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Lattice optimization for graphite moderated molten salt reactors using low-enriched uranium fuel

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ABSTRACT

The family of Generation-IV reactor concepts comprises of multiple promising designs, one of which is the molten salt reactor. These reactors have traditionally been chosen for the possible use of the thorium-uranium fuel cycle and used ^{7}LiF-BeF$_2$ carrier salt. This particular salt choice however presents several challenges due to the cost of highly depleted ^{7}Li isotope for the carrier salt, tritium production, and beryllium toxicity. Additionally, lack of developed and accepted safeguards methodology for thorium fuel cycle presents a barrier. While none of these issues are insurmountable, alternatives are worth investigating. The purpose of this paper is to analyze the more cost effective and regulatory amenable fuel salt choices by using low-enriched uranium fuel in the form of UF$_4$. Several eutectic mixtures are examined that avoid the use of ^{7}Li and Be while maintaining a melting point low enough to be compatible with standard structural materials. The optimal conditions for hexagonal lattice arrangements using nuclear graphite moderation are discussed for multiple fuel salt choices. The aim of this study is to present options of using simpler molten salt reactor alternatives focused on thermal single-fluid low-enriched uranium converter concepts.

1. Introduction

There are currently six contemporary reactor concepts that have been chosen by the Generation IV International Forum (GIF), one of these six modern designs is the molten salt reactor (MSR). The MSR concept is commonly chosen for its versatility, inherent safety, and potential as a breeder or iso-breeder that utilizes the ^{232}Th - ^{233}U fuel cycle (Weinberg et al., 1970; Furukawa et al., 2008; Nuttin et al., 2005). MSRs can also act as actinide burners (Fiorina et al., 2013; Ignatiev et al., 1606; Vergnes and Lecarpentier, 2013), often utilizing ^{7}LiF-BeF$_2$ (FLiBe) as their carrier salt of choice. These options encounter specific challenges with material availability and regulations in regards to the movement of fuel, which pose practical and economic barriers for the near-term deployment of MSRs. Some of these obstacles include: increased production of tritium, beryllium toxicity and cost, the availability and cost of lithium that is highly enriched with the ^{7}Li isotope, the small amount of available ^{233}U, the cost of starting the fuel cycle through the use of actinides, and possible proliferation concerns over the high percent ^{233}U fuel. These challenges can be addressed with research and other efforts, however this paper explores the possibility of reducing the number of near term barriers by using existing low enriched uranium technology operating in conjunction with carrier salts that have reduced or no tritium production (LeBlanc, 2010), other than from ternary fission itself which is not considered in this text.

In this paper the beginning of cycle (BoC) critical uranium enrichment level is given along with the estimated conversion ratio (CR) for an infinite hexagonal lattice configuration. Multiple fluoride based carrier salts are examined with the expected use of a single fluid denatured molten salt converter reactor that is operating in the thermal spectrum. The lattice configuration is described by the pitch of the fuel channel and the volumetric salt fraction.

MSRs can possibly be made more economical and more deployable through the simplification and partial expulsion of the fuel reprocessing system. The optimum fuel cycle choice falls outside of the scope of this paper, which focuses mostly on BoC criticality condition, however a fuel cycle study will be presented in a follow up paper. Preliminary results from this work have been reported at a conference (Chvala, 2014), however this study includes an additional salt option and provides denser coverage of the lattice space.

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Nomenclature

BoC	beginning of the cycle
CR	conversion ratio
FoM	figure of merit
LEU	low-enriched uranium
HEU	highly-enriched uranium
MSR	molten salt reactor
MSBR	molten salt breeder reactor
DMSR	denatured molten salt reactor
UT	the University of Tennessee

2. Lattice description

All lattice calculations and resulting data were performed using MCNP5 version 1.60 (X5 Monte Carlo Team, 2003; Brown et al., 2002) and were validated using Serpent2.1.23 beta (Leppänen et al., 2015) with ENDF/B-VII.0 libraries evaluated at 900 K (Chadwick et al., 2006). The library names used in both calculation codes are presented to allow for the duplication of results. The moderator used in this study is nuclear graphite. The graphite is assumed to have boron equivalent impurities of 2 ppm with a density of 1.8 g/cm^3 (Nuttin et al., 2005; Swank et al., 2010; Ball and Miller, 2007). The temperature of the graphite is 700 °C with proper thermal scattering treatment. The cross-section library used is ENDF/B-VII.0 and is evaluated at 900 K, while the library for thermal scattering is ENDF/B-VII.0sab evaluated at 1000 K.

2.1. Parameterization of lattice geometry

Each lattice is parameterized by the spacing of the fuel channels (pitch), and the fractional volume that contains the fuel salt (salt fraction). The hexagonal lattice, shown in Fig. 1, uses reflective boundary conditions.

This geometry relates the channel radius r with pitch p and salt fraction f using Eq. (1):

$$r^2 = p^2 \frac{\sqrt{3}}{2\pi} f \tag{1}$$

2.2. Fuel salt choices

The salts that are used in this study differ from other traditional MSR salt choices in that they do not contain any thorium, as shown in Table 1. By choosing salts that do not contain thorium, criticality may be reached with lower enriched uranium, since fertile uranium 238 replaces fertile thorium 232. The solubility of trifluoride actinides, in particular PuF_3, is not a limiting factor due to the thermal neutron spectrum resulting in low fissile loading, see Section 4.4.

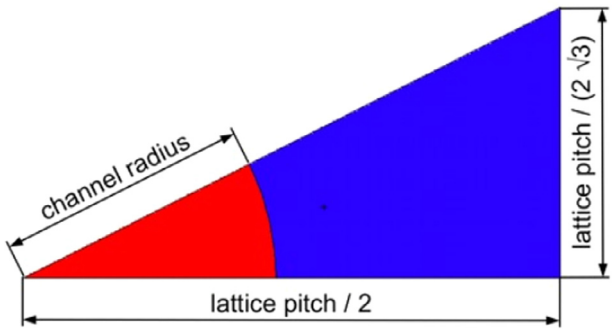

Fig. 1. Unit cell of a hexagonal lattice.

Table 1
Properties of the selected salts.

	Salt Composition	Melting Point [°C]	Density [g/cm^3]
1	72%^{7}LiF-16%BeF_2-12%UF_4	480	3.353
2	73%^{7}LiF-27%UF_4	490	4.340
3	78%NaF-22%UF_4	618	4.056
4	49%NaF-38%ZrF_4-13%UF_4	540	3.757
5	58%NaF-30%BeF_2-12%UF_4	525	3.208
6	74%NaF-12%BeF_2-14%UF_4	500	3.437
7	46%NaF-33%RbF-21%UF_4	470	4.026
8	50.5%NaF-21.5%KF-28%UF_4	490	4.326

Each salt choice is chosen with a large emphasis on their economic impact. The first salt in Table 1 is given for comparison, and gives an estimate for FLiBe salt where the thorium fraction is replaced with uranium. The second salt, 73%^{7}LiF-27%UF_4, holds the largest uranium capacity. This salt also simplifies laboratory development work due to the lack of beryllium, eliminating the need for safety protocol requirements related to the toxicity of beryllium. The third salt, 78%NaF-22%UF_4, has a higher melting point but no lithium content, which causes tritium production and is cost inefficient (Thoma et al., 1959). The fourth salt, 49%NaF-38%ZrF_4-13%UF_4, holds a decreased melting point while maintaining benefits from the exclusion of lithium and berylium (Thoma et al., 1959). The fifth salt, 58%NaF-30%BeF_2-12%UF_4, and the sixth salt, 74%NaF-12%BeF_2-14%UF_4, aim to reduce the parasitic absorption of zirconium by replacing zirconium with beryllium.

The seventh salt in Table 1, 46%NaF-33%RbF-21%UF_4, holds multiple benefits with a decreased melting point, large capacity of uranium, and does not cause any production of tritium (Thoma et al., 1959). The last salt, 50.5%NaF-21.5%KF-28%UF_4, does not contain any beryllium or lithium. This allows for a safer salt with no beryllium toxicity or chance of tritium production due to the presence of lithium. This salt is also less expensive as its constituents, sodium and potassium fluorides, are cheap.

The salt densities were calculated using the molar additions method (Cantor et al., 1968; Williams and Toth, 2005; Briggs, 1966), and phase diagrams were used to estimate the melting points of each salt (Thoma and Grimes, 1957; Thoma et al., 1959). All salt components are presented in their natural isotopic abundance, excluding ^{7}Li, which is depleted to 99.995%. The level of uranium is varied in order to reach a k_∞ of unity, allowing for the representation of a BoC critical system. The expected temperature of the fuel salt is 650 °C, which is used for all observed salt choices.

3. Criticality searches

Each lattice choice is defined by two variable parameters, the space in lattice pitch p and the salt fraction f. The enrichment level of uranium required for the infinite lattice to be critical is calculated for each parameter combination. There are approximately 1250 points sampled in each lattice pitch range from 1 to 60 cm