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Options for Burning LWR SNF in LIFE Engine

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We have pursued two processes in parallel for the burning of LWR SNF in the LIFE engine: (1) solid fuel option and (2) liquid fuel option. Approaches with both are discussed below. The assigned Topical Report on liquid fuels is attached. Ralph is the lead author, and substantial contributions by Kaufman, Turchi and Shaw have been made.

Solid Fuel Option

The most logical solid fuel option would leverage as much of the DUPIC fuel cycle as possible (Figure 1). As described in the literature, the key process of the DUPIC fuel cycle is the oxidation and reduction of PWR spent oxide fuel (known as OREOX) to prepare powder for CANDU fuel fabrication. This is a completely dry process without any separation of fissile isotopes from the spent PWR fuel.

It is assumed that the composition of PWR spent oxide fuel fed to the OREOX process consists of U (1,943 kg), Pu (18.1 kg), Np (1.06 kg), Am (1.20 kg), Cm (0.04 kg), and miscellaneous fission products (73.4 kg). The composition of the powder leaving the OREOX process for fabrication of the CANDU fuel consists of U (1,924 kg), Pu (17.9 kg), Np (1.05 kg), Am (1.19 kg), Cm (0.04 kg), and miscellaneous fission products (52.3 kg). This would also be the composition of dry powder available from PWR SNF for fabrication of LIFE fuel.

The primary waste stream coming from the DUPIC fuel fabrication process consists of metallic components from spent PWR fuel, fission gases, and semi-volatile fission products released from the fuel during treatment. Noble gases such as Kr and Xe are compressed in 50-litre cylinders for long-term storage and decay. Tritium and carbon are trapped on molecular sieves and barium hydroxide, respectively. These are then mixed with cement, which is poured into large drums for disposal. The discarded cladding (hulls) are also mixed with cement for disposal. Radioactive iodine is trapped on a silver zeolite and Cs and Ru are fixed on filters and vitrified for disposal.

During the past 6 months, computational models for TRISO and SHC (solid hollow-core fuel) have been developed at LLNL, which account to the extent possible for the effects of fuel irradiation on stress and failure. Predictions for enhanced TRISO particles and SHC pebbles indicate that stresses, due to fission gas accumulation, thermal gradients and irradiation-induced swelling of the materials can be maintained below those that would lead to failure of the pressure boundary (Figures 2 and 3). Even so, the materials team remains skeptical about the ability of any TRISO fuel to reach 99.9% FIMA. Based upon their polled opinions, they seem to have greater confidence in the (un-tested) SHC design. They are also intrigued about the possiblity of using an inert matrix fuel, though very little data at high burn-up exists for this option.

A consensus seems to be that at extreme levels of radiation damage, the fuels will be "reduced to powder." Direct use of the powder from the OREOX process without consolidation appears to provide an interesting avenue of pursuit. These powdered or granular fuels could be immobilized in a porous metallic matrix that would enable fission gas release for capture. If desired, this fuel form could also be encapsulated with an appropriate cladding, thereby forming a pressure boundary for fission gas containment as envisioned with both TRISO and SHC. Sacrificial materials and diffusion barriers will have to be used, as now proposed in the TRISO, to prevent attack of the containment materials by palladium and other fission products that would from low-melting eutectics or solid solution. Alternatively, the powders could be contained loosely in a spherical (or other shaped) shell, that would also serve as a pressure boundary for fission gas containment. With loose powders, a much finer powder with high thermal conductivity would be added as a filler to promote good thermal conductivity in the interstitial spaces. While the swelling of graphite layers in a fabricated fuel may be problematic, the use of graphite powders would probably not be precluded as filler. It is possible that the powders could sinter into a solid form over time, however, this is not considered to be problematic.

In addition to the direct incorporation of OREOX powder into solid LIFE fuels, it would also be possible to build spherical fuel assemblies with plenum volumes for the containment of powder and the collection of released fission gas. This is yet another engineering option worthy of investigation.

Liquid Fuel Option

The liquid fuel option would avoid problems associated with radiation damage of fuels at high burn-up, but is challenged by the need for continuous on-line processing to remove fission products. Noble fission gases must be removed and stored in canisters for disposal; metallic fission products must be removed to prevent them from plating out in the primary circulation loop; rare earths must be removed so that the solubility of plutonium can be maintained at a relatively high level. Such liquid fuels require oxidation-state (redox) control, which can be accomplished with active electronic systems (adder potentiostats).

Molten salt fuels have been considered for graphite-moderated molten-salt breeder reactors that operate on the thorium-uranium fuel cycle, and enable continuous reprocessing of the fuel (Table 1). Reprocessing is used to keep fission products at desirable levels. A widely referenced fuel salt consists of Li_2BeF_4 (FLIBE) with dissolved fissile UF_4 and a blanket salt consisting of Li_2BeF_4 with dissolved fertile ThF_4. These conceptual designs assumed that $NaBF_4$ and NaF would be used as secondary loop coolants. FLINABE and FLINAK based systems are also possible.

In-depth understanding must be developed through application of internationally accepted predictive codes such as THERMOCALC to predict the formation of various phases, including

precipitates such as plutonium trifluoride (PuF_3). The prediction of phase diagrams for complex liquid fuels, with large numbers of fission products, requires that the phase diagrams for several binary pairs of salts first be developed. For example, equilibrium phase diagrams have been predicted for: BeF_2-LiF (FLIBE), BeF_2-ThF_4, BeF_2-UF_4, BeF_2-ZrF_4, LiF-PuF_3, LiF-ThF_4, LiF-ZrF_4, ThF_4-UF_4 and several other systems (Figures 4 and 5). The LIFE Project has recently predicted a phase diagram for the most basic FLIBE-based liquid fuel, assumed to consist primarily of lithium, beryllium, uranium and plutonium fluorides. This phase diagram is shown in Figure 5, and reveals regions of stable operation for such liquid fuels. Such systems would have to be operated at compositions and temperatures where no solid-phase precipitates form.

Most of our practical knowledge pertaining to liquid fuel comes from the development of the molten salt breeder reactor (MSBR) [Foster and Wright 1973]. The MSBR concept depends upon molten fluoride salts containing ^{7}LiF, BeF_2, ThF_4 and $^{233}UF_4$ being pumped through a graphite moderated core where heat released by fission will raise the salt temperature to 700°C (~1300°F), close to the operating temperature of the LIFE engine. The heated salt then passes through an intermediate heat exchanger where its enthalpy is reduced by transfer of heat to a secondary salt, sodium fluoroborate, which in turn may transfer energy to steam at 3500 psia and 540°C (~1000°F).

Initially, a two-salt MSBR design was favored, with a fuel salt containing only fissile UF_4 and no fertile ThF_4, and a blanket salt containing only ThF_4 and no UF_4. The fuel and blanket salts were passed in close proximity to one another inside a graphite moderator block, where uranium-233 was bred from thorium. Unfortunately, problems due to dimensional instability of the graphite moderator block after long-term neutron irradiation were encountered.

Enabled by the physical separation of fissile and bred uranium in the graphite moderator block of the two-salt MSBR, a relatively simple on-line fuel reprocessing plant was possible [Figure 13.31, Foster and Wright 1973]. In the conceptual on-line salt reprocessing plant, fuel salt is fluorinated to convert dissolved UF_4 to gaseous UF_6, thereby allowing uranium separation from the liquid stream. The remaining salt is separated from rare earth and other fission products by vacuum distillation. Approximately 5% of the fuel salt is discarded in this process, with the need for LiF and BeF_2 makeup. In contrast, the blanket salt is fluorinated primarily for the removal of bred uranium from the fertile thorium. Approximately 5% of the blanket salt is discarded in this process, with the need for LiF, BeF_2 and ThF_4 makeup. The total salt leaving the loop is approximately 8%. A LIFE engine with liquid fuel would require a separation process for the continuous removal of rare earth fission products to prevent the precipitation of PuF3. Similar salt removal would therefore be expected in a LIFE engine.

A reactor design with a single-salt two-region core and an easily replaceable graphite core assembly evolved, due to the problems anticipated with dimensional stability of the graphite in the two-salt design. This design eliminated the need for complicated flow paths inside the moderator block. The value of k_∞ was found to peak at approximately 4% salt content and drop

to below unity at approximately 18% salt content. It was found that a core with 13% salt content would be critical with k_∞ of 1.034, while a reflector region having less than 37% salt content would be sub-critical with k_∞ of 0.392. A breeding ratio of 1.07 to 1.08 was predicted for such a system with low fuel costs and small fuel inventories.

In the conceptual MSBR, the strong neutron capture of protactinium-233 (σ = 43b) in the core of the core leads to the production of protactinium-234, which in turn would decay to non-fissionable uranium-234. It is therefore be necessary to remove ^{233}Pa (27.4-day half life) from the core so that it has time to decay to fissionable ^{233}U. A liquid-to-liquid extraction process may be the best means for separating protactinium and uranium from the molten salt. An electrolysis process is then used to selectively recycle the uranium from salt in the decay tank to the MSBR fuel salt. Residual protactinium is then trapped in solidified salt for discarding.

The removal of noble gases such as xenon from the core of the MSBR is also essential for efficient breeding. Specifically, helium purging is used to remove xenon-135 ($\sigma = 2.72\times10^6$b), reducing the concentration of this gaseous fission poison by an order-of-magnitude ($\times$10).

Tritium recovery and permeation from molten fluoride salts in the molten salt breeder reactor (MSBR) has been previously studied, and is believed to be analogous to the application of such salt in the LIFE engine. In the case of the MSBR, it was assumed that tritium was dissolved in the molten salt, with the formation of tritium fluoride (TF) suppressed. It was believed that tritium in the MSBR would permeate through the hot steel tubes of heat exchangers and steam generators at a rate of approximately 1 gram per day in the absence of permeation barriers, assuming that 1% of the helium coolant flow rate was processed for tritium recovery at 90% efficiency per pass (90% separation efficiency). Tritiated water in the steam system was considered to be a personnel hazard at concentration levels well below one part per million (< 1 ppm), a level that would be reached quickly without costly isotopic processing. The use of permeation barriers (coatings) on MSBR and steam generator tubes was said to be able to reduce the leak rate of tritium into the steam system by two orders-of-magnitude (x100). For the option wi\th the lowest estimated leak rate of 55 curies per day, it may be possible to purge the steam system continuously to prevent the accumulation of tritiated water.

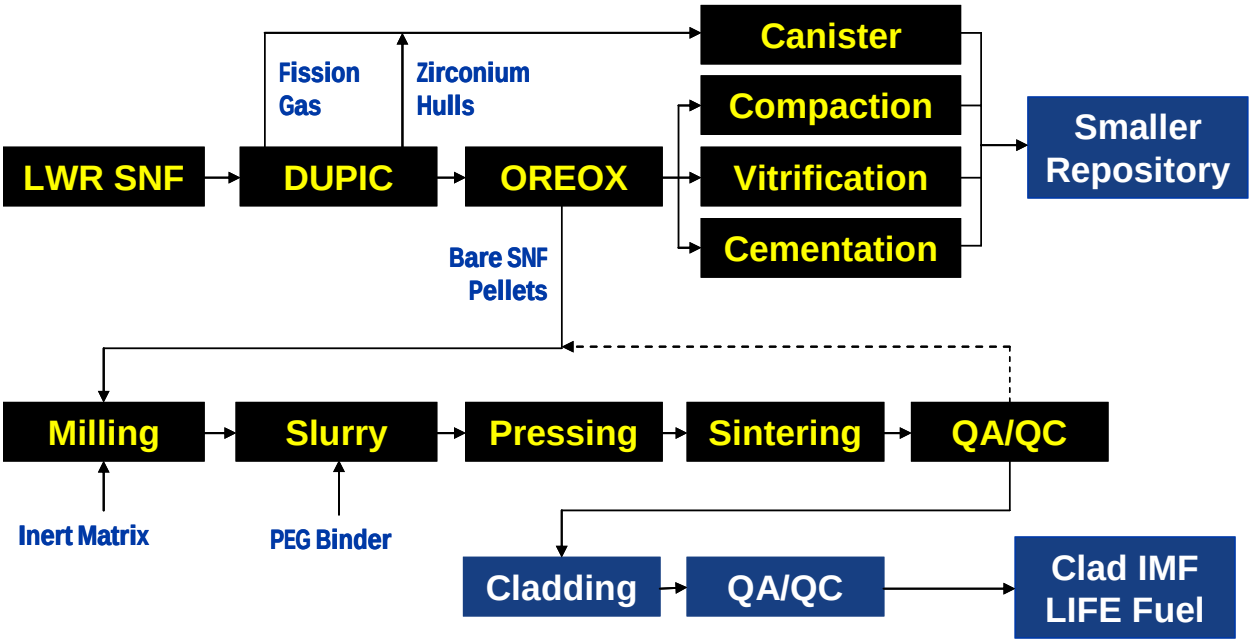


Figure 1 – Other processes, similar to those used for the DUPIC fuel cycle, may be needed for the conversion of LWR spent nuclear fuel into LIFE fuel.

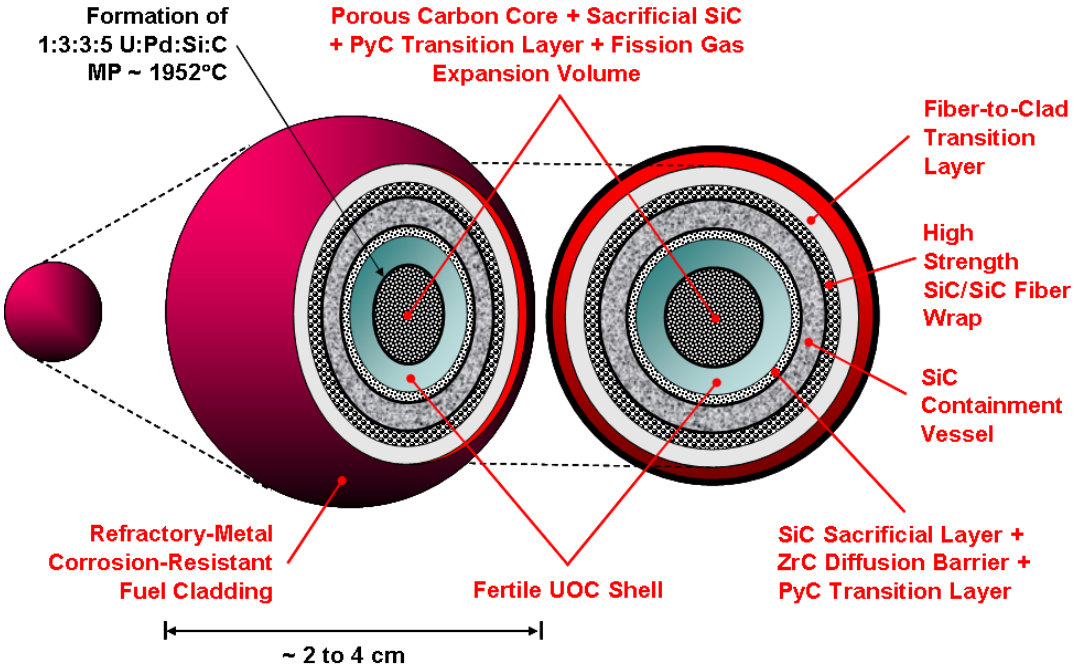
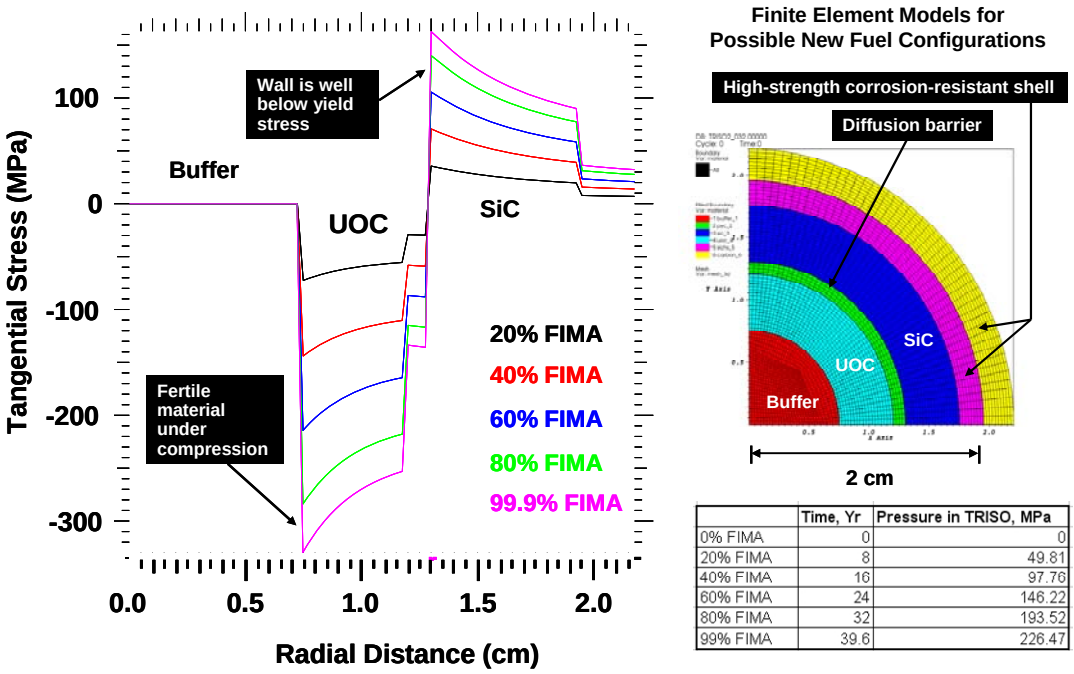


Figure 2 – A wide-variety of alternative materials and fuel designs are possible and are being evaluated. Higher mass fraction of fertile material can be achieved with new solid hollow core (SHC) fuel. The specific use of sacrificial SiC in fuels is attributed to R. J. Lauf et al. [1984].



	Time, Yr	Pressure in TRISO, MPa
0% FIMA	0	0
20% FIMA	8	49.81
40% FIMA	16	97.76
60% FIMA	24	146.22
80% FIMA	32	193.52
99% FIMA	39.6	226.47

Figure 3a – Predicted stress in the wall of solid hollow-core (SHC) fuel at fission gas pressures corresponding to 20, 40, 60, 80 and 99.9% FIMA.

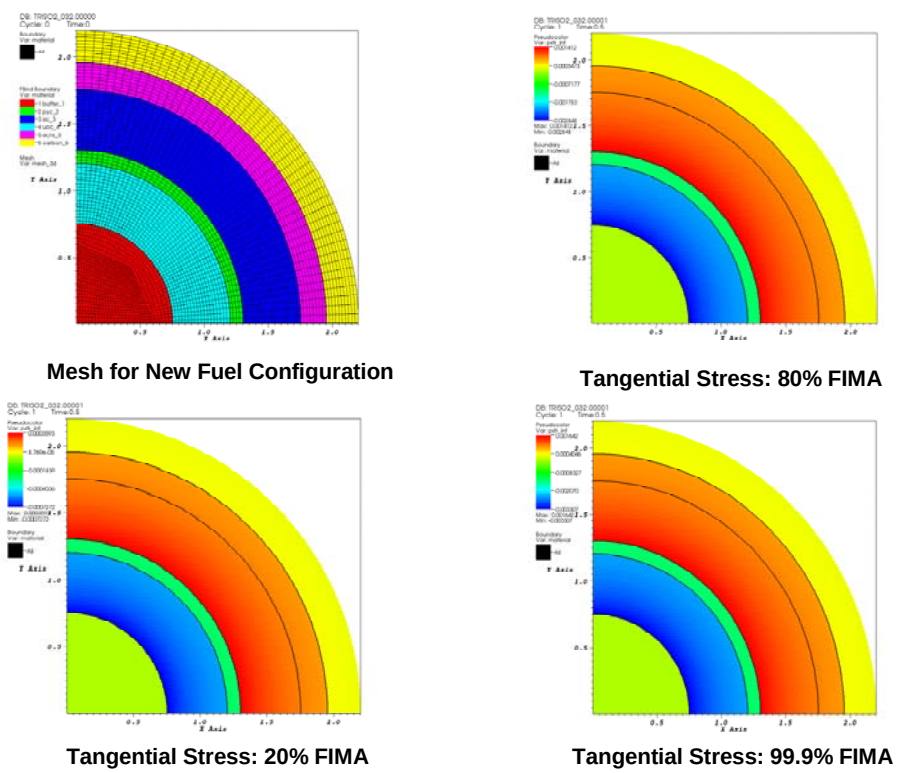


Figure 3b – Numerical analysis indicates a substantial strength margin in SHC pressure boundary. In all cases, the stress is well below the yield and ultimate tensile stresses of the wall material.

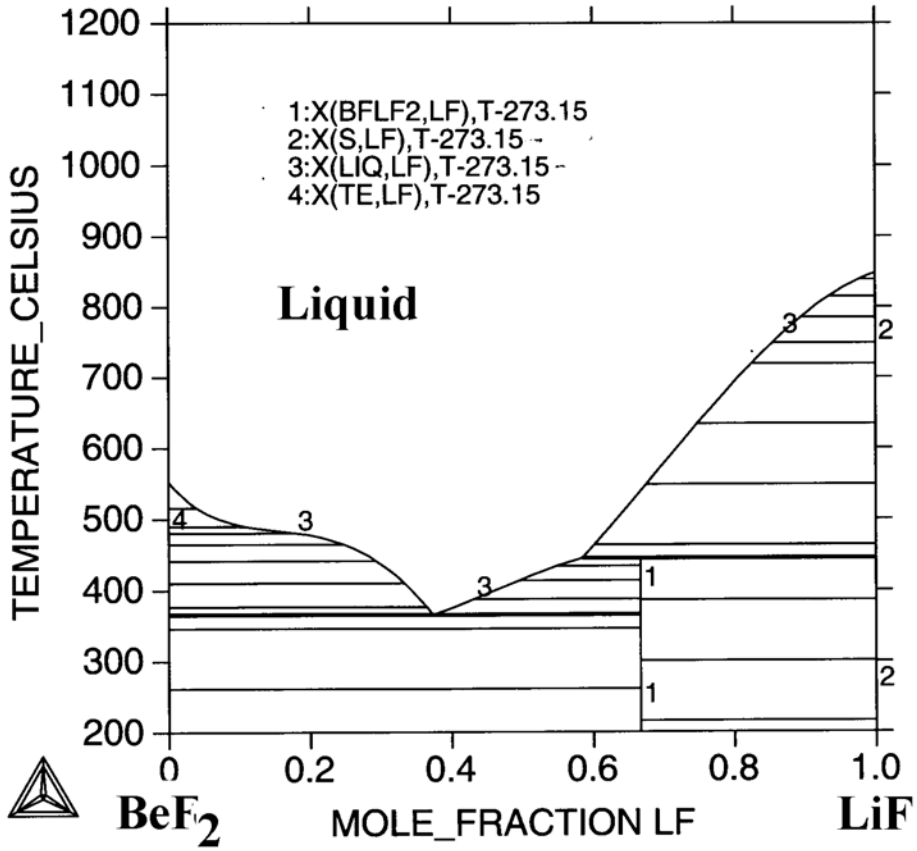


Figure 4 – A binary mixture of BeF_2 and LiF to form Li_2BeF_4 produces a eutectic composition with a low melting point of less than 400°C. This molten salt coolant is less prone to fire than liquid metal coolants, and can be used to breed tritium from the lithium. The corrosion of structural components of the LIFE engine in contact with this coolant must be understood.

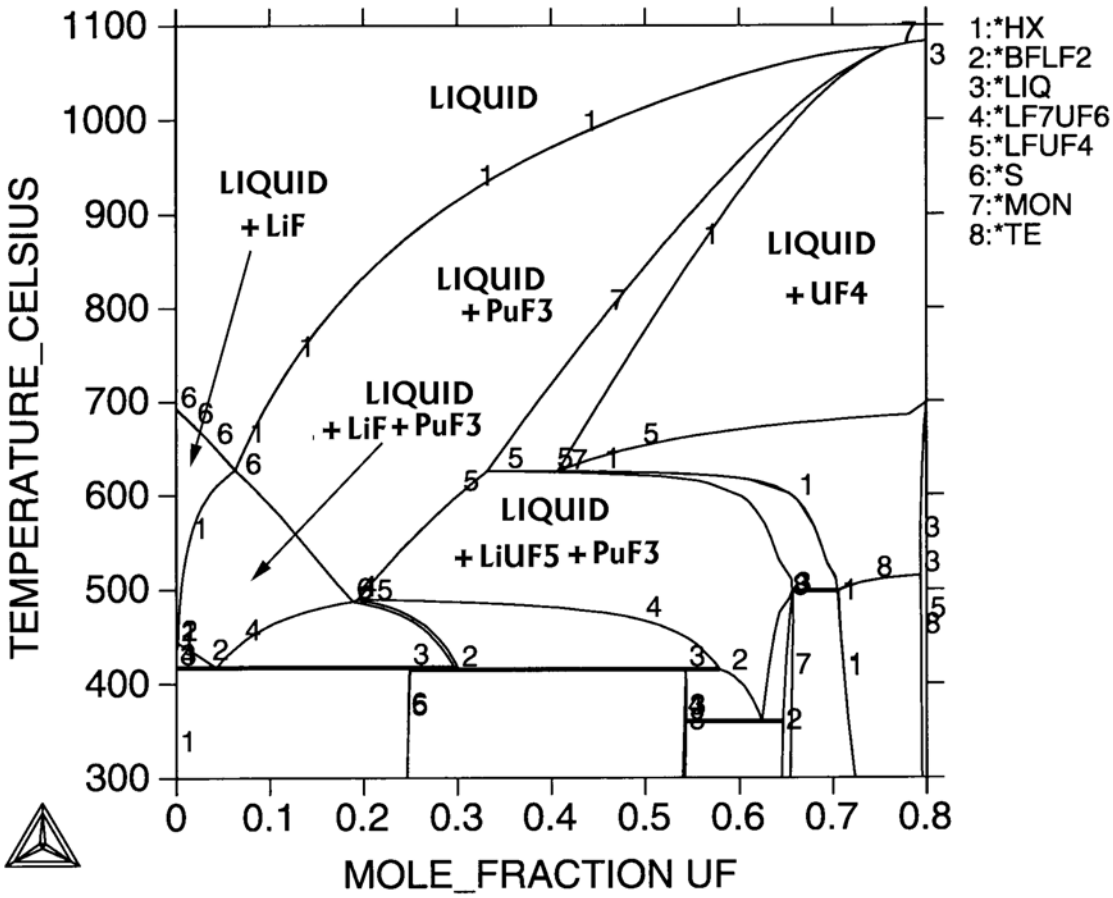


Figure 5 – Molten salt fuel is not damaged by radiation and enables direct digestion of fertile/fissile materials. Phase transformations and actinide solubility in LIFE coolants and liquid fuels: BeF_2-LiF, BeF_2-ThF_4, BeF_2-UF_4, BeF_2-ZrF_4, LiF-PuF_3, LiF-ThF_4, LiF-ZrF_4, ThF_4-UF_4 and higher order systems. Regions of stable operation revealed for liquid fuels in LIFE engine.

Table 1 – Blanket and Coolant Properties [Williams et al. 2006; Moir et al. 1985; Wright and Foster 1973; Rosenthal et al. 1971]

Salt Formula	Formula Weight	Melting Point	Latent Heat of Fusion	Vapor Pressure (900ºC)	Density	Heat Capacity (700°C)	Viscosity	Thermal Conductivity	Neutron Capture vs. Graphite	Moderating Ratio
	FW	MP	ΔH_f	P_{vap}	ρ	$\rho \times C_p$	μ	κ		
	(g/mol)	(ºC)	(cal/g)	(mm Hg)	(g/cm^3)	(cal/cm^3/ºC)	(cP)	(W/m-K)		
Li	6.9	179			554					
Be	9.0	1283			1820					
Na	23.0	97.81			970					
K	39.1	63.65			860					
LiF	25.9	842			2300					
BeF_2	47.0				1086					
LiF-BeF_2	33.0	460		1.2	1.94	1.12	5.6	1.00	8	60
LiF-NaF-BeF_2	38.9	315		1.7	2.00	0.98	5.0	0.97	20	22
KF-ZrF_4	103.9	390		2.8	2.80	0.70	5.1	0.45	67	3
Rb-ZrF_4	132.9	410		1.3	3.22	0.64	5.1	0.39	14	13
LiF-NaF-ZrF_4	84.2	436		5 2	2.79	0.84	6.9	0.53	20	13
LiF-NaF-KF	41.3	454		0.7	2.02	0.91	2.9	0.92	90	2
LiF-NaF-RbF	67.7	435		0.8	2.69	0.63	2.6	0.62	20	8
72LiF-16BeF_2-12ThF_4			63.00	0.1 torr	3.35		12	1.10		
70LiF-12BeF_2-18ThF_4					3.87					
71LiF-2BeF_2-27ThF_4			54.00		4.52		15-25	0.70		

Note: A wide variety of high temperature fluids exist that can be used to cool fission, fusion and fusion-fission hybrid systems, and include liquid metals and alloys, and fluoride salts. These include lithium, beryllium, sodium, potassium, alkali metal alloys, lithium fluoride, beryllium fluoride, lithium-beryllium fluoride (FLIBE = Li_2BeF_4), and mixed alkali-metal fluorides (FLiNaK = Li-Na-K-F). The presence of the lithium in the salt enables breeding of tritium, while the beryllium helps moderate neutrons. At the present time, the LIFE engine design specifies FLIBE for the primary coolant system, with the extraction of breed tritium from this fluid. The secondary cooling loop will use beryllium-free FLiNaK, which is both less expensive and less hazardous than the FLiNaK. The properties of many of these high-temperature heat-transfer fluids are summarized in Table 8 [Williams et al. 2006; Moir et al. 1985; Wright and Foster 1973; Rosenthal et al. 1971].

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