Molten salt reactor: Overview and perspectives

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Abstract

Recent molten salt reactor (MSR) developments in Europe and Russia address an advanced large power unit without graphite in the core. In new MSR concepts neutron spectrum is hard enough and the reprocessing rate strongly reduced compared to prior graphite moderated Th-U MSBR design developed at United States ORNL. The consideration done demonstrates the MSR potential as the system with flexible fuel cycle options which can operate with different loadings and make up scenarios based on transuranic elements from spent LWR fuel without and with Th-U support. This system may operate in the actinide recycler and transmuter, convertor as well as breeder modes without essential changes in the core/blanket configuration and find its place in any scenario of nuclear energy development.

Introduction

Nuclear energy systems employing liquid salt fluorides were investigated by Oak Ridge National Laboratory (ORNL, United States) in the 60s and 70s [1-3]. The favourable experience gained from the 8 MWt MSRE test reactor operated from 1965 to 1969 [1], and the development of chemical control of molten salt technologies and metallic materials compatible with fluoride melts prepared the technical base for MSR creation. Two main configurations of 1 000 MWe graphite moderated Th-U Molten Salt Breeder Reactor (MSBR) designs were developed at ORNL: *i)* single stream [2]; *ii)* two-fluid [3]. The technical feasibility of such systems, the key parameters of which are listed in Table 1 (first two columns), no longer raise any doubts. MSBR [3] possesses good characteristics, but demands continuous removal of soluble fission products (removal time for lanthanides is about 30 days). Creation of such system for intensive fission product clea-up in MSBR (first of all, for single stream one) is a challenge, in particular, remain difficulties on actinide losses to waste and selection of constructional materials for the fuel clean up unit.

In Russia, the molten salt programme was started in the second half of 1970s [4]. Last decade within ISTC #1606 main focus was placed on experimental and theoretical evaluation of single stream Molten Salt Actinide Recycler and Transmuter (MOSART) system fuelled only by transuranium elements (TRU) trifluorides from LWR spent fuel [5]. It was shown that optimum spectrum for MOSART is intermediate/fast spectrum of homogeneous core without graphite moderator. Promising configuration for 2 400 MWt MOSART is the homogeneous cylindrical core (3.6 m high and 3.4 m in diameter) with 0.2 m graphite reflector filled by 100% of molten 15LiF-58NaF-27BeF_2 or 73LiF-27BeF_2 salt mixture. It is feasible to design critical homogeneous core fuelled only by TRU from UOX and MOX PWR spent fuel while equilibrium concentration for trifluorides of actinides (about 1 mole% for the rare earth removal cycle 300 EFPD) is truly below solubility limit at minimal fuel salt temperature in primary circuit 600°C (about 3 mole %). The effective flux of such system is near 1×10^{15} n/cm^2/s [5]. Due to possibility of operation without additional neutron sources, MOSART loaded only by TRU from spent nuclear fuel has maximum capacity, high enough transmutation efficiency and can be loaded by the fuel of wide range of compositions.

- Recent molten salt Th-U breeder developments in Europe (CNRS, France) [6] also addresses advanced large power unit without graphite in the core and fast neutron spectrum (MSFR). In MSFR, prior ORNL concept has been revisited by removing graphite and BeF_2. The neutron spectrum is fast and the reprocessing rate strongly reduced down to 40 litres per day to get a positive breeding gain. The reactor is started with ^{233}U or with Pu and minor actinides (MA) mixture from PWR spent fuel. For MSFR as solvent system for fuel and blanket circuits it is offered to consider molten 78LiF-22ThF_4 ($T_{melting}$ = 565°C). Note that this solvent has essentially higher melting temperature and solubility for actinide trifluorides, in comparison with well established 72LiF-16BeF_2-12ThF_4 melt ($T_{melting}$ = 504°C). MSFR exhibit large negative temperature and void reactivity coefficients, a unique safety characteristic not found in solid-fuel fast reactors. ^{233}U production rate in MSFR is about 100 kg/yr. Compared to MSBR basic difficulty of MSFR is that it requires essentially higher starting loadings of fissile materials (5 060 kg of UF_4 or 11 200 kg of $TRUF_3$) and fuel concentrations for criticality. In MFSR starting loading concentration of UF_4 ($TRUF_3$) in fuel salt will make, accordingly, 2.5 (6.5) mole %. Because of limits on solubility of $TRUF_3$ molten 78LiF-16ThF_4-6.5$TRUF_3$ mixture can be realised only at the minimum temperature in fuel circuit >700°C [8,9]. In this case the maximum core temperature of fuel salt makes 850°C [6]. That will demand development of a new constructional material for MSFR fuel circuit since developed for MSBR and MOSART Ni-Mo alloys of Hastelloy N type are certified only for temperature of about 704°C. As advanced high performance material for MSFR design recently has been proposed new Ni-W-Cr alloy [6].
- Unification of MOSART system with Th containing molten salt blanket proposed within ISTC #3749 [7] can provide: *i)* its core operation with minor-actinide-bearing fuels basing on additional ^{233}U support; *ii)* effective production of ^{233}U required for starting uranium

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loading of MSFR. In the first case, when the core is processed unburned actinides are returned to the core along with any ^{233}U available from the blanket. In second case uranium processed from blanket is removed for MSFR loading, but in both cases thorium-bearing salt is returned to the blanket.

- From the point of view of structure such symbiosis MOSART with Th containing molten salt blanket can have advantage, since allows to use in a greater degree main feature of MSR which concern fuel cycle flexibility. MSR in this case can operate with more wide range of fuel loadings without core modification and thus can be included in different scenario of nuclear power development in different modes as actinide recycler and transmuter, Th-U converter or breeder. Beside this it can be realised on the base of existing technologies within technological margins.

Thus, R&D on MSR in Europe and Russia is focused last time on fast spectrum concepts which have been recognised as long-term alternatives to solid-fuelled fast neutron reactors with attractive features (strong negative feedback coefficients, easy in-service inspection, and simplified fuel cycle). Concepts are available for breeding and TRU burning. The basic characteristics of recent MOSART and MSFR concepts [5,6] compared to well established graphite moderated MSBR design [2,3] developed in 1970s are listed below.

Table 1: The basic characteristics of molten salt systems

Design	MSBR [2,3]		MSFR [6]		MOSART [5,7]	
Type	Breeder		Breeder		Burner/converter	
Neutron spectrum	Thermal		Fast		Fast	
Number of fluid streams	2	1	2	2	1	2
Thermal capacity, MW	2 250	2 250	3 000	3 000	2 400	2 400
Fuel salt temperature, °C	566/704	566/704	600/750	700/850	600/720	600/720
Fuel salt composition, mole %	68LiF 31BeF_2 0.2UF_4	72LiF 16BeF_2 12ThF_4 0.2UF_4	77.5LiF 20ThF_4 2.5UF_4	78LiF 16ThF_4 6.5$TRUF_3$	72LiF 27BeF_2 1$TRUF_3$	72LiF 27BeF_2 1$TRUF_3$
Blanket salt composition, mole %	71LiF 2BeF_2 27ThF_4	No	78LiF 22ThF_4	78LiF 22ThF_4	No	75LiF 5BeF_2 20ThF_4
Fuel cycle	U -Th	U -Th	U-Th	TRU-Th-U	w/o U,Th	TRU-Th-U
Soluble fission product removal time, EFPD	30-50	10-30	418	418	300	300

Transmuter mode

As the calculation tool MCNP-4B+ORIGEN2.1 code adapted to the specific features of MSR was used on all stages of the work. As the starting point of our neutronic and fuel cycle consideration MOSART core concept was chosen. Originally single fluid MOSART concept was considered as dedicated burner for homogeneous TRU recycling without Th and U support. Following TRU mixtures from spent UOX (1) and MOX (2) PWR fuel after one year cooling were considered for its core make up: *i)* ^{238}Pu-3.18%, ^{239}Pu-43.93%, ^{240}Pu-21.27%, ^{241}Pu-13.52%, ^{242}Pu-7.88%; ^{237}Np-6.42%; ^{241}Am-0.55%; ^{243}Am-2.33%; Pu/(Np+MA) = 9; *ii)* ^{238}Pu-2.77%; ^{239}Pu-48.36%; ^{240}Pu-19.97%; ^{241}Pu-8.30%; ^{242}Pu-6.25%; ^{237}Np-6.51%; ^{241}Am-5.56%; ^{243}Am-1.69%; Pu/(Np+MA) = 5.

Two-fluid MOSART configuration with the thorium containing blanket can be used for accumulation of leakage neutrons in the form of ^{233}U produced. The minimal TRU concentration in fuel salt and maximum ^{233}U production was obtained for ring core with height 3.2 m/outer radius 1.6 m supplied by inner 0.7 m radius blanket and outer 0.6 m thickness blanket (Figure 1). Fuel salt volume in the external part of primary circuit was reserved as 11 m^3. This system has equilibrium TRU concentration of about 2 mole % (Figures 2 and 3), ^{233}U production rate ~300 kg/year and it is able to produce ^{233}U start loading for MSFR (~5 t) during 16-18 years of

operation (Figure 4). When all ^{233}U produced in the blanket is recycled to MOSART fuel stream, $TRUF_3$ concentration at equilibrium in fuel salt will decrease down to 0.45 mole % after 8 years of operation (Figures 2 and 5).

After 10 years MOSART operation with ^{233}U recycling from blanket to the fuel stream there is an opportunity to decrease the Pu/(Np+MA) ratio in $TRUF_3$ make-up from 9 (UOX case) down to 2 (Figure 6). For make-up by $TRUF_3$ with Pu/(Np+MA) ≈ 1.5 ratio critical TRU concentration of 3 mole% in the fuel salt of this type is required (see Figure 5).

The calculations had done show the potential of Li,Be/F MOSART with Th containing molten salt blanket in heterogeneous schemes of recycling. Within technological margins it is possible to burn any composition of spent LWR fuel down to Pu/(Np+MA) = 1.5 ratio (see Figure 5). It seems that for more poor fuel compositions on the base of MA only, salt solvents systems without BeF_2 with higher solubility for trifluorides are required (for example, LiF-NaF-KF). Unfortunately, to get critical loading more than 30 t of MA will be required.

Figure 1: Ring core configuration

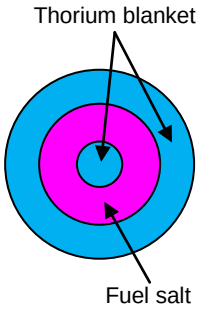


Figure 2: Concentrations for actinide and lanthanides trifluorides in fuel salt vs. time for ring core

Solid line – ^{233}U to stock, dashed line – ^{233}U recycling from blanket to fuel stream

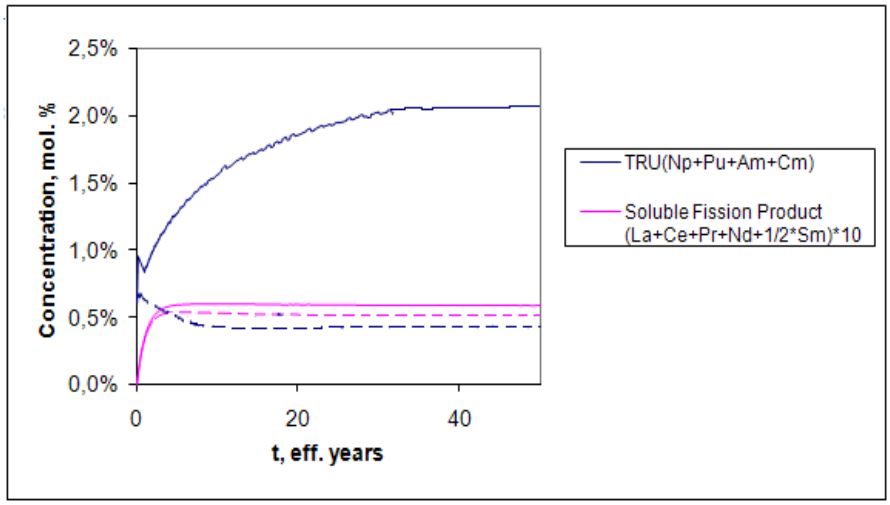


Figure 3: Equilibrium $TRUF_3$ concentration in fuel salt (in mole%) vs. time for ring core configuration

Solid line – ^{233}U to stock, dashed line – ^{233}U recycling from blanket to fuel stream

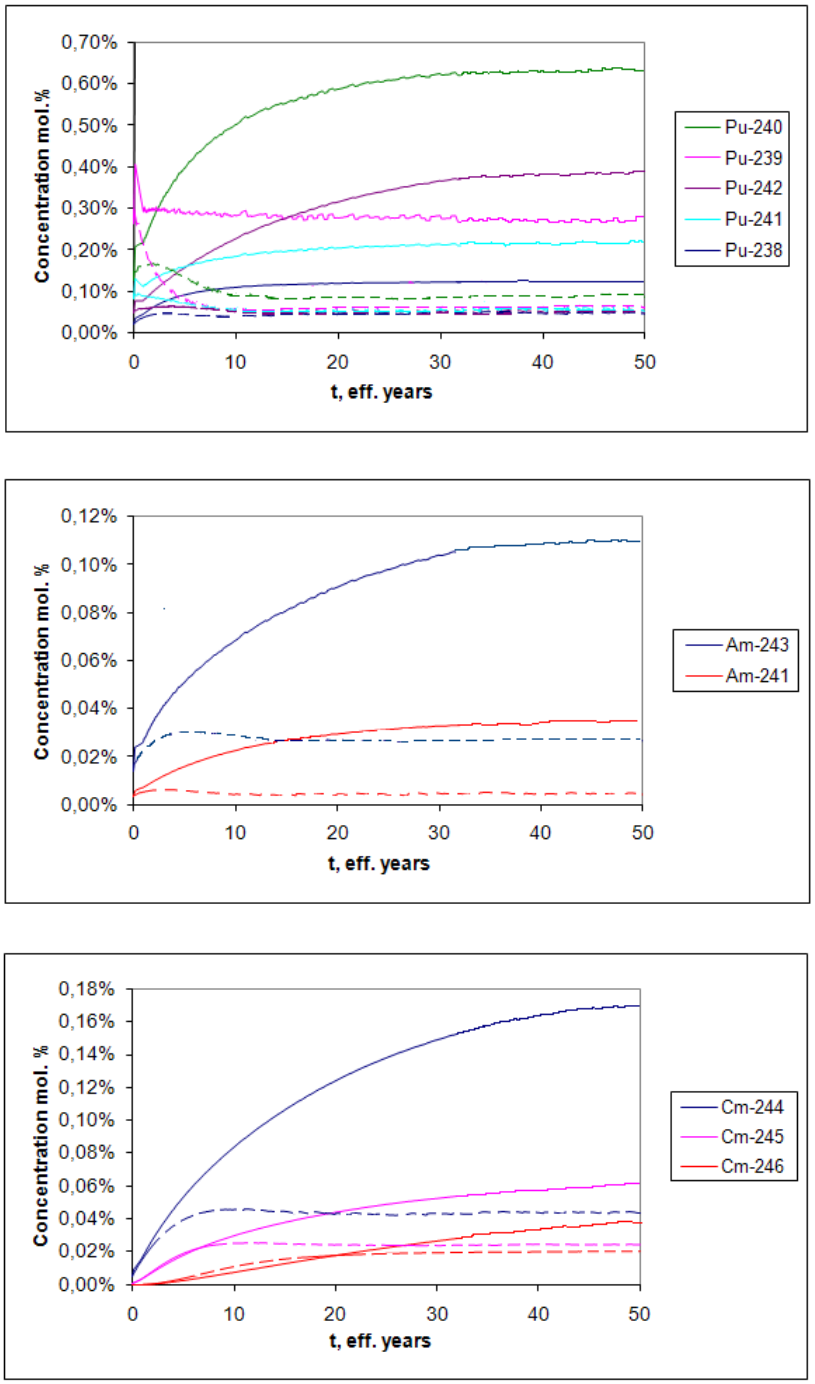


Figure 4: TRU loading and ^{233}U production for two-fluid ring core configuration

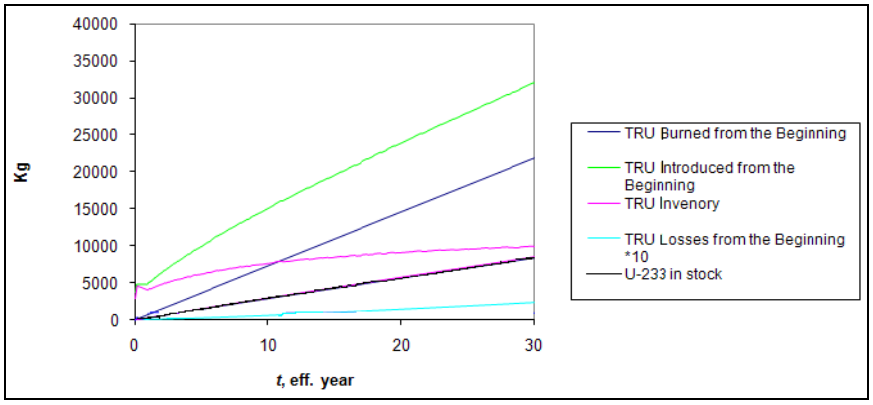


Figure 5: Critical $TRUF_3$ concentration required for transition to different feed loadings vs. Pu/(Np+MA) ratio in it

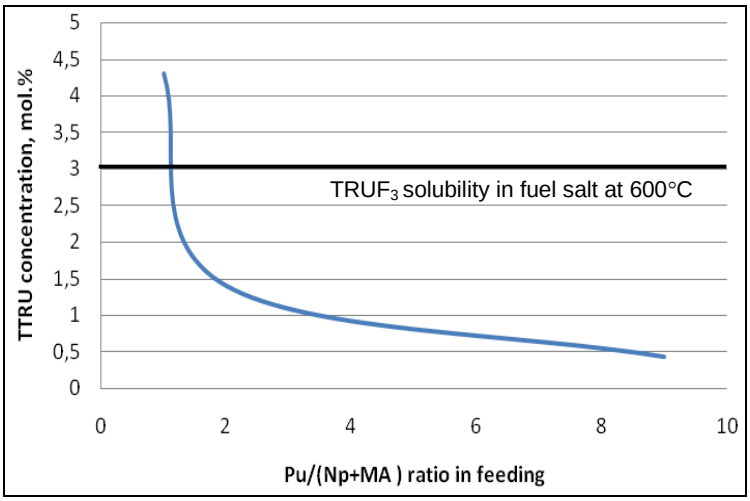
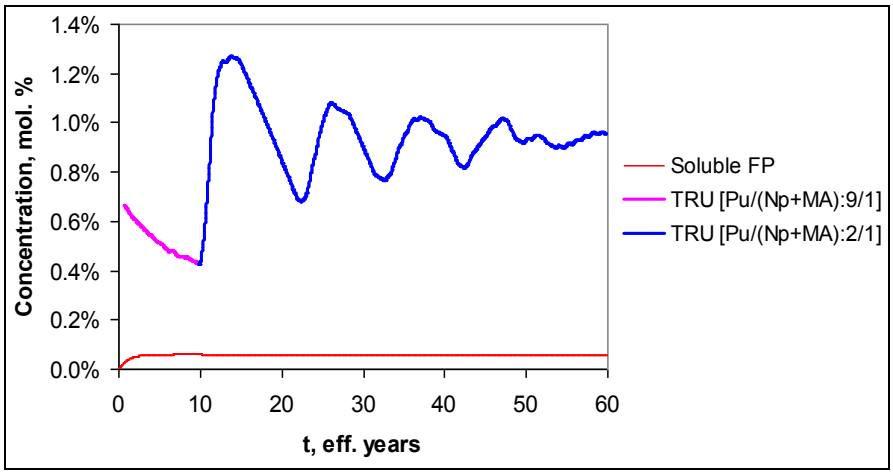


Figure 6: Concentrations of TRU and lanthanides trifluorides vs. time for ring core with fuel make up transition from Pu/(Np+MA) = 9 down to Pu/(Np+MA) = 2 after 10 years of operation



Breeder mode

After 10 years of MOSART operation with ^{233}U recycling from blanket to the fuel stream there is possibility for transition to breeding mode. This transition needs addition more than 10 mole % of ThF_4 in fuel salt and will lead to essential increase of critical $TRUF_3$ concentration in fuel salt. Calculation meanings of $TRUF_3$ concentrations necessary for transition for different molten salt solvents are collected in the Table 2. As can see required $TRUF_3$ concentration for transition to breeder mode in molten $80LiF\text{-}3BeF_2\text{-}12.5ThF_4$ salt mixture will be about 4.5 mole % (Figure 7). The doubling time for the same case will be 35-40 years (Figure 8).

Table 2: Critical $TRUF_3$ concentration in the fuel salt

Salt solvent system, mole %			Fuel addition, mole %
LiF	BeF_2	ThF_4	$TRUF_3$
73	12	11	4.0
73	5	16.6	5.4
83	2	10.9	4.1
80	3	12.5	4.5

Figure 7: $AnF_{3(4)}$ concentrations (in mole%) vs. operation time after transition to breeder mode in molten $80LiF\text{-}3BeF_2\text{-}12.5ThF_4$ salt mixture

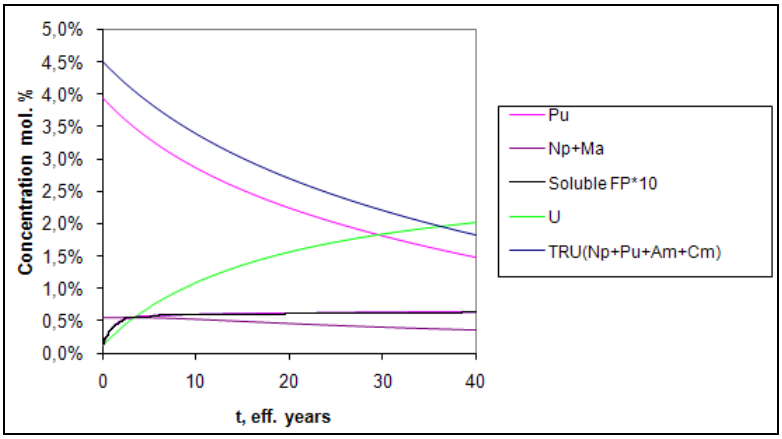
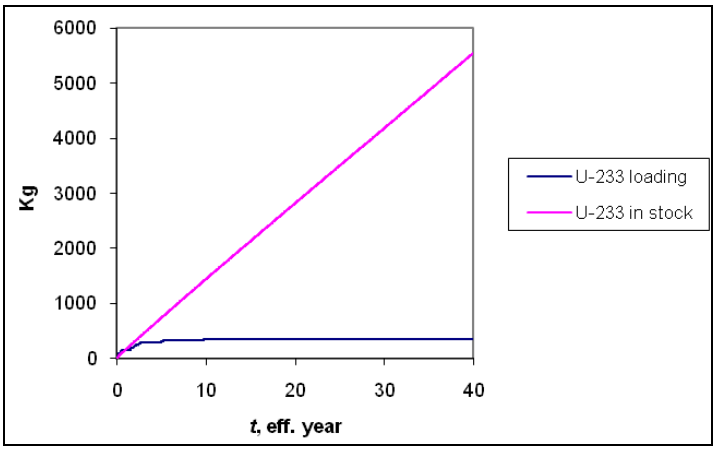


Figure 8: ^{233}U production in breeding mode



Core/blanket metallic materials

The metallic material used in constructing the primary circuit of a MSR will operate at temperatures up to 720 or even 800°C. The operating lifetime of a Gen-IV MSR should be in the range of 60 yrs, with a 90% load factor. Thus the metal must have moderate oxidation resistance, must resist corrosion by the salt, and must not be subject to severe embitterment by neutrons. In non moderated two fluid MSFR concepts walls separating fuel and blanket streams will present in the highest flux.

Early materials studies led to the development of a Ni-Mo alloy Hastelloy N with max ASME section VIII temperature of 704°C, for use with fluoride salts. The weakest point of this alloy is high-temperature embitterment under helium production as a result of neutron reactions (radiation-induced embitterment). The main reactions sources of He are: $^{58}Ni + n \rightarrow {}^{55}Fe + {}^{4}He$; $^{60}Ni + n \rightarrow {}^{57}Fe + {}^{4}He$; $^{10}B + n \rightarrow {}^{7}Li + {}^{4}He$ and two-stage reaction $^{58}Ni + n \rightarrow {}^{59}Ni + \gamma$; $^{59}Ni + n \rightarrow {}^{56}Fe + {}^{4}He$. Neutronic spectrum on the dividing walls surface is mixed (see Figure 9).

In case of modified Hastelloy N with decreased boron content, after five years in the most sensitive region (wall between the inner blanket and fuel salt streams in the central axial cross-section) will be produced about 60 appm of He (Figure 10). Specifying a critical level of helium that would cause embitterment is not presently possible. Experimental data are required to asses radiation induced embitterment.

Alternative structural alloys for MSFR core internals to expand upper operating temperature to 800°C would be low activation nanocomposited ferritic materials with Ni cladding. Longer-range alternative to the modification of existing commercial alloys is the design of completely new alloys utilising the principal of creating a high number density of stable nano-scale clusters or particles, thereby establishing a very high density of point defect recombination sites and traps for He. Efforts are in progress to adapt an approach similar to ferritic and ferritic-martensitic alloy systems to design and development of high strength, radiation-stable Ni-based alloys.

Figure 9: Neutron spectrum on the walls separating core and blanket streams

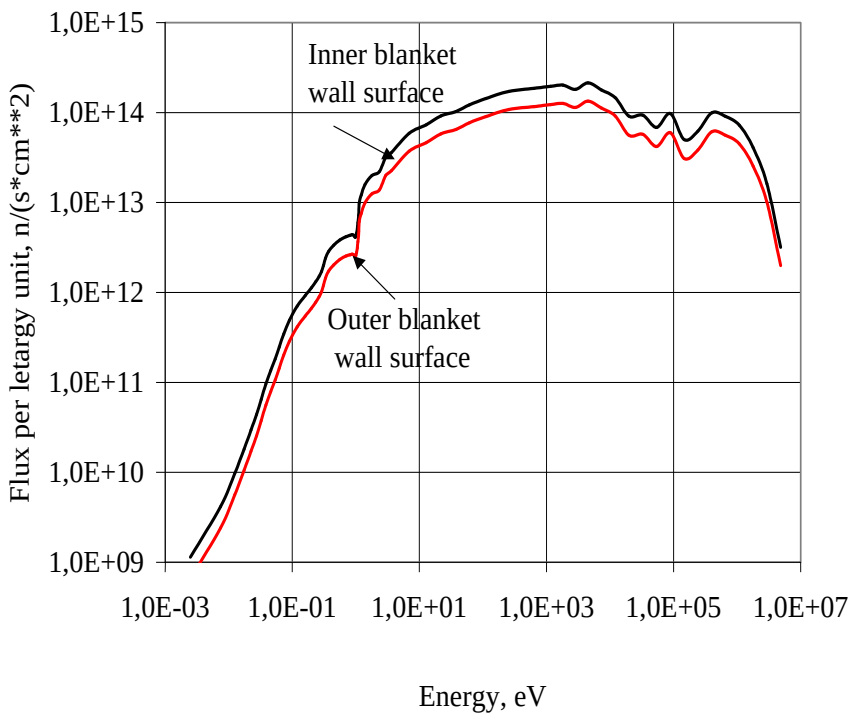


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Figure 10: He production in Ni-Mo alloy (B content ≤ 0.001%)

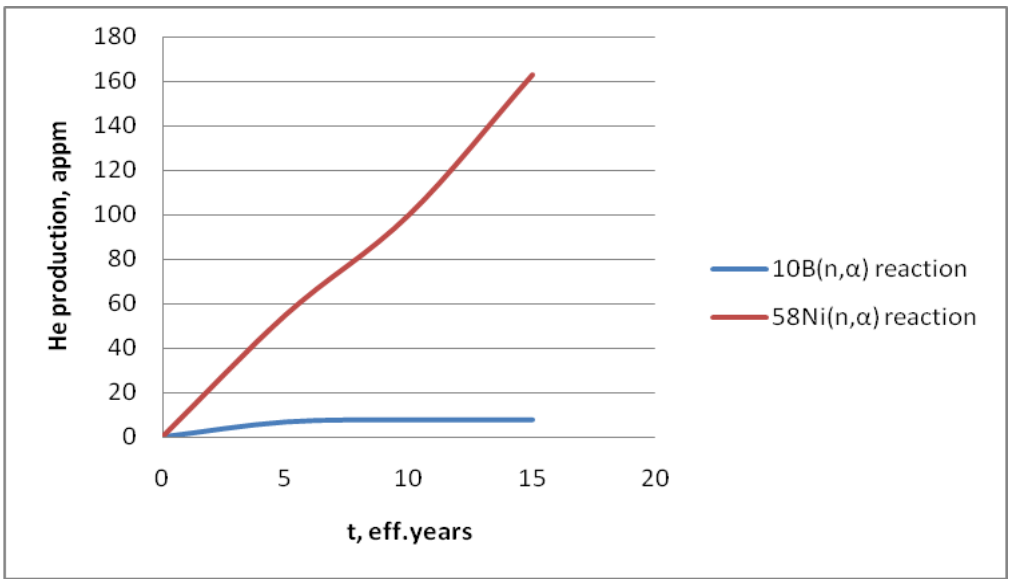

Table 3: Average neutron flux in the central axial cross-section and neutron reaction rates

Location	Energy group	Neutron flux, n/sm^2·s	Reaction	Reaction rates, barn·s^{-1}
Wall between core and inner blanket	E < 1 eV 1eV < E < 0.1 MeV E > 0.1 MeV Total	7.7E+12 1.3E+15 2.6E+14 1.6E+15	^{10}B(n,α) ^{58}Ni(n,γ) ^{58}Ni(n,α) ^{59}Ni(n,α) ^{60}Ni(n,α)	6.4E+16 9.4E+13 8.4E+11 1.9E+15 1.4E+11
Wall between core and outer blanket	E < 1 eV 1eV < E < 0.1 MeV E > 0.1 MeV Total	4.8E+12 8.5E+14 1.7E+14 1.0E+15	^{10}B(n,α) ^{58}Ni(n,γ) ^{58}Ni(n,α) ^{59}Ni(n,α) ^{60}Ni(n,α)	4.2E+16 6.0E+13 5.6E+11 1.2E+15 9.9E+10

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