FAST MOLTEN SALT REACTOR –TRANSMUTER FOR CLOSING NUCLEAR FUEL CYCLE ON MINOR ACTINIDES

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ABSTRACT

Creation fast critical molten salt reactor for burning-out minor actinides and separate long-living fission products in the closed nuclear fuel cycle (NFC) is the most perspective and actual direction. The reactor on melts salts - molten salt homogeneous reactor with the circulating fuel, working as burner and transmuter long-living radioactive nuclides in closed NFC, can serve as an effective ecological cordon from contamination of the nature long-living radiotoxic nuclides. High-flux fast critical molten-salt nuclear reactors in structure of the closed nuclear fuel cycle of the future nuclear power can effectively burning-out / transmute dangerous long-living radioactive nuclides, make radioisotopes, partially utilize plutonium and produce thermal and electric energy. Such reactor allows solving the problems constraining development of large-scale nuclear power, including fueling, minimization of radioactive waste and non-proliferation. Burning minor actinides (MA) in MSR is capable to facilitate work solid fuel power reactors in system NP with the closed nuclear fuel cycle and to reduce transient losses at processing and fabrications fuel pins. At substantiation MSR-transmuter/burner as solvents fuel nuclides for molten-salt reactors various salts were examined, for example: $LiF - BeF_2$; $NaF - LiF - BeF_2$; $NaF - LiF$; $NaF – ZrF_4$; $LiF - NaF - KF$; $NaCl$.

RRC "Kurchatov institute" together with other employees have developed the basic design reactor installations (RI) with MSR – burner long-living nuclides for fluoride fuel composition with the limited solubility minor actinides ($MAF_3 < 2$ mol %) and have estimated its basic characteristics. On the basis of these data employees RRC KI and VNIPIET carry out conceptual binding reactor installations with MSR – burner to the project of a factory on processing 500 tons spent fuel (SF) of reactors of type VVER-1000 in a year. During a settlement-experimental research in RRC KI it is shown, that fluoride fuel composition with high solubility minor actinides ($MAF_3 > 10$ mol %) allows to develop in some times more effective molten-salt reactor with fast neutron spectrum – burner/ transmuter of the long-living radioactive waste. In high-flux fast reactors on melts salts within a year it is possible to burn ~300 kg minor actinides per 1 GW thermal power of reactor. The technical and economic estimation given power-technological complex shows on economic efficiency of use such burner/ transmuter. After separation from spent fuel power reactors minor actinides go on burning out in molten salt reactor.

The offered concept power-technological complex with high-flux fast reactor on melts salts, intended for burning out and transmutation long-living radiotoxic nuclides, at practical realization will allow minimizing quantity of the long-living radioactive waste in system of a

nuclear power. Accommodation of such reactors at the enterprises of a fuel cycle will provide with their energy and will facilitate the decision of a problem of radioactive waste management with the minimal losses. Small share MSR (5-7) % from full electric power in structure of the future nuclear power provides practically full burning of all minor actinide.

1. FAST CRITICAL MOLTEN SALT REACTOR CONCEPT

The most perspective and actual direction, in opinion of authors, is a creation fast critical molten salt reactor for burning-out minor actinides and separate long-living fission products in the closed nuclear fuel cycle (NFC). High-flux fast critical molten-salt nuclear reactors in structure of the closed nuclear fuel cycle of the future nuclear power can effectively burning-out / transmute dangerous long-living radioactive nuclides, make radioisotopes, partially utilize plutonium and produce thermal and electric energy. Such reactor allows solving the problems constraining development of large-scale nuclear power, including fueling, minimization of radioactive waste and non-proliferation. Burning minor actinides (MA) in MSR is capable to facilitate work solid fuel power reactors in system NP with the closed nuclear fuel cycle and to reduce transient losses at processing and fabrications fuel pins.

During a settlement-experimental research in RRC "Kurchatov Institute" it is shown, that fluoride fuel composition with high solubility minor actinides (MAF_3 > 10 mol %) allows to develop in some times more effective molten-salt reactor with fast neutron spectrum – burner/ transmuter of the long-living radioactive waste. The power-technological complex with high-flux fast reactor on melts salts with the general thermal power 2.5 GW(th) will provide burning out more than 700 kg in a year Np, Am, Cm, thus its electric power will make 1.1 GW(e). Table 1 shows the efficiency of high flux MSR (HFMSR) and high flux fast MSR (HFFMSR) for incineration of minor actinides from VVER-1000 spent fuel.

Table 1. High flux molten salt reactor-transmuter efficiency

Actinide	Amount discharged from reactor VVER-1000, kg/year	Fluoride Fuel Composition (MAF_3 < 2 mol %)		Fluoride Fuel Composition (MAF_3 > 10 mol %)	
		Heavy-nuclei rate required for feeding HFMSR, kg/year	Number of attended reactors as VVER-1000	Heavy-nuclei rate required for feeding HFFMSR, kg/year	Number of attended reactors as VVER-1000
Pu	216.8	450.3	8	–	20
Np	13.3	103.9		237.7	
Am	28.1	220.3		503.9	
Cm	0.3	2.3		5.3	

The technical and economic estimation of the given power-technological complex shows an economic acceptability of use MSR- burner/ transmuter long-living radioactive waste.

2. MODELLING OF FAST MSR EQUILIBRIUM

Molten salt reactor is proposed for incineration minor actinides from power reactor VVER-1000 type spent fuel. Spent fuel parameters for 4.4% U-235 initial enrichment, 4.0% burning and 10 years cooling are listed in tables 2 and 3.

Table 2. Actinides in power reactors spent fuel, g/kg

Actinide	Mass
Np	0.488
Pu	8.24
Am	0.6092
Cm	8.92E-03
Total	9.346

Table 3. Nuclide fractions in spent fuel

Np-237	1.000E+00	**Pu-236**	4.910E-09	**Am-241**	8.921E-01	**Cm-242**	1.378E-05
		Pu-238	1.680E-02	**Am-242m**	7.665E-04	**Cm-243**	1.987E-02
		Pu-239	6.107E-01	**Am-243**	1.072E-01	**Cm-244**	8.824E-01
		Pu-240	2.235E-01			**Cm-245**	9.706E-02
		Pu-241	9.970E-02			**Cm-246**	6.380E-04
		Pu-242	4.930E-02			**Cm-247**	4.737E-06
						Cm-248	2.197E-07
Total	1.000E+00		1.000E+00		1.000E+0 0		1.000E+0 0

Neptunium, americium and curium are used as MSR feed. Total annual feed mass in 1000 kg corresponds to MSR unit power ~ 1 GW(e). Actinide masses in feed are listed in tables 4, 5.

Table 4. Actinide masses in annual MSR feed

Actinide	kg/year
Np	441.182
Am	550.754
Cm	8.064
Total	1000.0

Table 5. Nuclide fraction in annual MSR feed

Nuclide	Fraction	MSR Feed, kg/year
Np-237	4.41182E-01	4.41182E+02
Am-241	4.91315E-01	4.91315E+02
Am-242m	4.22157E-04	4.22157E-01
Am-243	5.90171E-02	5.90171E+01

Cm-242	1.11124E-07	1.11124E-04
Cm-243	1.60197E-04	1.60197E-01
Cm-244	7.11598E-03	7.11598E+00
Cm-245	7.82733E-04	7.82733E-01
Cm-246	5.14494E-06	5.14494E-03
Cm-247	3.82039E-08	3.82039E-05
Cm-248	1.77149E-09	1.77149E-06
Total	1.0	1000.0

2.1. MSR calculational model

MSR calculational model (Fig. 1) is R-Z cylinder. Inside the reactor vessel there are homogeneous core with fuel salt, top graphite reflector, bottom graphite reflector and side graphite reflector including fuel salt inlet. Core is divided into three parts: C1 – top core, C2 – middle core and C3 – bottom core. At present state of calculations all core parts are the same composition. Reactor material parameters are listed in Table 6.

Table 6. Reactor material parameters

Zone and material	Temperature, K	Density, g/cm^3	Nuclear density, 1/barn-cm
Core and fuel salt inlet			
Fuel salt composition:			
(11% mol. MA in fuel salt)			
$0.4115 \cdot LiF + 0.103 \cdot NaF + 0.3755 \cdot KF + 0.11 \cdot MAF_3$	950	2.96	
Li-6			6.16577E-06
Li-7			1.05646E-02
F-19			3.13398E-02
Na-23			2.64590E-03
K-nat			9.64598E-03
MA			2.81547E-03
Graphite reflector	800		
C			8.760E-2
Reactor vessel	700		
C			3.44E-05
S			2.32E-05
Ti			6.45E-04
Mn			5.60E-04
Ni-58			5.12E-02
Ni-60			1.89E-02
Ni-61			8.05E-04
Ni-62			2.52E-03
Ni-64			6.18E-04
Mo			9.89E-03

Average neutron flux in MSR core and fuel salt inlet is assumed $2 \cdot 10^{15}$ neutron/(cm^2·sec). The time intervals during which fuel salt passes through reactor (core and fuel salt inlet) and through outer circuit is equal, so average neutron flux in system is $1 \cdot 10^{15}$ neutron/(cm^2·sec).

Equilibrium state for MSR with given feed, fluoride fuel composition (MAF_3 11 mol %) and average neutron flux in system was obtained. Calculations were performed using MCNP5 code [1] and nuclear data was obtained using NJOY99 system [2] from library ENDF/B-VI [3, 4]. The total number of histories in calculation was $1.2 \cdot 10^6$, so the statistical deviation of neutron multiplication factor Keff was about 0.0004.

2.2. Equilibrium state for MSR

For given reactor parameters and feed (in assumption that fuel salt is in core during 10 seconds and then 10 seconds out of core) the heavy metals equilibrium was calculated about 18 tons. Table 7 shows the equilibrium masses of heavy metals.

Table 7. Equilibrium masses of heavy metals

Element	Equilibrium mass in core, kg	Equilibrium mass out of core, kg
Th	0.157	0.157
Pa	0.014	0.014
U	1351.704	1351.704
Np	2534.735	2534.735
Pu	9415.769	9415.769
Am	3381.377	3381.377
Cm	1126.647	1126.647
Bk	0.237	0.237
Cf	2.007	2.007
Total	17812.65	17812.65

Neutron-physical calculation of MSR model with equilibrium fuel composition was performed and neutron multiplication factor obtained is equal Keff = 1.0215±0.0004. Neutron spectra were calculated in three cores C1, C2, C3 (each core was divided into 2 parts – central part with radius 50 cm and peripheral part) and fuel salt inlet. Spectra are shown in Figures 2, 3, 4 and 5. One can see that neutron spectrum is thermal in fuel salt inlet, harder in top and bottom cores and fast in central part of middle core. In core total volume 17.5 m^3 the heavy metal mass was about 19.9 tons, total power release ~ 2.86 GW and average power density ~ 160.9 MW/ m^3.

For estimation of radial non-uniformity of power release the core was divided into radial cylindrical zones: 1 cm thickness for first 10 cm of the core outer part and 10 cm thickness for the core inner part. In each of this zone volume averaged power release was obtained and normed on total volume averaged power release in core. Calculated values versus center points of radial cylindrical zones are shown in Figure 6.
For estimation of axial non-uniformity of power release the core was divided into axial cylindrical zones: 1 cm thickness for first 10 cm of the core top and bottom and 10 cm thickness for the core inner part. In each of this zone volume averaged power release was

obtained and normed on total volume averaged power release in core. Calculated values versus center points of axial cylindrical zones are shown in Figure 7.

Fission products equilibrium was also calculated in assumption that:

- The heavy metals equilibrium is a constant source of fission products;
- Fission products (excluding Kr and Xe) are continuously removed from the fuel composition;
- Fission products disposal cycle during which 100% equilibrium amount is removed was adopted 100 days, 1 year и 3 years;
- Kr and Xe are totally removed from fuel salt at the core outlet;
- Fission products individual yields and decay parameter are obtained from libraries ENDF/B-VI и JENDL-3.2 [5];
- Fission products neutron reaction rates are calculated using MCNP5 code and ENDF/B-VI library.

In equilibrium state for given power 989.785 kg/year fission products are generated. Fission products equilibrium mass in core depends on disposal cycle and comes to ~ 92.7 kg (1.1 % to heavy metal nuclides) for 100 days cycle, ~ 337 kg (3.9% to heavy metal nuclides) for 1 year cycle and ~1009.4 kg (11.8% to heavy metal nuclides) for 3 years cycle. Besides ~ 71 g/day Kr and ~ 517 g/day Xe are released. The results are shown in Table 8.

Table 8. Fission products equilibrium amounts

Element	Equilibrium mass in core, kg			Disposal rate, g/day		
	FP cycle 100 days	FP cycle 1 year	FP cycle 3 years	FP cycle 100 days	FP cycle 1 year	FP cycle 3 years
Ge	0.004	0.014	0.042	0.039	0.039	0.039
As	0.001	0.004	0.011	0.012	0.011	0.010
Se	0.197	0.719	2.156	1.970	1.969	1.967
Br	0.070	0.253	0.734	0.704	0.693	0.670
Rb	0.004	0.015	0.045	0.042	0.042	0.041
Sr	0.577	1.905	5.278	5.765	5.215	4.817
Y	0.601	1.235	2.466	6.013	3.381	2.251
Zr	10.321	36.343	107.239	103.208	99.500	97.868
Nb	0.417	0.843	1.061	4.167	2.308	0.968
Mo	10.309	40.428	125.265	103.094	110.685	114.319
Tc	3.134	11.538	33.102	31.342	31.590	30.209
Ru	14.159	46.524	129.900	141.585	127.377	118.549
Rh	2.333	11.091	34.692	23.331	30.366	31.661
Pd	7.747	31.149	103.648	77.475	85.282	94.591
Ag	1.040	3.687	10.865	10.395	10.093	9.915
Cd	0.948	3.663	11.846	9.479	10.028	10.811
In	0.067	0.250	0.735	0.671	0.684	0.671
Sn	0.496	1.783	5.349	4.965	4.881	4.882
Sb	0.200	0.656	1.677	1.996	1.797	1.530
Te	2.432	8.281	24.489	24.318	22.673	22.349
I	1.464	4.551	12.643	14.640	12.461	11.538
Cs	0.605	2.136	6.092	6.051	5.849	5.560
Ba	1.640	4.706	13.241	16.403	12.884	12.084

La	2.486	8.864	26.308	24.863	24.270	24.009
Ce	11.063	35.446	95.043	110.627	97.045	86.738
Pr	3.307	13.494	41.738	33.069	36.945	38.091
Nd	11.382	45.947	148.197	113.815	125.796	135.247
Pm	1.513	4.891	10.038	15.133	13.390	9.161
Sm	3.010	12.220	41.586	30.102	33.457	37.952
Eu	0.700	2.479	7.298	7.000	6.786	6.660
Gd	0.365	1.543	5.699	3.647	4.225	5.201
Tb	0.046	0.159	0.440	0.459	0.436	0.401
Dy	0.032	0.132	0.490	0.320	0.360	0.447
Ho	0.001	0.004	0.012	0.011	0.011	0.011
Er	0.001	0.004	0.013	0.009	0.010	0.012
Total	92.672	336.957	1009.436	926.7	922.5	921.2

Criticality calculations were performed for various fission products equilibrium amounts in core. The results are shown in Table 9.

Table 9. Fission products effect on neutron multiplication factor Keff

Calculation mode	Keff	Δ(1/K)
Without fission products	1.0215 ± 0.0004	
Fission products cycle 100 days	1.0198 ± 0.0004	-0.17%
Fission products cycle 1 year	1.0155 ± 0.0004	-0.59%
Fission products cycle 3 years	1.0043 ± 0.0004	-1.71%

3. CONCLUSION

1) Fast molten salt reactor–transmuter with fluoride fuel composition with high solubility minor actinides (MAF_3 > 10 mol %) is proposed for closing nuclear fuel cycle on minor actinides. The technical and economic estimation of the power-technological complex with MSR shows an economic acceptability of use MSR - burner/transmuter long-living radioactive waste.

2) Reactor calculational model is designed and equilibrium state modeling is performed for incineration minor actinides from power reactor VVER-1000 type spent fuel. In the equilibrium state MSR is fed by Np, Am, Cm and does not need Pu feed.

3) It is shown that neutron spectrum is hard in the core center and softer in the core periphery.

4) In core total volume 17.5 m^3 the heavy metal mass was about 19.9 tons, total power release ~ 2.86 GW and average power density ~ 160.9 MW/ m^3. About 990 kg/year fission products are generated. Fission products equilibrium amounts in core depend on fission product disposal rate and vary from ~ 100 kg to 1000 kg.

5) Fission products effect on Keff varies from -0.17 % to -1.71 % Δ(1/K).

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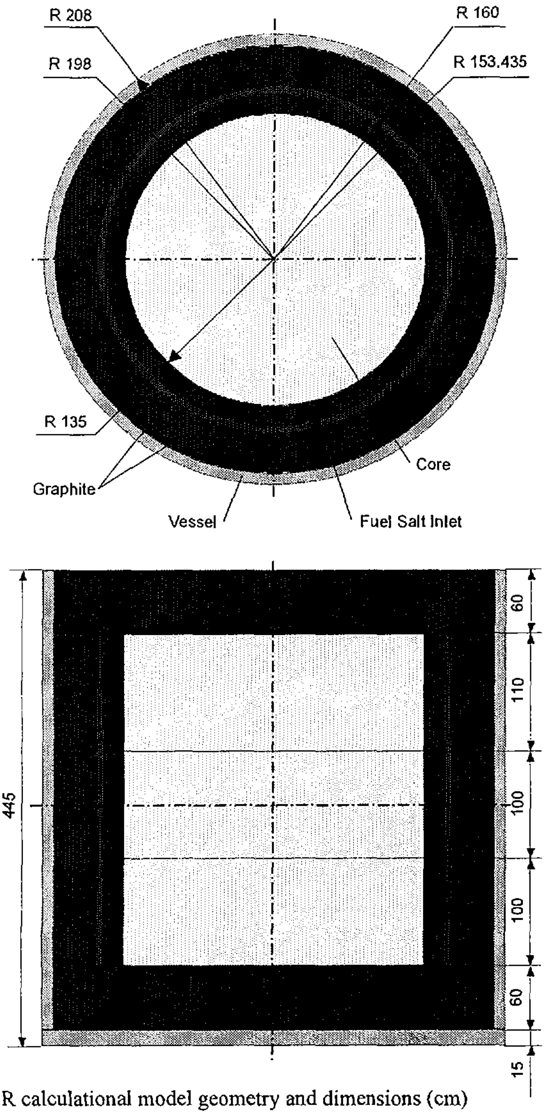

Figure 1. MSR calculational model geometry and dimensions (cm)

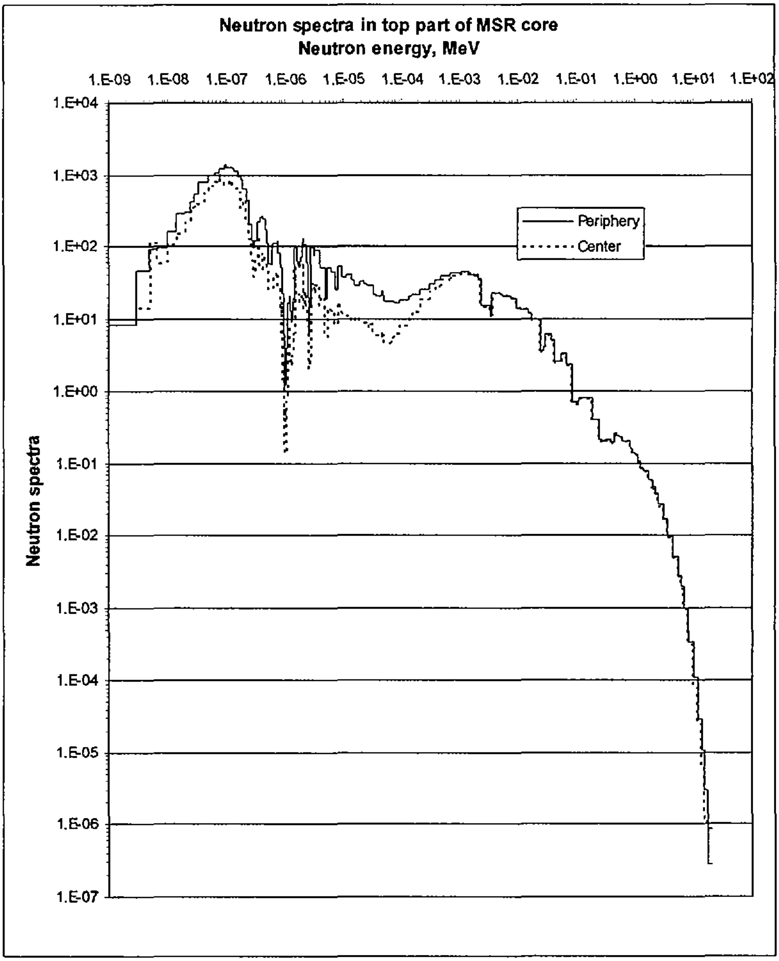

Figure 2. Neutron spectra in the top part of the MSR core (C1)

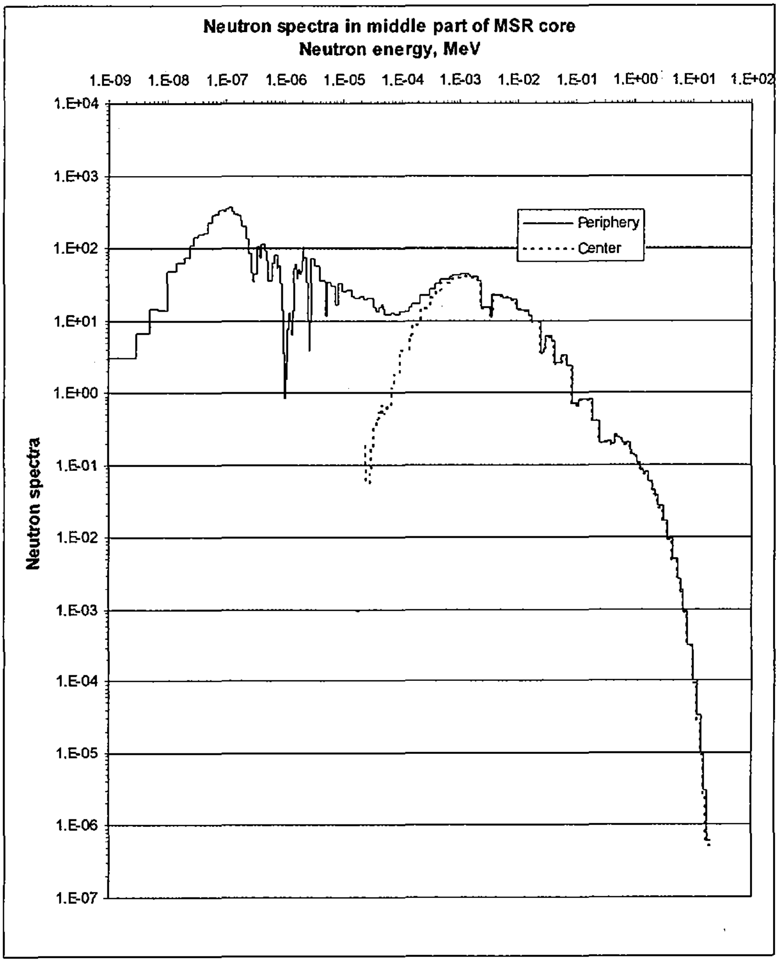

Figure 3. Neutron spectra in the middle part of the MSR core (C2)

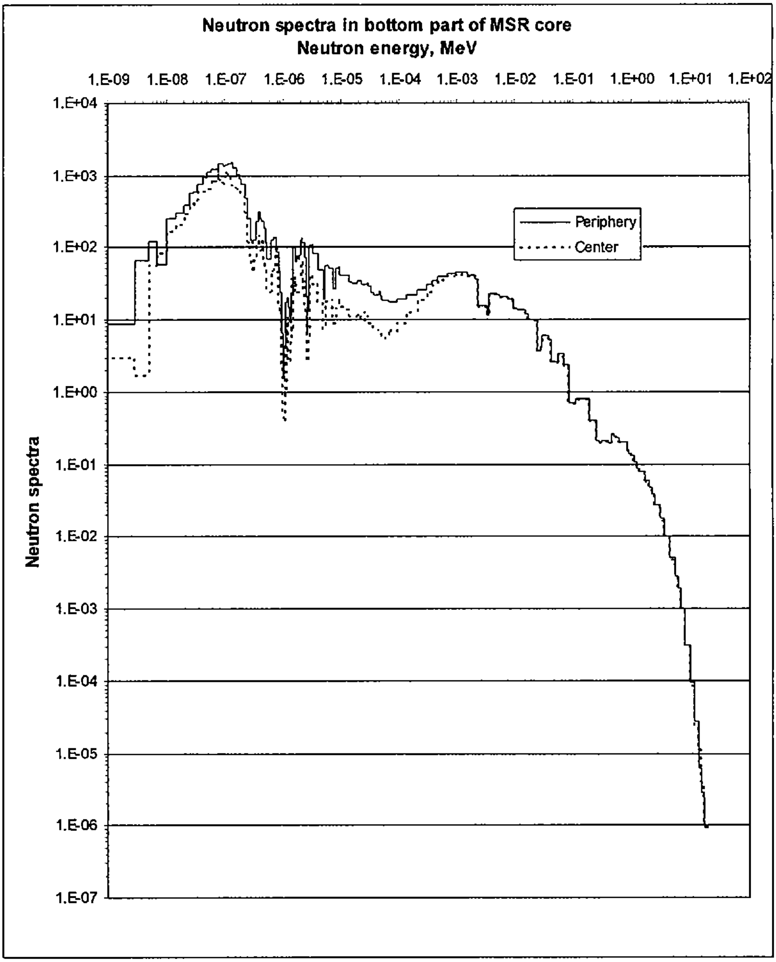

Figure 4. Neutron spectra in the bottom part of the MSR core (C3)

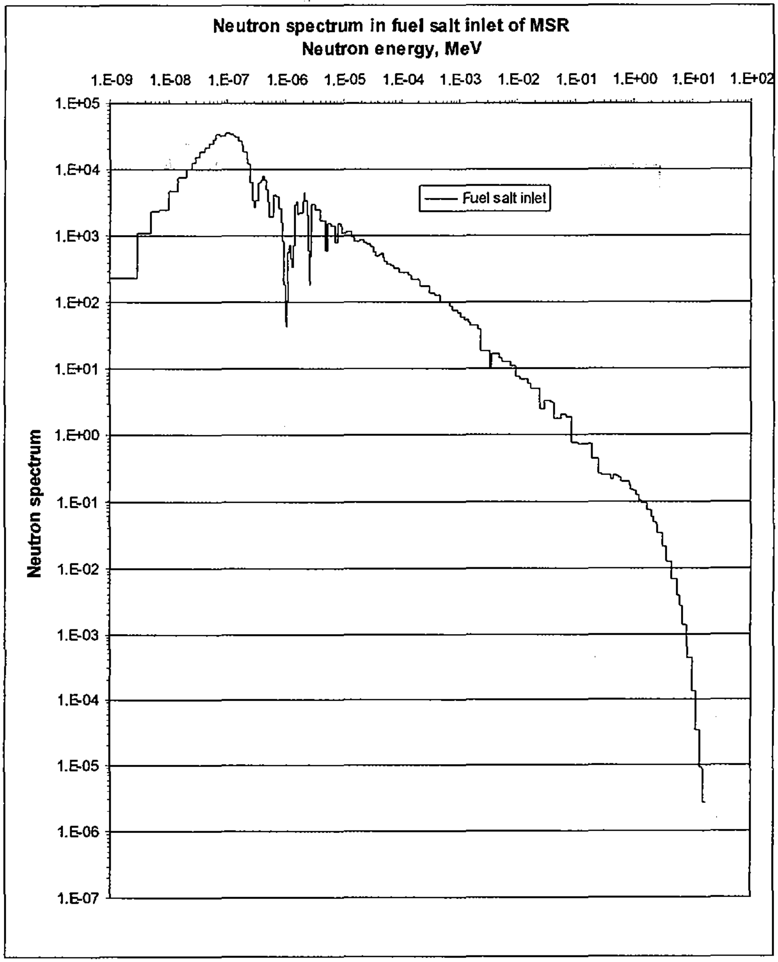

Figure 5. Neutron spectrum in the fuel salt inlet of the MSR

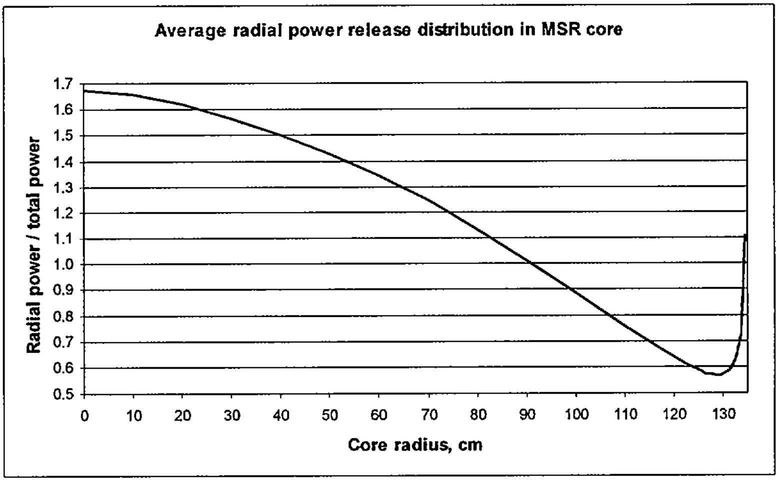

Figure 6. Average radial power release in core

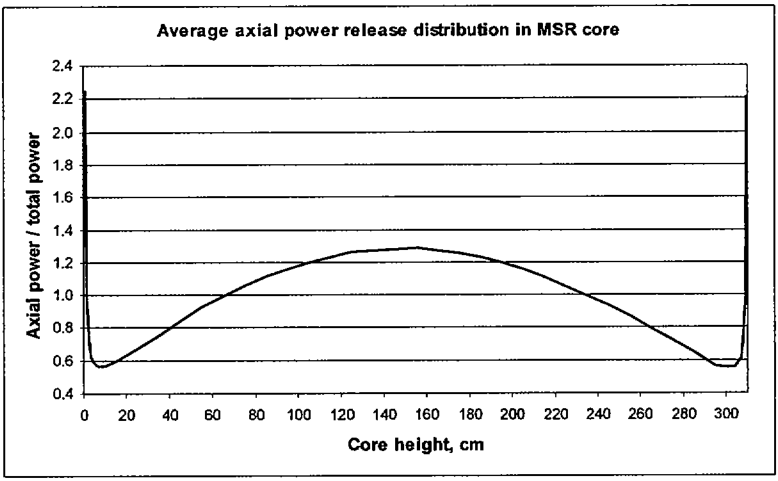

Figure 7. Average axial power release in core