Transmutation of Transuranic Elements and Long Lived Fission Products in Fusion Devices

Y. Gohar

Fusion Power Program
Technology Division
Argonne National Laboratory
9700 S. Cass Avenue, Argonne, IL 60439, USA

International Workshop on Blanket and Fusion Concept for the Transmutation of Actinides
General Atomics, San Diego, California, U.S.A.
21-23 March 2001

FUSION power program

TECHNOLOGY DEVELOPMENT

Transmutation of Transuranic Elements and Long Lived Fission Products in Fusion Devices

Objective

Develop a complete, economical, attractive, proliferation resistant solution for the pressing problems of disposing of spent nuclear fuel, transuranic elements, and highly enriched uranium. Such solution is developed to compute favorably with the other options under consideration.

Complete

- **The transuranic elements and the highly enriched uranium isotopes are utilized without leftover to store or guard.**
- **The long-lived fission products are transmuted to eliminate the need for its storage.**

Economical

- **The energy content of the transuranic elements and the highly enriched uranium is fully utilized.**
- **The generated energy produces revenue for the system.**
- **The required new resources are relatively reduced to improve the lifecycle cost.**
- **The R&D requirements are significantly minimized to reduce the total cost and the deployment time.**

Transmutation of Transuranic Elements and Long Lived Fission Products in Fusion Devices
(Continued)

Attractive

- The required D-T fusion power is very small, which can be realized with the current technologies.
- The volume of the radioactive waste generated from the system is relatively small.
- Such fusion devices provide the opportunity to develop fusion energy for the future.
- This application enhances the public acceptance of the fusion energy.
- The requirements of the geological repository site are significantly reduced. The site is used to dispose of the radioactive waste streams produced during the operation.

Proliferation Resistance

- Minimize pure high fissile material streams. Pure high fissile material streams are the main concern with respect to proliferation resistance.
- Enhance intrinsic barriers to proliferation such as material attractiveness and ease of recovery.

Material Inventory

- Spent Nuclear fuel
  - In the United State, about 70,000 tons of spent fuel will be accumulated by the year 2015 (67,000 tons uranium, 600 tons TRU, and 2400 tons of fission products).
  - At the present time, the cumulative amount of plutonium discharged from the world's civilian reactors is about 1400 tons.
- Excess Weapons Materials
  - The agreed arms reduction agreements between the United State and Russia resulted in more than 100 tons of excess weapons plutonium.
  - About 1400 tons of highly enriched uranium was generated during the cold war.

Transmutation Options

Neutron Spectrum

Fast neutrons have neutronics advantages for transuranic transmutation:

- **Transuranic elements have better ratio of fission to parasitic capture for fast neutrons than for thermal neutrons.**
- **Neutron loss to the fission products is relatively small for fast neutrons.**
- **Power peaking is less for fast neutrons.**
- **Neutron leakage is thermalized for transmuting the long-lived fission products (Tc^{99}, I^{129}, etc.).**

Coolant

Molten salts, liquid metals, and helium are the appropriate coolants for achieving fast neutron spectrum. Fusion blankets using these coolants are under development for fusion energy systems:

Flibe	**Breeder and coolant**
Lithium Lead Eutectic	**Breeder and coolant**
	Breeder with other coolants
Helium	**Coolant**
Bismuth Lead Eutectic	**Fission reactor coolant**
	Spallation target material for Accelerator Transmutation of Waste (ATW)

Transmutation Options
(Continued)

Fusion Blanket Concepts

Fusion blanket concepts operating at optimal transmutation rate require the following features:

- Constant content of the transuranic elements

The large change in the transmutation rate during operation, the desire to operate at constant output power, and the need for high transmutation rate require constant transmutation rate. This can be achieved by maintaining constant material composition or increase the fusion power to compensate for the material composition change.

- High availability for enhanced economics

Minimizing the down time for loading and shuffling transuranic materials, burnable poison loading, and blanket maintenance contributes significantly to this objective.

- Increase or eliminate the transuranic burnup limit

This eliminates the processing requirements to extract the unutilized transuranic elements or reduces the initial inventory of the transuranic elements for once through operating scenario. Also, it increases the blanket availability for power generation.

This requires blanket concepts with mobile transuranic materials. Molten salts, liquid metal eutectics, or pebble bed blanket concepts can operate in this mode.

Why Fusion?

- Fusion provides the optimum neutron source for such application. The spatial distribution, the neutron energy spectrum, and the neutrons intensity are quite satisfactory for transuranic transmutation.

- The total lifecycle cost is extremely competitive with the other options under consideration (MOX fuel program, ATW program, etc.).

- Modest fusion requirements are needed to solve the transmutation problem in a reasonable time.

- Can be demonstrated with small driven fusion devices using the current technologies.

- Japan, Europe, and Russia are using the transuranic elements for energy production, which fusion will achieve.

- These fusion devices will provide an opportunity to develop fusion energy for the future.

- This application will enhance the public acceptance of the fusion energy with respect to the environmental impact.

Flibe Blanket Concept

- **Molten fuel salt (Flibe) was used for fission reactors. Also, Flibe is under development for fusion reactors and it is an option for the Accelerator Transmutation of Waste (ATW) program.**

- **Flibe technologies were developed at ORNL for the MSBR program in the 60's and at ANL for molten salt processing program in the 90's. Also, the fast breeder program at ANL developed molten salt technologies for the IFR fuel cycle in the 90's.**

- **Uranium and transuranic fluorides are dissolved in the flibe of the self-cooled blanket concept, which simplifies the geometrical configuration.**

- **As a design option, a separate coolant can be used to cool the slowly circulating Flibe.**

- **Chemical control methods are required to insure compatibility with the structural material and low tritium permeation rate from the blanket system (two methods were successfully tested for fluoride salt).**

Flibe Blanket Concept
(Continued)

Flibe blanket concept has several attractive features for this application including:

- **The blanket achieves the elimination goal, the top rated solution for disposing the transuranic elements and the highly enriched uranium.**
- **The thermal power can be maintained at a constant level by adjusting the flibe composition.**
- **This blanket concept operates at a low-pressure, which simplifies the mechanical design.**
- **Development and fabrication costs of the transuranic and the uranium materials are eliminated.**
- **Burnup limit due to radiation damage is eliminated.**
- **Heat is generated within the flibe material, which simplifies the heat removal process.**
- **Flibe has a negative temperature coefficient related to the blanket reactivity, which enhances the safety performance.**
- **The operational record of the molten salt (Flibe) reactor at ORNL was successful. Uranium, thorium, and plutonium were utilized for generating power, which accumulated operational experience.**
- **Flibe are chemically and thermally stable under reactor operating conditions, which minimize the radioactive waste generation.**
- **The liquid blanket concept minimizes the total radioactive waste relative to the other blanket options.**

Flibe Molten Salt Blanket Concepts

- **Flibe molten salt (Li_2BeF_4) is well characterized for nuclear application with U, Th, and Pu.**
- **Physical Properties**

– Melting point, °C	459.1
– Thermal conductivity, W/cm. °C	0.010
– Viscosity, centipoises	0.116 exp [3755/T(°k)]
– Electrical conductivity at 500 , ohm^{-1}cm^{-1}	9.2 10^{-3}
– Heat capacity, cal/g. °C	0.57
– Density, g/cm^3	2.214 - 4.2 x 10^{-4} T(°C)

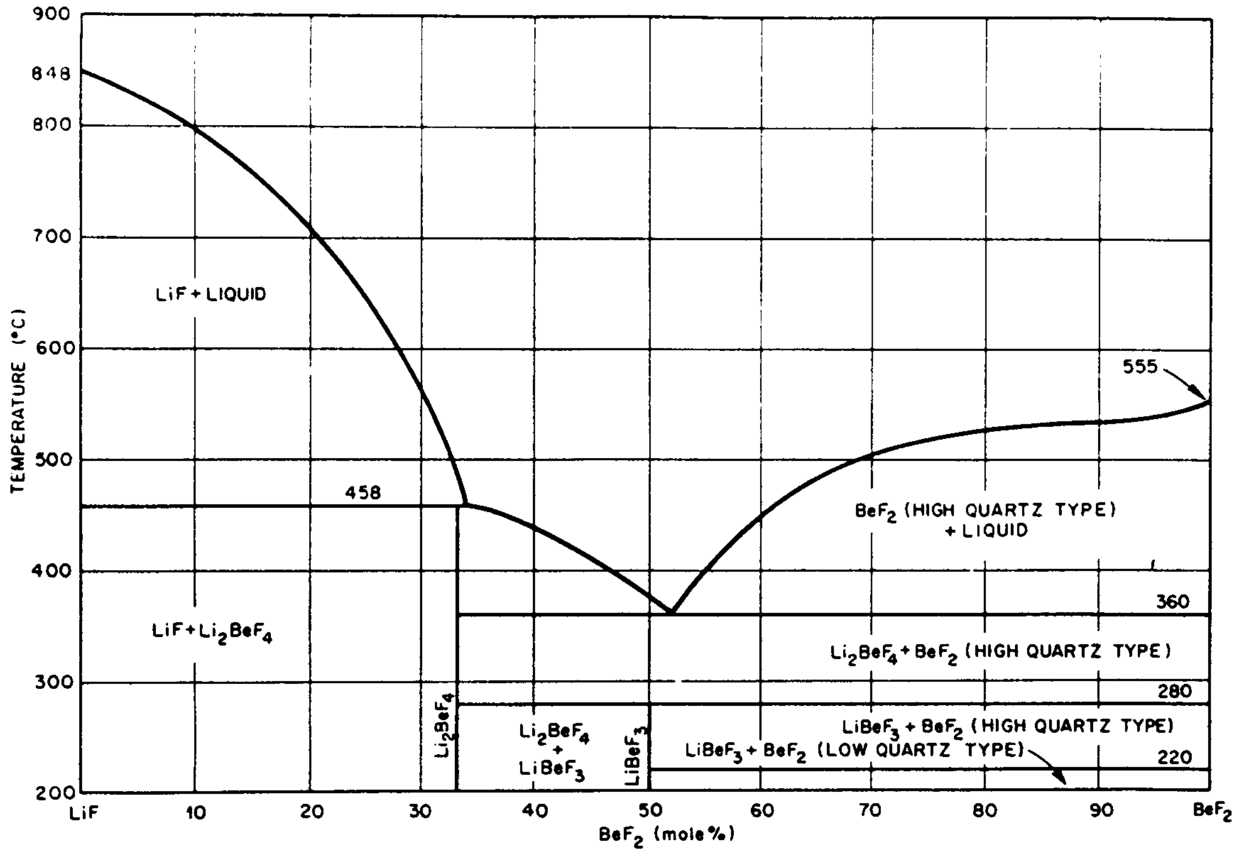
- **Flibe Chemistry with plutonium and fission products was studied for the MSBR in the 60's.**
  - **PuF_3 is the most suitable form to use in the Flibe.**
  - **PuF_4 has higher solubility than PuF3 in the Flibe but it is strongly oxidizing.**

$$Cr + 2PuF_4 \rightarrow CrF_2 + 2PuF_3$$

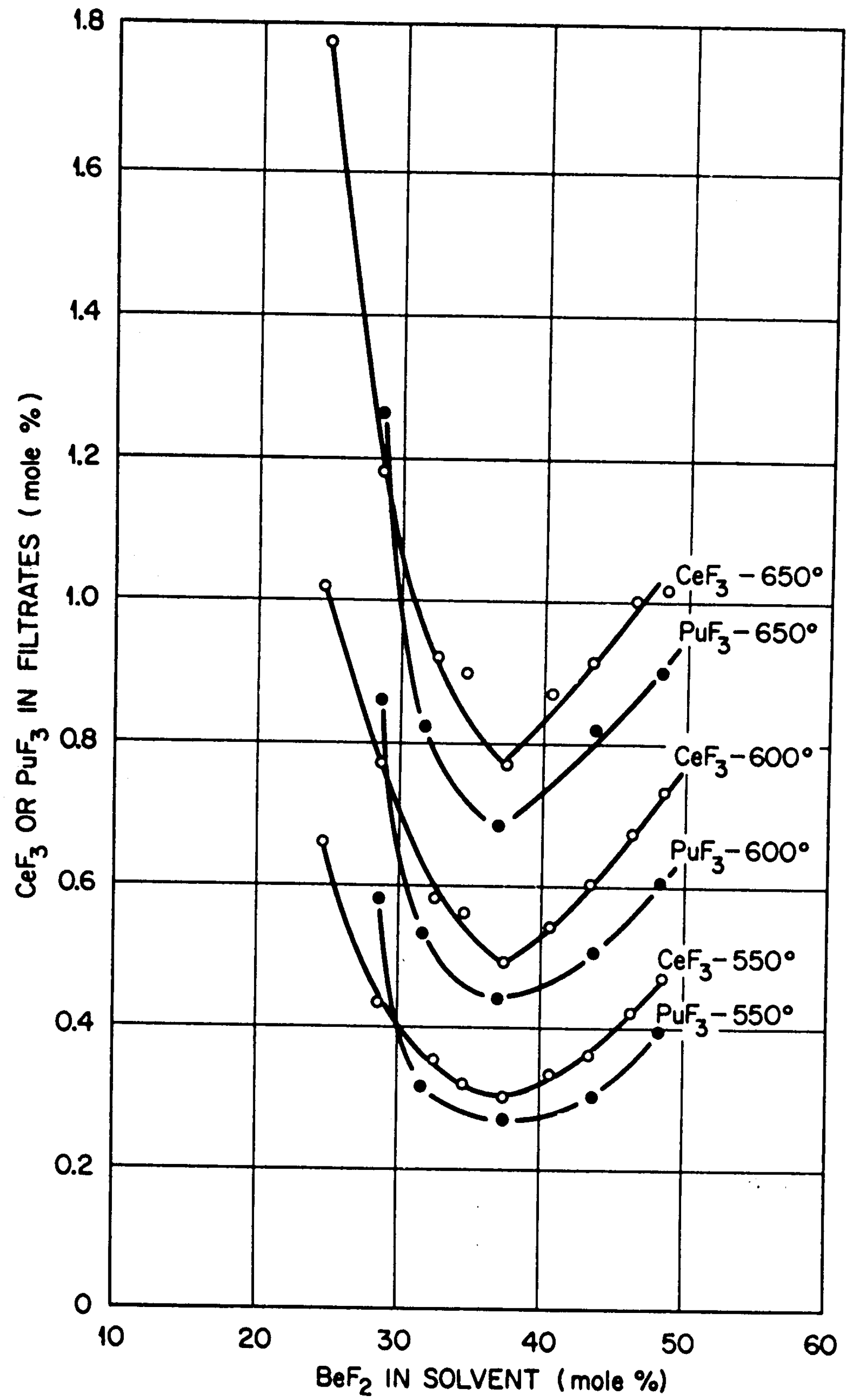
  - **PuF_6 gas.**
- **PuF_3 solubility in LiF-BeF_2 system was measured for compositions ranging in BeF_2 from 28.7 to 48.3 mole in the temperature range of 450 to 650 °C**
- **PuF_3**

Density, g/cm3	9.32
Melting point, °C	1425

LiF – BeF_2 Phase Diagram



Solubility of CeF_3 and PuF_3 in LiF-BeF_2 molten salt at different temperatures – From ORNL-TM-2256.



Liquid Metal Eutectic Blanket Concepts

- **Liquid metal eutectics are considered as coolant, tritium breeder, and uranium and transuranic carrier similar to Flibe.**

- **The main advantage of the liquid metal eutectics is the low melting point relative to Flibe, which significantly improves the structural material performance and simplifies the thermal hydraulic design. In addition, the use of beryllium and fluorine is eliminated.**

- **This blanket type has harder neutron spectrum, which improves the blanket neutronics performance.**

- **Austenitic and ferritic steels can be used as structural material.**

- **A coating is preferred for this concept to operate satisfactory. The development is underway for the lithium and lithium-lead fusion blanket concepts. A self-healing Oxide coating is developed for structural material corrosion control for the bismuth-lead spallation targets.**

- **Initial examination shows that lead and lithium have adequate solubility for uranium and transuranics.**

Computational Tools at ANL

- MONK Code: Monte Carlo transport code for lattice, core, shield, and burnup analyses. The burnup capabilities are integrated in the code. Several nuclear data libraries are available for MONK analyses:
  - ENDF/B-VI and JEF 2.2 evaluated nuclear data files.
  - Quasi-continuous energy libraries (13193 or 8220 groups).
  - Multigroup libraries (172 and 69 groups).
  - Explicit representation of the fission products where about 99% of the neutronically important process are represented in the library.
- MCNP/REBUS Codes
  - Transport and depletion capabilities.
  - Libraries based on ENDF/B-VI.
- DRAGON Code: Collision probability lattice code
  - Lattice and depletion capabilities.
  - 69-group library based on ENDF/B-VI.
- DRAGON/DIF3D/REBUS and DANTSYS Codes: Deterministic calculations
  - Multi-group libraries based on ENDF/B-VI.
  - Lattice, core, and burnup capabilities.

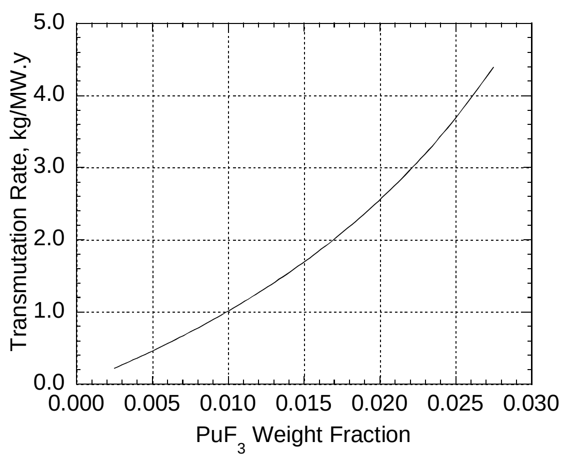
Analyses of the Flibe Blanket Concept

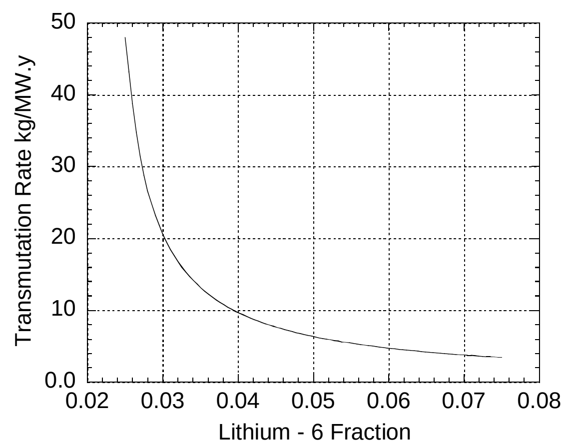
- Assessment was performed to define the Flibe blanket configuration and characterize its performance including material, neutronics, and thermal-hydraulic aspects.

- Blanket radial thickness, PuF_3 concentration in Flibe, and lithium-6 fraction in lithium were studied parametrically to define the blanket performance parameters including:

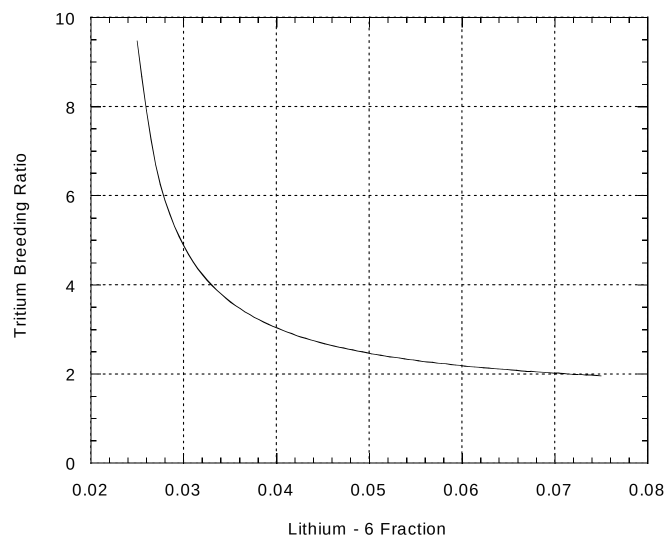
  – Pu transmutation rate, kg/MW.y of fusion power
  – Blanket energy multiplication factor
  – Total energy multiplication factor
  – Local tritium breeding ratio
  – Blanket shielding characteristics
  – System reactivity

- Plutonium transmutation rate as function of the PuF_3 weight fraction and the lithium-6 fraction in the Flibe molten salt:

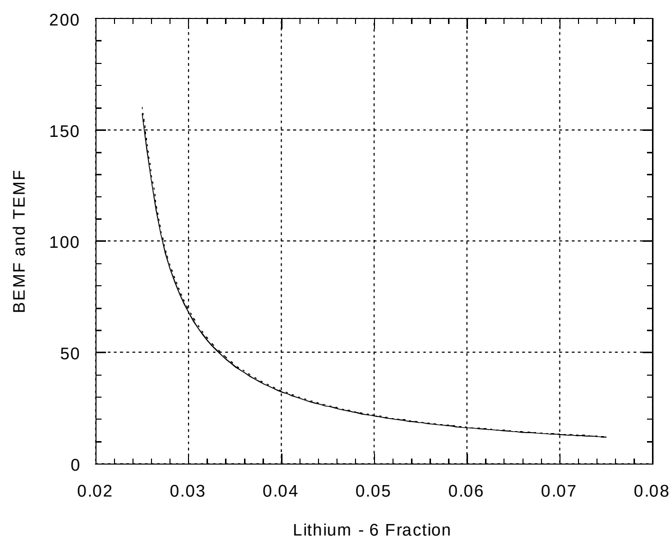




Analyses of the Flibe Blanket Concept
(Continued)

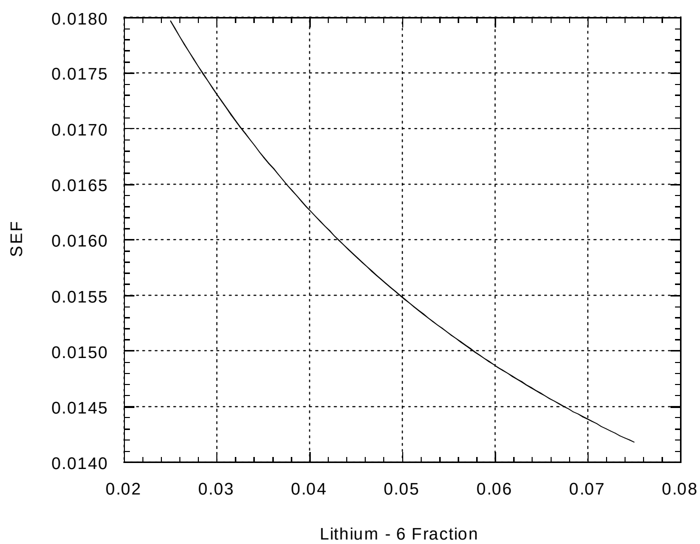


Tritium breeding ratio as function of the lithium-6 fraction.

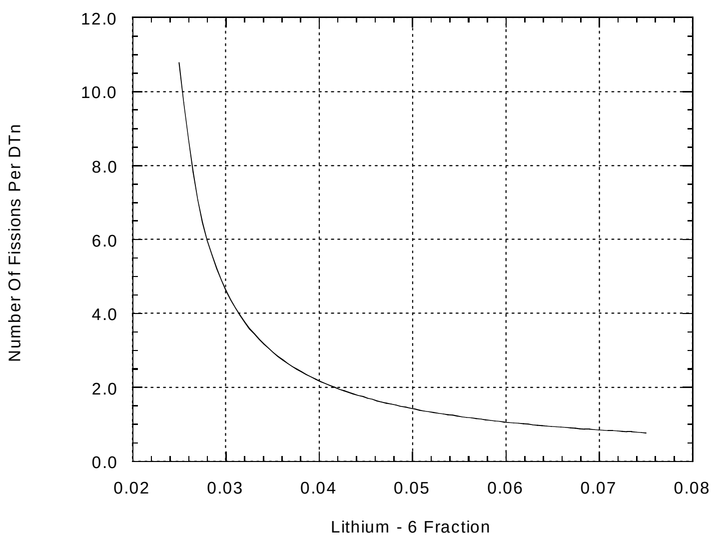


Blanket energy multiplication factor and total energy multiplication factor as function of the lithium-6 fraction.

Analyses of the Flibe Blanket Concept
(Continued)



Shield energy fraction as function of the lithium-6 fraction.



Number of fission reactions per fusion neutron as function of the lithium-6 fraction.

Flibe Blanket Performance Parameters With Different Plutonium Weight Fractions And Lithium-6 Enrichments

PuF_3 weight fraction	0.0025	0.0275	0.024	0.024	.00051	0.0056
Lithium-6 enrichment	Natural	Natural	2.5	Natural	0.0	0.25
Blanket energy multiplication factor	1.765	15.052	157.2	11.98	242.6	264.0
Total energy multiplication factor	1.819	15.262	160.0	12.15	251.5	270.9
Local tritium breeding ratio	1.214	2.180	9.469	1.956	0.488	10.740
Shield energy fraction	0.030	0.014	0.018	0.014	0.0352	0.0252
Fission reactions per D-T neutron	0.049	0.986	10.78	0.770	16.301	17.902
Pu transmutation rate, kg/MW.y	0.219	4.390	47.98	3.428	72.560	79.689
System Reactivity	0.125	0.741	0.969	0.691	0.979	0.981

Simple Poloidal Flibe Blanket Configuration

Blanket Configuration

First wall:	Type 316 stainless steel – 0.5 cm thick
Breeder/coolant:	Flibe with PuF_3 – 50 cm
Structure:	Type 316 stainless steel – 1.0 cm thick
Shield:	Steel shield (80% Type 316 Austenitic steel and 20% H_2O)

Blanket Parameters

Blanket thickness, cm	51.5	51.5
Lithium – 6 fraction	0.0	0.0025
PuF_3 weight fraction	0.00051	0.0056
Blanket energy multiplication factor (BEMF)	242.6	264.0
Local tritium breeding ratio (TBR)	0.488	10.74
Transmutation rate, kg/MW.y	72.56	79.69
Shield energy fraction	0.035	0.025
Neutron wall loading, MW/m^2	0.1	0.1
Blanket poloidal length, m	5	5
Surface heat flux, MW/m^2	0.025	0.025
Flibe temperature change, °C	100	100
Flibe velocity, m/s	1.06	1.15
Flibe inlet and outlet from the top		

Conclusions

- **Fusion blankets with liquid carrier for the transuranic elements and the uranium isotopes achieve the elimination goal, which is the top rated solution for the disposition of the spent nuclear fuel, the transuranic elements, and the highly enriched uranium.**

- **This type of blankets achieves a transmutation rate up to 80 kg/MW.y of fusion power.**

- **The energy from the transmutation process is utilized to produce revenue for the system.**

- **This fusion solution reduces the requirements for a geological repository, which is a major advantage.**

- **Flibe molten salt, liquid lead, lithium-lead eutectic, and bismuth-lead eutectic are identified as the most promising liquids for this application. Fusion blankets using these materials are under development for power reactors with the exception of bismuth-lead eutectic, which is being developed for spallation targets.**

- **A 334-MW of fusion power for thirty years with an availability factor of 0.75 can dispose of the 70,000 tons of the US inventory of spent nuclear fuel generated up to the year 2015. A fusion device with a few MWs of fusion power can demonstrate this application.**

Conclusions
(Continued)

- **This application will provide an excellent opportunity to develop the fusion energy for the future by building small fusion devices.**

- **These fusion devices have an operating flexibility, which can be achieved by adjusting the blanket parameters to maintain an acceptable thermal output for generating revenue. The small fusion power and the operating flexibility provide a mechanism to develop the fusion energy for the future.**

- **Further analyses and studies are needed to develop such system considering previous work including non-fusion options.**