Transmutation and Production Rates of Elements in Flibe and Flinabe with Impact on Chemistry Control

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DISCLAIMER

TRANSMUTATION AND PRODUCTION RATES OF ELEMENTS IN FLIBE AND FLINABE WITH IMPACT ON CHEMISTRY CONTROL

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ABSTRACT

Neutronics calculations were performed for blanket designs using the molten salts Flibe and Flinabe to determine the transmutation rates of constituent elements and the rates of production of other elements. At least from mass balance considerations no free fluorine will be left provided that the recombination reactions with freed Be, Li, Na, and tritium are fast enough. However, more than 95% of the tritium bred will be in the form of TF. In addition, O and N are produced. A REDOX reaction needs to be established to control the TF activities. The Be used for neutron multiplication can be used for the REDOX control to reduce TF to T_2. *The thermodynamics for the reaction between TF and Be is an important process to be demonstrated.*

I. INTRODUCTION

The molten salt Flibe has been considered as a breeding material and coolant candidate in fusion systems.[1-3] The low viscosity Flibe being widely considered consists of LiF and BeF_2 with molecular ratio of 2:1. It has the attractive features of low activation, low tritium retention, small density change on melting, low chemical reactivity with air and water, and low electrical conductivity which alleviates the MHD problems encountered in magnetic confinement fusion systems. In addition, Flibe has good neutron attenuation properties. On the other hand, it has a relatively high melting point (459°C) and low thermal conductivity resulting in reduced heat transfer capability.[4] In addition, tritium permeation caused by the low tritium solubility in Flibe is a major safety concern.[5]

Transmutation of the constituent elements leads to the production of the highly corrosive free fluorine. In addition, the less corrosive TF will be produced as a result of combination of some of the freed F and the generated tritium. While TF is compatible with some structural materials, free F is not compatible with any structural material. Controlling the activities of free F and TF is essential for the Flibe to be considered as a viable breeder/coolant candidate. In an earlier work, we performed preliminary calculations to determine the transmutation rates of the constituent elements of Flibe in a fusion neutron environment.[6] In this work, a more detailed analysis is performed for a 60-cm thick blanket that includes a 6-cm thick front multiplier zone required for tritium self-sufficiency. The structural material is the nano-composited ferritic (NCF) steel alloy 12YWT and the Be multiplier is used. Rates of all energetically possible nuclear reactions were calculated using the most recent FENDL-2 cross section data.[7] In addition to neutron destruction rates of constituent elements, the production rates of other elements were calculated.

The molten salt Flinabe that consists of LiF, BeF_2 and NaF with the molecular ratio of 1:1:1 has recently been proposed for beam line protection in Heavy Ion Beam fusion systems.[8] The phase diagram shows that the melting temperature of the Flinabe is only ~300°C and the vapor pressure would be low near the melting temperature. Flinabe was considered in the past as the breeding material but received less attention due to concerns with tritium breeding. New calculations suggested that, by adding about 12-cm thick front blanket zone that includes about 60% Be, tritium self-sufficiency can be achieved.[9,10] For comparison, Flibe needs about 6-cm thick front zone to achieve the same tritium breeding ratio (TBR). The thickness of the front Be zone can be reduced by enriching Li in ^{6}Li. As a result, Flinabe is currently being assessed as a breeder/coolant in fusion blankets. It is of particular interest in designs with liquid walls where it is desired to have a low vapor pressure to reduce plasma contamination and a low melting temperature to allow the blanket to operate at a wider temperature window. In this work, we perform detailed calculations for a fusion blanket utilizing Flinabe to determine the transmutation and production rates compared to those in Flibe. The impact on required chemistry control for Flibe and Flinabe is investigated.

II. TRANSMUTATION AND PRODUCTION RATES IN FLIBE

Calculations have been performed for a typical fusion blanket. We used the most recent FENDL-2 cross section data in 175 neutron energy groups.[7] The data include all partial reactions needed to determine the transmutation rates. The first wall (FW) and blanket design includes a 0.5-cm thick FW made of the NCF alloy 12YWT which is capable of operating at elevated temperatures up to about 800°C.[11] The front 6 cm of the blanket consists of 60% Be multiplier, 5% NCF structure, and 35% Flibe. The rest of the 60-cm thick blanket includes 5% NCF and 95% Flibe.

A representative 40-cm thick shield (80% NCF, 20% water) is included to adequately account for neutron reflection into the blanket. The low viscosity Flibe (Li_2BeF_4) is used with natural Li. The calculated local TBR for this design is 1.4.

Table I lists all possible neutron reactions with the constituents of Flibe (Be, F, ^{6}Li, and ^{7}Li). The reaction products are also listed. Decay products resulting from radioactive decay of radioactive reaction products are indicated. The (n,2n), (n,t), and (n,α) reactions result in the destruction of Be atoms producing He, T and Li. The (n,d), (n,t), (n,α), (n,n'p), (n,n'α), (n,2n), and (n,γ) reactions result in the destruction of F atoms producing He, H, T, O and Ne atoms. The (n,α), (n,n'd), and (n,2nα) reactions result in the destruction of ^{6}Li producing He, H, and T atoms. The (n,n'α), (n,γ), and (n,2nα) reactions result in the destruction of ^{7}Li. He, H, and T atoms are produced.

Table I. Transmutation Reactions for Flibe Constituents

Be
^{9}Be (n,2n) 2 ^{4}He
^{9}Be (n,t) ^{7}Li
^{9}Be (n,α) ^{6}He ($T_{1/2}$ = 807 ms) $\rightarrow$ ^{6}Li
^{9}Be (n,γ) ^{10}Be
F
^{19}F (n,p) ^{19}O ($T_{1/2}$ = 26.9 s) $\rightarrow$ ^{19}F
^{19}F (n,d) ^{18}O
^{19}F (n,t) ^{17}O
^{19}F (n,α) ^{16}N ($T_{1/2}$ = 7.13 s) $\rightarrow$ ^{16}O
^{19}F (n,n'p) ^{18}O
^{19}F (n,n'α) ^{15}N
^{19}F (n,2n) ^{18}F ($T_{1/2}$ = 1.83 h) $\rightarrow$ ^{18}O
^{19}F (n,γ) ^{20}F ($T_{1/2}$ = 11 s) $\rightarrow$ ^{20}Ne
^{6}Li
^{6}Li (n,α) ^{3}H
^{6}Li (n,2nα) ^{1}H
^{6}Li (n,n'd) ^{4}He
^{6}Li (n,p) ^{6}He ($T_{1/2}$ = 807 ms) $\rightarrow$ ^{6}Li
^{6}Li (n,γ) ^{7}Li
^{7}Li
^{7}Li (n,n'α) ^{3}H
^{7}Li (n,2nα) ^{2}H
^{7}Li (n,γ) ^{8}Li ($T_{1/2}$ = 0.84 ms) $\rightarrow$ ^{8}Be $\rightarrow$ 2 ^{4}He
^{7}Li (n,d) ^{6}He ($T_{1/2}$ = 807 ms) $\rightarrow$ ^{6}Li
^{7}Li (n,2n) ^{6}Li

Table II gives the destruction rates for the Flibe constituent elements. The results are given in units of atoms/s per cm^3 of Flibe and are normalized to 1 MW/m^2 neutron wall loading. The peak and average destruction rates are given. The peak to average ratio is the highest for F (6.9) where destruction is dominated by high energy neutrons and the lowest (5.4) for Li in which most of the transmutations are caused by relatively low energy neutrons. Table III lists the production rates for elements produced from the destruction of the Flibe constituent elements. Notice that in addition to T and He, a large amount of O and N is produced in the Flibe upon irradiation in a fusion system. O and N are produced at about 7% and 14% of the rate of tritium breeding. This should be taken into account when assessing the compatibility of Flibe with structural materials in the fusion radiation environment.

Table II. Destruction Rates (atoms/cm^3 s) for Flibe Constituent Elements

	Be	F	Li
Peak	7.23×10^{11}	2.34×10^{12}	7.73×10^{12}
Average	1.23×10^{11}	3.40×10^{11}	1.43×10^{12}

Table III. Production Rates of Elements Produced from Transmutation of Constituent Elements in Flibe

Be

	Atoms/cm^3 s	
	Peak	Average
T	1.46×10^{10}	1.67×10^{9}
He	1.32×10^{12}	2.20×10^{11}
Li	6.38×10^{10}	1.26×10^{10}

F

	Atoms/cm^3 s	
	Peak	Average
T	4.46×10^{10}	5.41×10^{9}
H (excluding T)	3.88×10^{11}	5.17×10^{10}
He	1.84×10^{12}	2.78×10^{11}
O	7.17×10^{11}	1.05×10^{11}
N	1.59×10^{12}	2.29×10^{11}
Ne	3.02×10^{10}	5.91×10^{9}

Li

	Atoms/cm^3 s	
	Peak	Average
T	7.59×10^{12}	1.41×10^{12}
H (excluding T)	1.64×10^{11}	2.53×10^{10}
He	7.74×10^{12}	1.43×10^{12}

Total in Flibe

	Atoms/cm^3 s	
	Peak	Average
T	7.65×10^{12}	1.42×10^{12}
H (excluding T)	5.52×10^{11}	7.70×10^{10}
He	1.09×10^{13}	1.93×10^{12}
Li	6.38×10^{10}	1.26×10^{10}
O	7.17×10^{11}	1.05×10^{11}
N	1.59×10^{12}	2.29×10^{11}
Ne	3.02×10^{10}	5.91×10^{9}

III. TRANSMUTATION AND PRODUCTION RATES IN FLINABE

The blanket design used is similar to that used for Flibe with the exception that the thickness of the front multiplier zone of the blanket is increased to 12 cm. This is needed because of the reduced tritium breeding capability of Flinabe compared to Flibe.[9,10] The Flinabe ($LiNaBeF_4$) is used with natural Li. The calculated local TBR for this design is similar to that for the Flibe design. In addition to the neutron reactions with Be, F, ^{6}Li, and ^{7}Li listed in Table II, all nuclear reactions with Na listed in Table IV were accounted for. The (n,α), (n,n'p), (n,n'α), (n,2n), and (n,γ) reactions result in destruction of Na atoms producing He, H, T, F, Ne and Mg atoms.

Table IV. Transmutation Reactions for Sodium in Flinabe

^{23}Na (n,p) ^{23}N ($T_{1/2}$ = 37.6 s) → ^{23}Na
^{23}Na (n,α) ^{20}F ($T_{1/2}$ = 11.4 s) → ^{20}Ne
^{23}Na (n,n'p) ^{22}Ne
^{23}Na (n,n'α) ^{19}F
^{23}Na (n,2n) ^{22}Na ($T_{1/2}$ = 2.6 y) → ^{22}Ne
^{23}Na (n,γ) ^{24}Na ($T_{1/2}$ = 15 h) → ^{24}Mg

Table V gives the destruction rates for the Flinabe constituent elements. The peak to average ratio is the highest for F and Na (6.3) and the lowest (4.3) for Li. The lower peaking in destruction rates in Flinabe is due to the softer neutron spectrum due to using twice as much separate Be multiplier. This is also reflected in about 12% lower destruction rates for Be and F and about 27% higher Li destruction rate in Flinabe as shown in Fig. 1. The production rates for elements produced from the destruction of the Flinabe constituent elements are given in Table VI. In addition to T and He, a large amount of O and N is produced in the Flinabe. The added Na in the Flinabe results in the production of additional modest amounts of Mg and Ne. Production of O and N in Flinabe is about 13% lower than in Flibe as shown in Fig. 2. On the other hand, a factor of 8 more Ne is produced in Flinabe. These results should be taken into account when assessing the compatibility with structural materials in the fusion radiation environment.

Table V. Destruction Rates (atoms/cm^3 s) for Flinabe Constituent Elements

	Peak	Average
Be	$5.91x10^{11}$	$1.09x10^{11}$
F	$1.89x10^{12}$	$2.98x10^{11}$
Li	$7.88x10^{12}$	$1.82x10^{12}$
Na	$3.75x10^{11}$	$5.93x10^{10}$

Table VI. Production Rates of Elements Produced from Transmutation of Constituent Elements in Flinabe

Be

	Atoms/cm^3 s	
	Peak	Average
T	$1.16x10^{10}$	$1.45x10^{9}$
He	$1.12x10^{12}$	$2.05x10^{11}$
Li	$5.29x10^{10}$	$1.12x10^{10}$

F

	Atoms/cm^3 s	
	Peak	Average
T	$3.54x10^{10}$	$4.72x10^{9}$
H (excluding T)	$2.46x10^{11}$	$4.49x10^{10}$
He	$1.49x10^{12}$	$2.43x10^{11}$
O	$5.79x10^{11}$	$9.19x10^{10}$
N	$1.28x10^{12}$	$1.99x10^{11}$
Ne	$2.93x10^{10}$	$6.92x10^{9}$

Li

	Atoms/cm^3 s	
	Peak	Average
T	$7.82x10^{12}$	$1.81x10^{12}$
H (excluding T)	$6.63x10^{10}$	$1.12x10^{10}$
He	$7.88x10^{12}$	$1.82x10^{12}$

Na

	Atoms/cm^3 s	
	Peak	Average
H (excluding T)	$2.17x10^{11}$	$2.96x10^{10}$
He	$1.11x10^{11}$	$1.66x10^{10}$
Ne	$2.92x10^{11}$	$3.95x10^{10}$
Mg	$7.78x10^{10}$	$1.92x10^{10}$
F	$5.33x10^{9}$	$5.93x10^{8}$

Total in Flinabe

	Atoms/cm^3 s	
	Peak	Average
T	$7.87x10^{12}$	$1.82x10^{12}$
H (excluding T)	$5.29x10^{11}$	$8.57x10^{10}$
He	$1.06x10^{13}$	$2.28x10^{12}$
Li	$5.29x10^{10}$	$1.12x10^{10}$
O	$5.79x10^{11}$	$9.19x10^{10}$
N	$1.28x10^{12}$	$1.99x10^{11}$
Ne	$3.21x10^{11}$	$4.64x10^{10}$
Mg	$7.78x10^{10}$	$1.92x10^{10}$
F	$5.33x10^{9}$	$5.93x10^{8}$

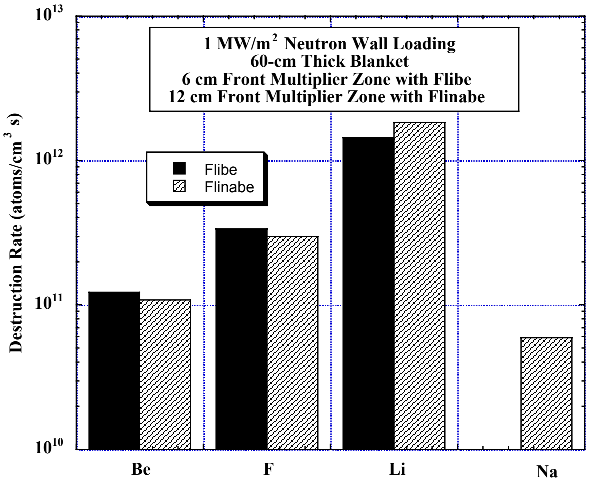

Fig.1. Comparison of Destruction Rates of Constituent Elements in Flibe and Flinabe.

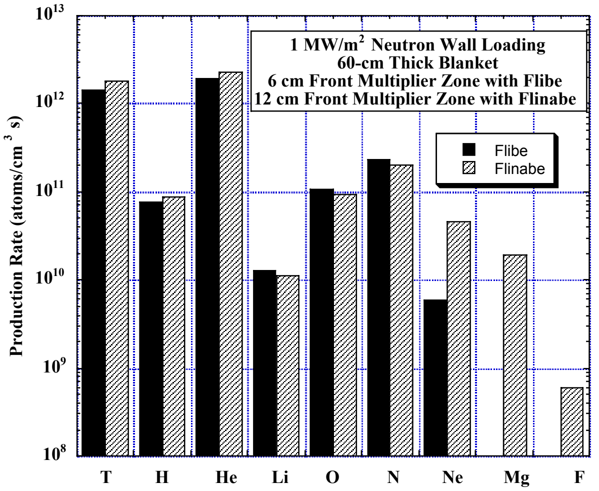

Fig. 2. Comparison between Elemental Production Rates in Flibe and Flinabe.

IV. ATOMIC BALANCE

The atomic balance of the transmutation products was evaluated to determine the expected amount of the corrosive free F and TF generated in a fusion radiation environment. Table VII depicts the atomic balance for the Flibe and Flinabe. The rates given are averaged over the 60-cm thick blanket and are normalized to 1 MW/m^2 neutron wall loading. Free tritium is produced as a result of tritium breeding in Li and (n,t) reactions with other constituent elements. Fluorine transmutation results in freeing Li atoms from LiF, Be atoms from BeF$_2$, and Na atoms from NaF. The free Li is incremented by the amount produced as a transmutation product of Be. For each Li atom transmuted a free F atom is produced from LiF. In addition, two F atoms are freed from BeF$_2$ for every Be atom transmuted. In Flinabe, more F is freed from NaF following Na transmutation. The free F generated will recombine with Li to form LiF, with Be to form BeF$_2$, and with Na to form NaF. The remaining free F will combine with the free tritium to form TF. The results in Table VII indicate that at least from mass balance considerations there will be no free F left. It remains to be investigated whether the kinetics of these combination reactions is fast enough. The results show that in the case of Flibe more than 95% of the T bred in the blanket will be in the form of TF. In the Flinabe blanket more than 98% of the T bred will be in the form of TF. These are very encouraging results since free F is very corrosive. Hence we only have to worry about chemistry control of the less corrosive TF.

Table VII. Atomic Balance of the Transmutation Products

	Atoms/cm^3 s	
	Flibe	Flinabe
Free T	1.42×10^{12}	1.82×10^{12}
Free Li from F transmutation	1.70×10^{11}	7.45×10^{10}
Free Li as transmutation product of Be	1.26×10^{10}	1.12×10^{10}
Total free Li	1.83×10^{11}	8.57×10^{10}
Free Be from F transmutation	8.50×10^{10}	7.45×10^{10}
Free Na from F transmutation	--	7.45×10^{10}
Free F from Li transmutation	1.43×10^{12}	1.82×10^{12}
Free F from Be transmutation	2.46×10^{11}	2.18×10^{11}
Free F from Na transmutation	--	5.93×10^{10}
Free F as transmutation product of Na	--	5.93×10^{8}
Total free F available	1.68×10^{12}	2.10×10^{12}
F combined with Li to form LiF	1.83×10^{11}	8.57×10^{10}
F combined with Be to form BeF$_2$	1.70×10^{11}	1.49×10^{11}
F combined with Na to form NaF	--	7.45×10^{10}
F left combining with T to form TF	1.33×10^{12}	1.79×10^{12}
Free T left	7.00×10^{11}	3.00×10^{10}

The calculations were performed for designs with natural Li. Enriching the Li in ^{6}Li enhances tritium breeding with the TBR maximizing at 40-50% ^{6}Li.[9] We assessed the impact of Li enrichment on the atomic balance of transmutation products. Calculations were performed for a Flibe blanket with 40% ^{6}Li and a reduced multiplier zone of 4 cm thickness. This design yields the same TBR as in the design with natural Li. Lithium enrichment was found to have only a minor impact on the mass balance with the conclusion regarding free F and TF being the same.

V. CHEMISTRY CONTROL

The chemistry control process is to control the activities of the TF and free F. Both TF and free F are corrosive to many structural materials. A possible method is to use the Be in the blanket, and maybe additional Be in the heat transport loop, to reduce both TF and F_2 to BeF_2 and T_2. The free energy of formation for the TF and BeF_2 are -66.2 kcal/g-atom of fluorine and -106.9 kcal/g-atom of fluorine, respectively, at 1000 K.[12] Therefore, from a thermodynamics point of view the chemical reaction of

$$Be + 2TF \rightarrow BeF_2 + T_2$$

will proceed. Based on this reaction, the equilibrium TF concentration in the Flibe will be about 10^{-12} mole fraction. At this very low concentration Flibe will not react with any structural material. However, it is important to demonstrate the kinetics of this reaction so that the proper chemistry condition can be reproduced in the experimental program. This along with the transmutation rates calculated here define the experimental parameters to study the thermodynamics and kinetics in the experiment being set up at INEEL under the JUPITER-II collaboration.

VI. SUMMARY AND CONCLUSIONS

One major concern with using Flibe and Flinabe in fusion blankets is that transmutation of the constituent elements leads to the production of the highly corrosive free F and the relatively less corrosive TF. Controlling the activities of free F and TF is essential for consideration as viable breeder/coolant candidates. Neutronics calculations were performed for blanket designs using either Flibe or the low melting point Flinabe to determine the transmutation rates of constituent elements and the rates of production of other elements. The results showed that at least from mass balance considerations no free F will be left provided that the recombination reactions with freed Be, Li, Na, and produced tritium are fast enough. However, more than 95% of the tritium bred will be in the form of TF. The results indicate also that O and N are produced at about 7% and 14% of the rate of tritium breeding. Enrichment of Li was found to have minor impact on mass balance with the conclusions remaining the same. The results imply that we only have to worry about chemistry control of the less corrosive TF. Be can be used to reduce both TF and F_2 to BeF_2 and T_2.

The transmutation rates and the thermodynamics define the chemistry conditions of the Flibe and Flinabe under neutron irradiation. These conditions need to be reproduced in the experimental setup at INEEL to study the thermodynamics and kinetics of the REDOX reaction. After the REDOX condition is established, the material corrosion experiment can be carried out to assess the compatibility between the molten salt, with proper chemistry control, and different structural materials.

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