylindrical channels with a diameter of 5.25 cm. The channels form a triangular lattice with a pitch of 10 cm. In this manner the graphite to salt ratio is set to 3:1, so all of the five regions contain the same amount of salt. The core is surrounded with a 47 cm thick graphite reflector, thus the overall diameter of the core is 6 m. The reason for this design is that the graphite moderated region offers a themal spectrum, while the innermost region can provide a hard spectrum, especially in the case of an ADS.

The assumed nuclear power is 2 500 MW (about 1 000 MW electric power), the salt inlet temperature is 890 K, while the average temperature in the core is 940 K. The initial salt composition is the following: 32.17 mol% BeF_2 + 67.13 mol% LiF + 0.7 mol% $(Pu+MA)F_3$. The solubility limit for Pu and minor actinides (MA) in Be/LiF salt is about 1%. 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).

This goes into the first region. The 1 year burnt out goes to the 2nd, the 2 year to the 3rd etc. The order of the subsequent regions during burn-up (load pattern) is a target of the optimisation, as well. There is no continuous feed of fuel, but the evolving fission products are continuously removed using a chemical process. [7]

Figure 3. **Core map and region numbering of the examined version of the MRMSR** (left) **and MRADMS** (right)



Preliminary calculations were performed to investigate the advantages offered by MRADMSs. The same geometry was assumed as above, with the only modification that the cylindrical beam guide and lead spallation source was placed in the centre of the core. The beam guide is a tube 20 cm in diameter with vacuum inside and stretches from the top to 10 cm above the midplane of the core, where it connects to the cylindrical lead spallation source with the same diameter which ends at the bottom of the core (see Figure 3).

As burn-up calculations for MRADMSs have not been performed yet, the same salt composition was used as obtained from the burn-up calculations for MRMSR. In order to decrease the k_{eff} to about 0.98, the height of the core was decreased. The nuclear power was decreased in the same ratio to preserve the power density. The energy of the proton beam bombarding the target was 1 GeV. The current was determined to be sufficient for the required power.

Methods

The calculations were performed with the aid of the codes of the SCALE system. [8] The modules BONAMI and NITAWL were used to produce the AMPX working library, which contains resonance self-shielded problem dependent cross-sections at the given temperatures. The XSDRNPM 1-D discrete ordinates code produced a weighted AMPX library and calculated the neutron flux, spectrum and k_{eff}. The one dimensional cylindrical geometry is sufficient because the spatial changes along the flow direction do not influence the burn-up in a fluid fuel reactor. 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 burn-up of liquid fuel reactors since, besides the nuclear reactions, it is also capable of modelling the continuous feed and blending of different partial flows.

In the case of the MRMSR, an iterative burn-up calculation scheme was applied. 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. The required libraries were created from these compositions, the power ratios were determined and the burn-up 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.

The compositions obtained from the burn-up calculations were used to perform MCNP [9] calculations to determine the k_{eff}, spectra and power ratios in the middle of the burn-up cycle. In the case of the ADSs the sufficient core height reduction was determined to achive a sub-critical state (k_{eff} ≅0.98). Then the beam guide and the spallation target was inserted into the modified geometry and the MCNPX [10] was used to simulate the sub-critical system driven by a high energy (1 GeV) proton beam directed into the lead target. The spectra and power ratios were calculated. The preliminary nature of the ADS calculation must be emphasised here. The above outlined is not the final design for a multi-region accelerator-driven molten-salt sub-critical system only an attempt to determine the expectable neutronics properties of such systems.

Results

Two different load patterns were examined in order to find the optimal way to equalise the power ratio in the regions. In the first one the fuel is loaded into the regions in the following order: 5 4 3 2 1 (see Figure 3 for numbering). Since in this case the fresh fuel is loaded into the thermal region, the power in this region becomes extremely high (Figure 4). After one year, the fissile material content of the fuel becomes so low that the inner regions will not produce significant amount of energy. This extremely unbalanced power distribution influences the shape of the spectra, as well. It can be seen in Figure 5 that the high fissile material (mainly ^{239}Pu) content of the salt prevents the appearance of a real thermal spectrum in Region 5, while the evolving thermal peaks in the inner regions show that the presence of light elements such as Li and Be in the salt has an important moderating effect when the fissile material content is low. The application of an accelerator basically affects the power distribution because the power ratio of the innermost region increases to a high degree. However, there are still significant differences between the power of the regions. The spectral changes due to the introduction of the spallation source can be extracted from Table 2 and Figure 6. It can be seen that the accelerator can only slightly increase the fast neutron flux, which is reasonable, as in the case of k_{eff}=0.98 only about 2% of the neutrons come from the spallation reactions, but it can increase the average neutron energy in high extent. While in critical reactors the average neutron energy is slightly above 2 MeV, in the innermost region of the MRADMS it can be 3 or 4 times higher.

The other investigated load pattern (3 4 5 2 1) seems capable of partly solving both the power distribution and spectral problems. As shown in Figure 4, the power ratios of the regions are far more equalised than in the previous case. An other advantageous development is that Region 3 has a quite hard, while Region 5 has a real thermal spectrum, which proves that significantly different spectra can exist in an MRMSR (see Figure 7). Unfortunately, in Region 4 and 5 the thermal peak appears again. That is why the application of the spallation source does not result in a really hard spectrum. (see Figure 8).

Figure 4. **Power ratio of the regions in different versions of MRMSR**

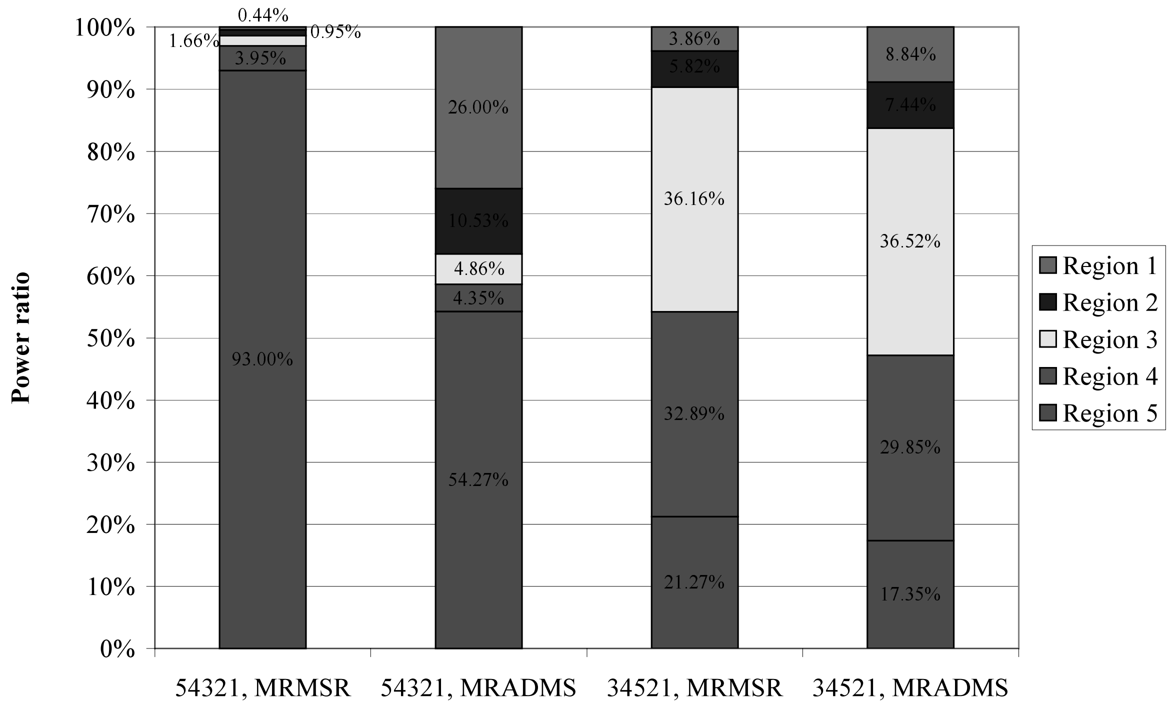


Figure 5. **Spectra in the regions of a MRMSR with load pattern 54321**

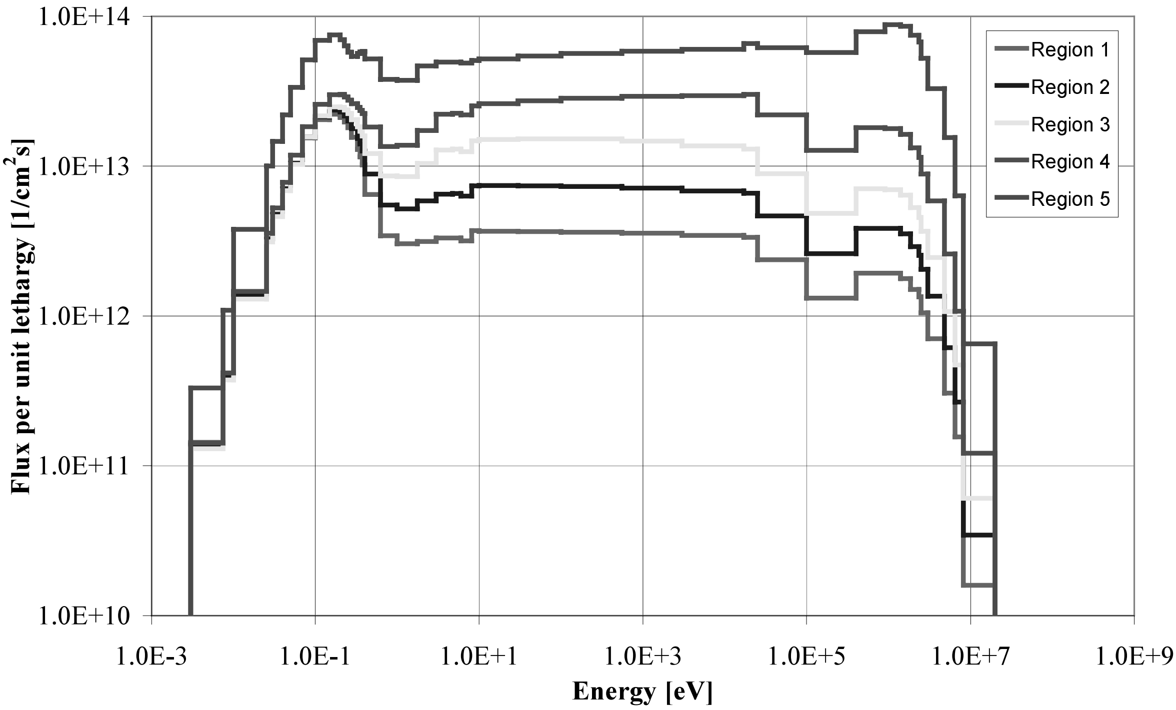


Figure 6. **Spectra in the regions of a MRADMS with load pattern 54321**

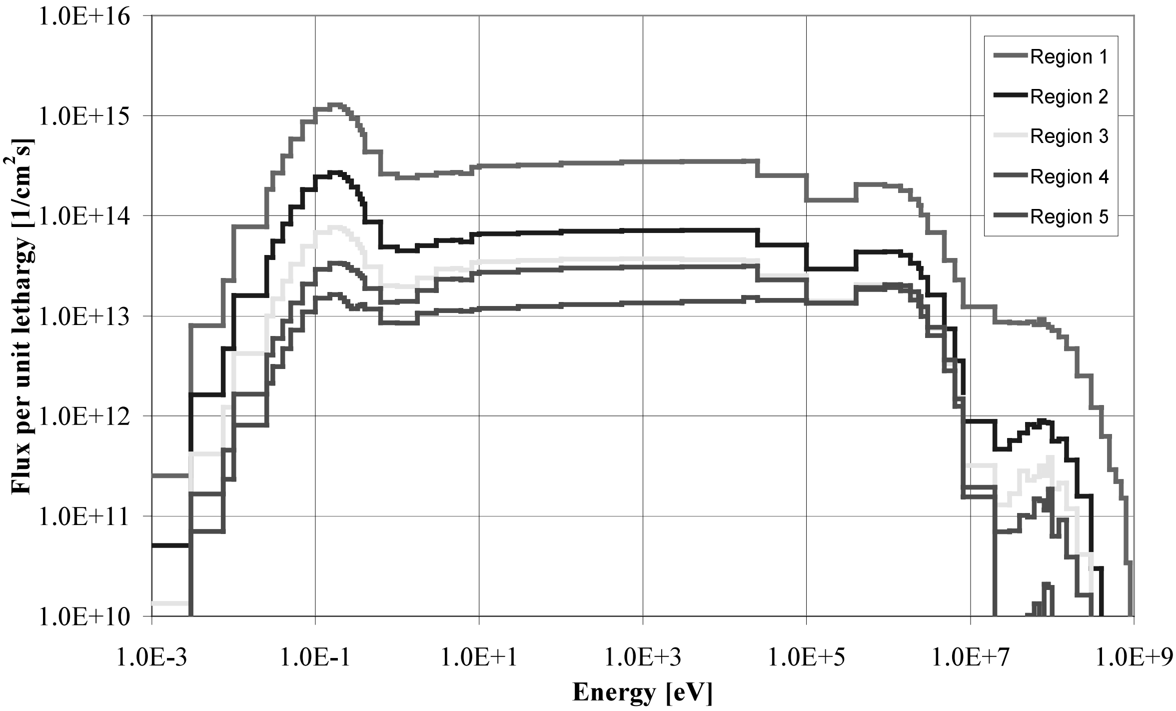


Figure 7. **Spectra in the regions of a MRMSR with load pattern 34521**

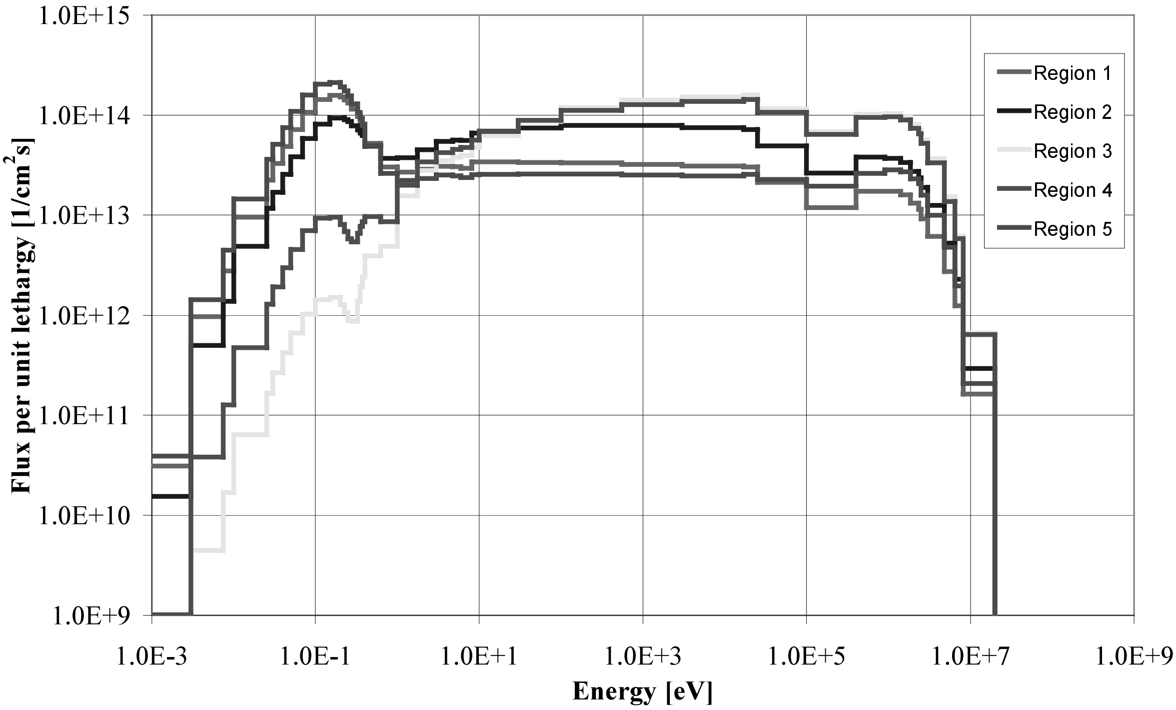


Figure 8. **Spectra in the regions of a MRADMSS with load pattern 34521**

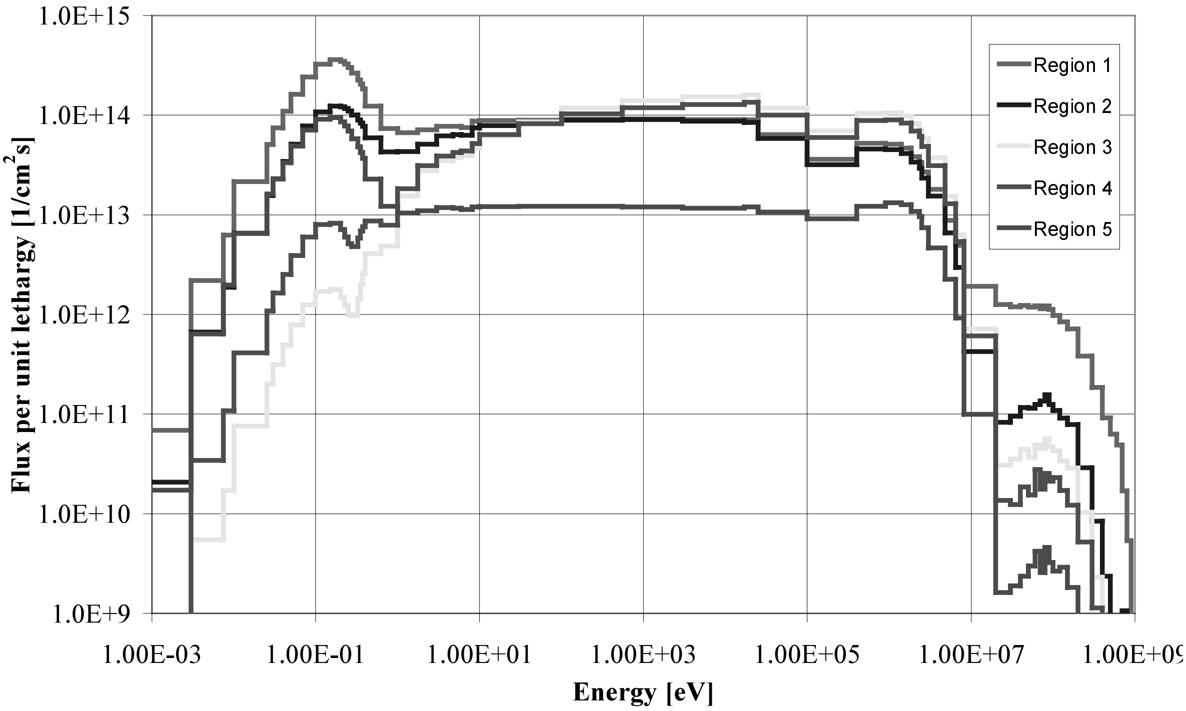


Table 2. **Ratio of fast flux (above 1 MeV) to total flux (%) and average neutron energy above 1 MeV (MeV) in different versions**

	MRMSR, 54321		MRADMS, 54321		MRMSR, 34521		MRADMS, 34521	
Region 1	2.75%	2.22 MeV	4.04%	8.74 MeV	3.00%	2.21 MeV	3.67%	6.09 MeV
Region 2	3.52%	2.20 MeV	4.07%	4.44 MeV	4.15%	2.16 MeV	4.34%	2.58 MeV
Region 3	3.82%	2.20 MeV	4.43%	3.78 MeV	8.76%	2.16 MeV	8.91%	2.23 MeV
Region 4	5.19%	2.15 MeV	5.37%	2.90 MeV	8.41%	2.16 MeV	8.45%	2.20 MeV
Region 5	10.82%	2.22 MeV	11.05%	2.28 MeV	4.61%	2.19 MeV	4.69%	2.23 MeV

Discussion

It can be stated that the above described version of the multi-region molten-salt reactor and accelerator-driven sub-critical system are suitable to realise the multi-step time-scheduled transmutational strategy, though several problems are still to be solved.

In order to avoid the moderating effect of the light elements in the salt, a new salt composition should be found. A mix of NaF and ZrF, or chloride based salts can be good candidates.

It must be ensured that the graphite moderated region remains over-moderated during the whole burn-up cycle because this is a basic safety requirement for fluid fuelled reactors. This requires the precise tuning of the moderator to salt volume ratio. Fortunately, over-moderating results in a well thermalised spectrum, which is advantageous for the transmutational efficiency.

Several other load patterns should be investigated to achieve a smooth power distribution. Further improvement may be achieved with the modification of the region volumes and fuel concentrations, if needed, however, this method needs chemical processing during refuelling. Another different strategy is that the actinides are never removed from the last region, only loaded from the previous one, while the composition reaches the equilibrium. This solution can raise the power ratio of the last region and seems advantageous for the transmutational efficiency, as well.

Further calculations on the transmutational efficiency should be performed, in a similar manner as this problem was studied in our previous papers. [11-13] During the optimisation of the power distribution, spectral parameters, etc., the viewpoint of the transmutational efficiency should always be considered.

Conclusion

The paper investigates some reactor physics properties of the multi-region molten-salt reactors and sub-critical systems on a 5-region version as an example. It can be concluded that it is possible to influence the power distribution and spectral properties by the variation of the load pattern. It was proved that significantly different spectra can be formed in the regions of the reactor, and the accelerator plays an important role in the hardening of the spectrum. However, other salt compositions should be investigated due to the moderating effect of the light elements. Further calculations are needed to determine the best core layout, load pattern and important parameters of the reactor.

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