THE EFFECTS OF VARIATIONS IN LITHIUM (^{7}Li) AND URANIUM (^{235}U) ISOTOPE LEVELS ON A SMALL FLUORIDE SALT-COOLED, HIGH-TEMPERATURE REACTOR WITH UO_2 FUEL PINS

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ABSTRACT

Lithium beryllium fluoride, FLiBe (66.7 mole % LiF and 33.3 mole % BeF_2) has become the baseline coolant for fluoride salt-cooled high-temperature reactors (FHRs). Natural lithium contains 92.5% ^{7}Li and 7.5% ^{6}Li. The presence of ^{6}Li can present challenges as it is an unfavourably strong neutron absorber. Hence, the objective of this study is to perform sensitivity analysis on ^{7}Li enrichment in order to better understand the effects on neutronic performance, multiplication factor and coolant temperature coefficient of reactivity (TCR), for a pin-type fuel assembly in a small FHR. The calculations carried out for this study use the WIMS infinite lattice code and a UO_2 pin-type fuel assembly design. The first case is performed at 19.75 wt.% ^{235}U enrichment while the ^{7}Li enrichment is varied over seven different values, namely 100%, 99.999%, 99.995%, 99.99%, 99.9%, 99.1% and 99.0%. Second test is performed at 7 wt.% ^{235}U and the ^{7}Li enrichment is only varied over six values, namely 100%, 99.999%, 99.995%, 99.99%, 99.9% and 99.1%. Additional variation of the ^{7}Li enrichment is unnecessary because the system becomes subcritical when the ^{7}Li enrichment is at 99.1% or lower. Results show that the neutronic performance is not only affected by the ^{7}Li enrichment, but also sensitive to the UO_2 fuel enrichment, especially at lower fuel enrichments. For 19.75 wt.% UO_2, the reactivity is incremented by 11 pcm while the coolant TCR is improved by 0.013 pcm for every 0.001% increase in ^{7}Li enrichment. On the other hand, for 7 wt.% UO_2, the reactivity is incremented by 35 pcm and the coolant TCR is improved by 0.014 pcm for every 0.001% increase in ^{7}Li enrichment.

ABSTRAK

Litium berilium fluorida, FLiBe (66.7 mole% LiF dan 33.3 mole% BeF2) telah menjadi penyejuk asas bagi reaktor suhu tinggi yang disejukkan dengan garam fluorida (FHRs). Litium semulajadi mengandungi 92.5% 7Li dan 7.5% 6Li. Kehadiran 6Li dapat menimbulkan cabaran kerana penyerap neutron yang tidak baik. Oleh yang demikian, objektif kajian ini adalah untuk melakukan analisis kepekaan terhadap pengayaan 7Li untuk lebih memahami kesan ke atas prestasi neutron, faktor pendaraban dan koefisien kefaktor reaktiviti suhu (TCR) untuk pemasangan bahan pin-pin dalam FHR yang kecil. Pengiraan yang dilakukan untuk kajian ini

menggunakan kod kekisi WIMS tak terhingga dan reka bentuk perhimpunan bahan api pin-UO2. Kes pertama dilakukan pada pengayaan 19.75 wt.% 235U manakala pengayaan 7Li bervariasi dalam tujuh nilai yang berbeza iaitu 100%, 99.999%, 99.995%, 99.99%, 99.9%, 99.1% dan 99.0%. Ujian kedua dilakukan pada 7 wt.% 235U dan pengayaan 7Li hanya bervariasi daripada enam nilai iaitu 100%, 99.999%, 99.995%, 99.99%, 99.9% dan 99.1%. Perubahan tambahan pengayaan 7Li tidak perlu kerana sistem menjadi subkritis apabila pengayaan 7Li berada pada 99.1% atau lebih rendah. Keputusan menunjukkan bahawa prestasi neutron tidak hanya dipengaruhi oleh pengayaan 7Li, tetapi juga sensitif terhadap pengayaan bahan bakar UO2, terutama pada pengayaan bahan bakar yang lebih rendah. Untuk 19.75 wt.% UO2, kereaktifan meningkat sebanyak 11 pcm manakala TCR penyejuk diperbaiki dengan 0.013 pcm untuk setiap peningkatan 0.001% dalam pengayaan 7Li. Sebaliknya, untuk 7% berat UO2, kereaktifan meningkat sebanyak 35 pcm dan penyejuk TCR meningkat sebanyak 0.014 pcm untuk setiap peningkatan 0.001% dalam pengayaan 7Li.

Keywords: FLiBe, $^{7}Li_2BeF_4$, fluoride salt, lithium enrichment, high-temperature reactor, FHR.

INTRODUCTION

Fluoride salt-cooled high-temperature reactors (FHRs), also known as solid-fuelled molten salt reactors, are one of the new reactor concepts proposed in the Generation IV International Forum [1]. The main distinguishing features of the FHR are its high operating temperature, lithium beryllium fluoride salt (Li_2Be_4F) coolant, known as FLiBe, and graphite moderator. One of the active efforts to develop the FHR is currently spearheaded by Charles Forsberg and Per Peterson's group, which involves researchers from Massachusetts Institute of Technology, the University of California at Berkeley (UCB), University of Wisconsin (UW) and others [2]. Most FHR studies have focused on the use of tristructural-isotropic (TRISO) particulate fuels that are compacted with a graphite matrix to form various fuel shapes. Concerns about the high potential fabrication cost of TRISO fuels have motivated our interest in the possible alternative use of more conventional pin-type UO_2 fuel in an FHR [3].

Some of the special characteristics of FLiBe are its transparency and its high heat capacity. In addition, the salt does not experience any rapid chemical reactions, and it is chemically compatible with structural metals and graphite. Compared to other coolants, FLiBe has the highest heat capacity, enabling it to transfer a large amount of energy with less coolant volume.

In addition, it can be used for high temperature operation (above 700°C) and at low system pressure (less than 1 MPa) because of its relatively high boiling point and low melting point. Neutronically, FLiBe is an effective neutron moderator because of the low atomic mass of lithium and beryllium. At the same time, FLiBe, in general, has small neutron cross-sections compared to other materials in a core such as fuel and graphite, hence making it quite transparent to neutrons [4]. FLiBe is the most neutronically favourable salt, which can be seen from its moderating ratio, *MR*. The moderating ratio and total neutron capture rate relative to graphite for basic materials in a core such as coolants, graphite and fuels are presented in Table 1.

TABLE 1. Neutron efficiency for various coolants and core materials [4, 5].

Coolant/Material	Total neutron capture relative to graphite	Moderating ratio, *MR*
$^{7}Li_2BeF_4$ (FLiBe)	8	60
Sodium	47	2
Helium	-	45
Light Water	75	246
Heavy Water	0.2	11449
Graphite	1	863
UO_2	3583	0.1

Naturally, lithium contains 92.5% ^{7}Li and 7.5% ^{6}Li. ^{6}Li is a strong neutron absorber and its presence can be a concern because it will reduce moderating ratio, *MR*. Figure 1 shows a comparison of the ^{6}Li and ^{7}Li microscopic elastic scattering and total cross-sections. From this plot, it can be seen that the ^{6}Li total cross-section has totally a different characteristic and higher magnitude at lower energies than the ^{6}Li elastic scattering cross-section, implying that the ^{6}Li(n,t) capture reaction is significantly more likely at lower energies. On the other hand, the ^{7}Li elastic scattering cross-section has a somewhat similar pattern to its total cross-section, which signifies that ^{7}Li is a weak neutron absorber. The ^{6}Li isotope has a large thermal neutron absorption cross section (about 1,000 barn at 0.1 eV), which is undesirable for neutron economy [6].

Another issue related to ^{6}Li is tritium production. Other than ^{9}Be and ^{7}Li reactions, ^{6}Li is one of the primary contributors to tritium production in the beginning of the coolant operating. Tritium produced in the coolant is not only a radiological concern, but it also poses a serious corrosion issue due to its initial form of tritium fluoride (TF), which is a strong oxidant. In addition, depending on the chemical redox potential in the salt and/or the occurrence of chemical reactions, tritium can also exist in the form of tritium gas, T_2. Unlike T_2, TF does not diffuse through metals. Tritium gas T_2, on the other hand, can diffuse through most metals at normal FHR operating temperatures (~700°C) and release to atmosphere, presenting an off-site concern.

A sensitivity analysis on ^{7}Li enrichment seems necessary to better understand the effect on neutronic performance (multiplication factor, k and coolant temperature coefficient of reactivity, TCR) for a pin-type fuel assembly in a small FHR. This work, however, will not investigate the ^{7}Li enrichment effects on the tritium production.

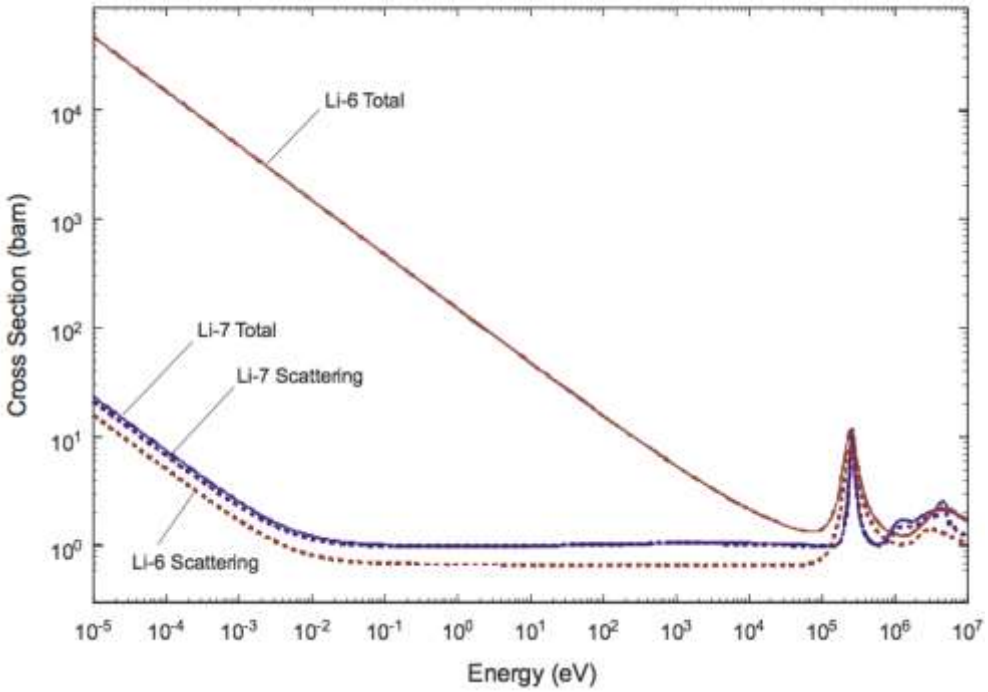

Figure 1. Microscopic cross-sections of ^{6}Li and ^{7}Li. Reproduced from Ref. 2.

METHODS

Design Model and Test Cases

The studied fuel assembly design is inspired by the Oak Ridge National Laboratory (ORNL) 125 MWth Small modular Advanced High Temperature Reactor (SmAHTR) concept [7]. The fuel assembly, as shown in Figure 2, consists of 72 fuel and 19 graphite pins with a diameter of 2.2 cm. The fuel pin pitch is 3.08 cm. The fuel cladding is made of graphite with a thickness of 0.3 cm. Unlike the SmAHTR concept, the fuel used in this work is a conventional UO_2 ceramic fuel pin and not a TRISO fuel compact.

Two fuel enrichments are analysed: (Case 1) 19.75 wt.% ^{235}U and (Case 2) 7 wt.% ^{235}U. The former is a typical value set as the maximum limit for low-enriched uranium (LEU). The latter is the minimum enrichment required for the fuel assembly design to achieve supercriticality, as, in our preliminary study, it was observed that the studied design with UO_2 fuel pins has a weak thermal spectrum. Therefore, the design requires a higher enrichment (7 wt.%) than the typical enrichment range (3 wt.% to 5 wt.%) for light water reactors to be supercritical.

For Case 1, the ^{7}Li enrichment is varied over 100%, 99.999%, 99.995%, 99.99%, 99.9%, 99.1% and 99.0%. Each variation is run at full power for 15,300 days since the fuel enrichment is high. For Case 2, the ^{7}Li enrichment is only varied over six values, namely 100%, 99.999%, 99.995%, 99.99%, 99.9% and 99.1%, due to its lower ^{235}U content (7 wt.%). A decrease in ^{7}Li content below 99.1% causes the system to be subcritical at this fuel enrichment.

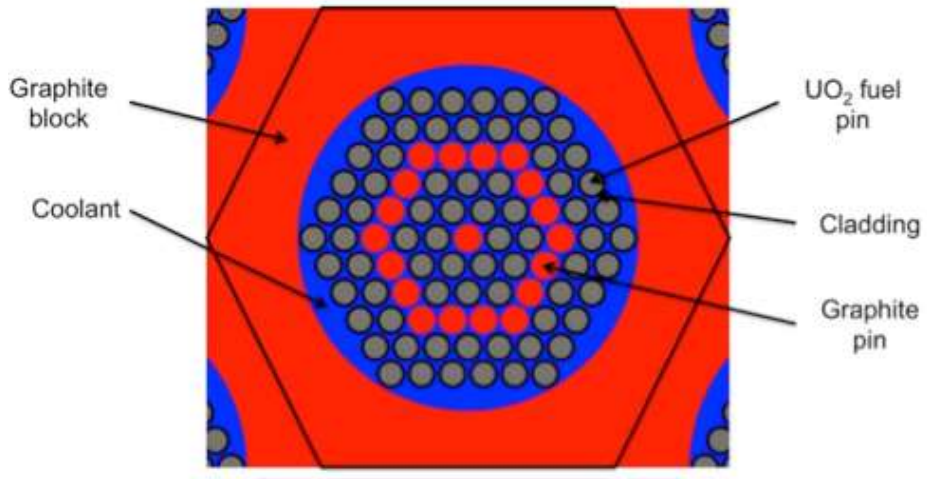

Figure 2. A fuel assembly in an infinite lattice with solid cylindrical UO_2 fuel pins.

Reactor Physics Code: WIMS

The WIMS reactor physics code is used to perform 2-D infinite fuel assembly lattice calculations. WIMS is a deterministic code developed by ANSWERS, Amec Foster Wheeler that can solve problems such as fuel depletion analysis and multiplication factor, k, calculations [8]. A calculation sequence in WIMS involves a set of methods, also called modules. Operations in WIMS start with data preparation and the reading of data libraries. The ENDF/B-VII data library is selected as the main cross-sections library as it is commonly used for FHR studies [4, 9]. Then, using the HEAD module, all the dimensions and material properties are initially set. Prior to using a flux-solving module, a service module such as COND is used to perform an energy condensation of cross-section data and flux distributions. For example, it can condense 172 energy groups to a smaller number of groups, hence reducing calculation time.

Later, the CACTUS module is used to solve the neutron transport equation. CACTUS solves the multigroup neutron transport equations using the method of characteristics, which is a numerical method in which the differential form of the Boltzmann equation is integrated along explicit lines (or tracks) through the geometry. The neutron flux is then acquired by the addition of the contributions made by a discretisation of these lines. At the end of the CACTUS module, the assembly infinite multiplication factor, k_{inf}, of the model is calculated. The effective multiplication factor is then estimated by applying 4% leakage probability. This value seems reasonable as it is within the typical neutron leakage probabilities, 3% and 5%, used in PWR [10] and FHR [11] assessments, respectively. The assembly k_{inf} must be 1.04 at the critical (end-of-cycle) burnup point.

To find the coolant TCR, the temperature of the coolant is perturbed by ±30 K. The coolant temperature coefficient is defined as the change in reactivity per degree change in coolant temperature, calculated as:

$$\alpha_{\text{coolant}} = \frac{k_{\text{inf}}(T_{c2}) - k_{\text{inf}}(T_{c1})}{k_{\text{inf}}(T_{c2}) \square k_{\text{inf}}(T_{c1}) \square (T_{c2} - T_{c1})} \square 10^5 \qquad (1)$$

where T_{c1} and T_{c2} are the two coolant temperatures, while $k_{\text{inf}}(T_{c1})$ and $k_{\text{inf}}(T_{c2})$ are the corresponding k_{inf} values. The density of FLiBe is sensitive to temperature. Hence, the coolant density is also adjusted when its temperature is changed, with the assumption that the internal pressure of the core is uniform. All the reactivity coefficients were measured in pcm per K.

RESULTS

Figures 3 and 4 show plots of the variation of the effective multiplication factor, k_{eff}, against burnup for the 19.75 wt.% and 7 wt.% ^{235}U cases, while the differences in the reactivity between other ^{7}Li enrichments and the baseline enrichment of FLiBe (99.995% ^{7}Li - 0.005% ^{6}Li) for both the 19.75 wt.% and 7 wt.% ^{235}U cases are presented in Figures 5 and 6. Additionally, using the k_{eff} plots, the percentage differences in cycle length and differences in discharge burnup for each case are determined and presented in Figures 7 and 8.

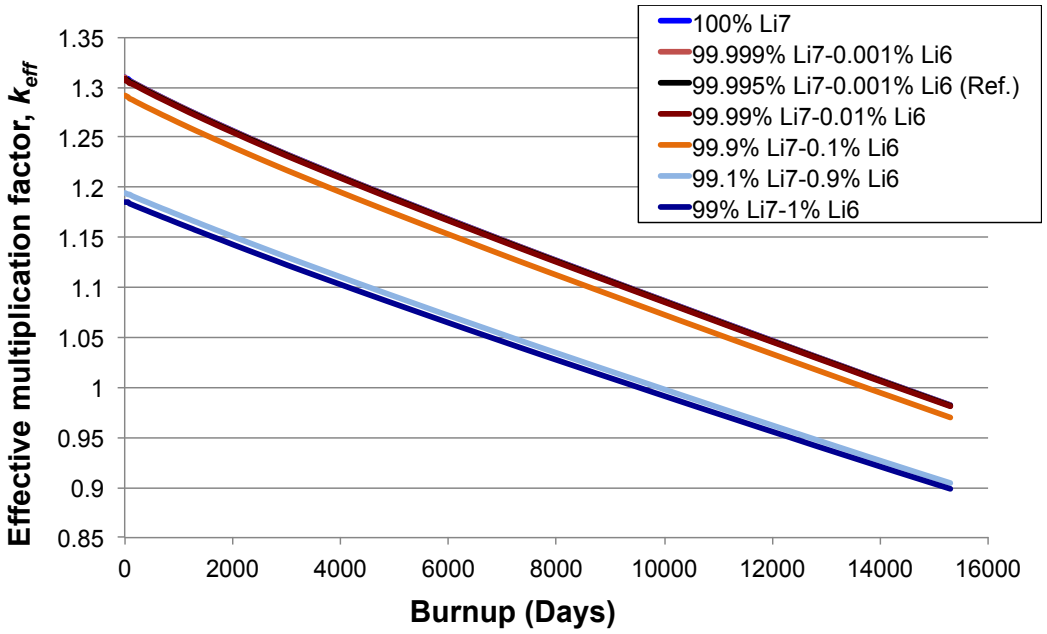

Figure 3. Effective multiplication factor, k_{eff}, as a function of burnup (days) for Case 1: 19.75 wt.% UO_2 at various Li enrichments in FLiBe.

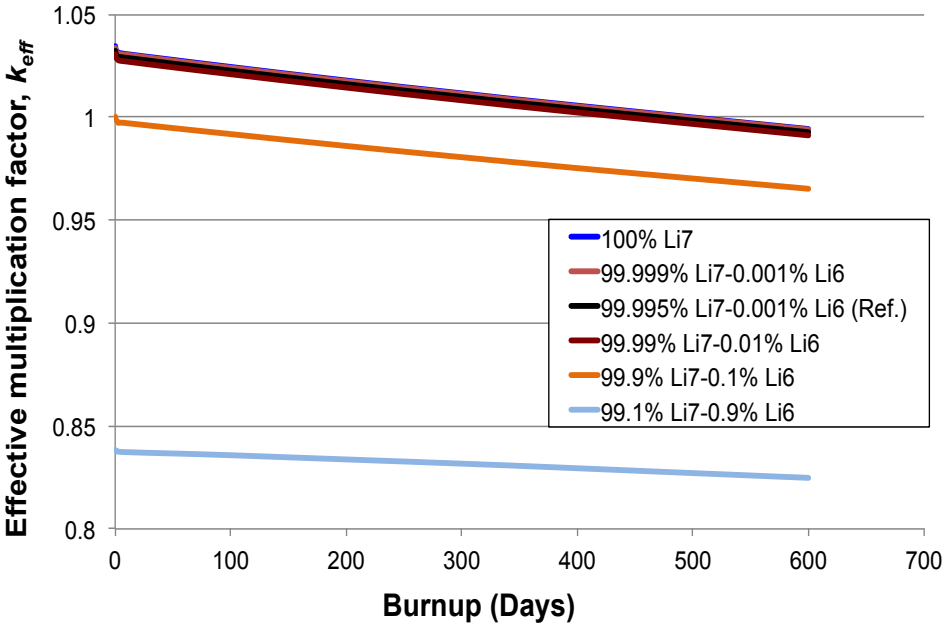

Figure 4. Effective multiplication factor, k_{eff}, as a function of burnup (days) for Case 2: 7 wt.% UO_2 at various Li enrichments in FLiBe.

Both Figures 3 and 4 show that the beginning-of-life (BOL) k_{eff} decreases when the ^{7}Li enrichment is reduced. At 99.9% ^{7}Li, the fuel assembly design with 7 wt.% ^{235}U is only supercritical at BOL before quickly becoming subcritical at the next depletion step. When the ^{7}Li enrichment is further reduced to 99.1%, the system can no longer achieve supercriticality. On the other hand, for the test case with 19.75 wt.% ^{235}U, at 99.1% ^{7}Li the system is still in a supercritical state for a long period of time. This is mainly due to the fact that, at higher fuel enrichment, more neutrons are being produced from the larger amount of fissile material which then can counter the neutron loss from ^{6}Li capture reactions.

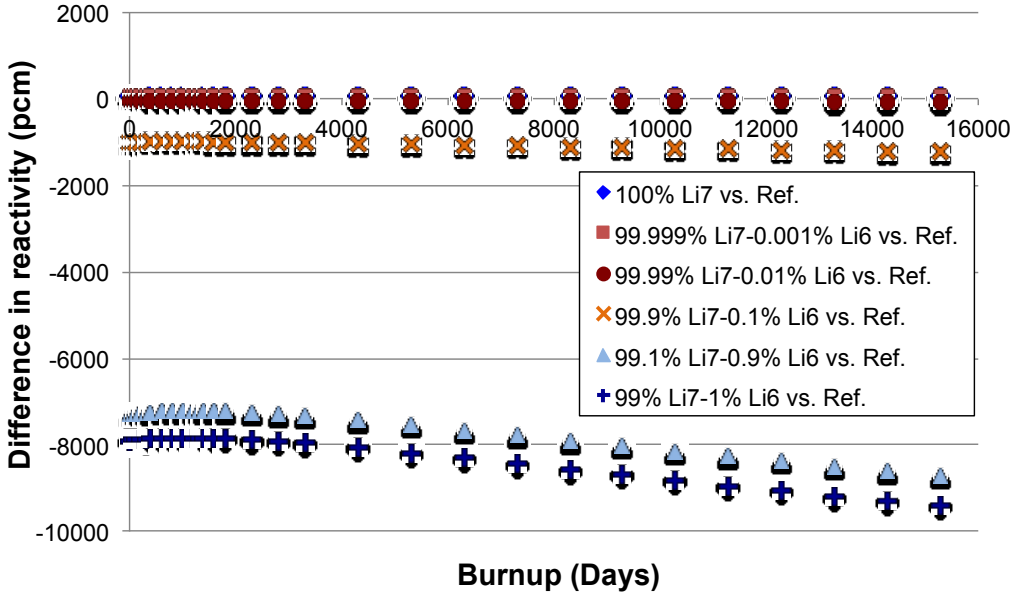

Figure 5. Difference in reactivity for the 19.75 wt.% UO_2 case between various ^{7}Li enrichments and the baseline enrichment in FLiBe.

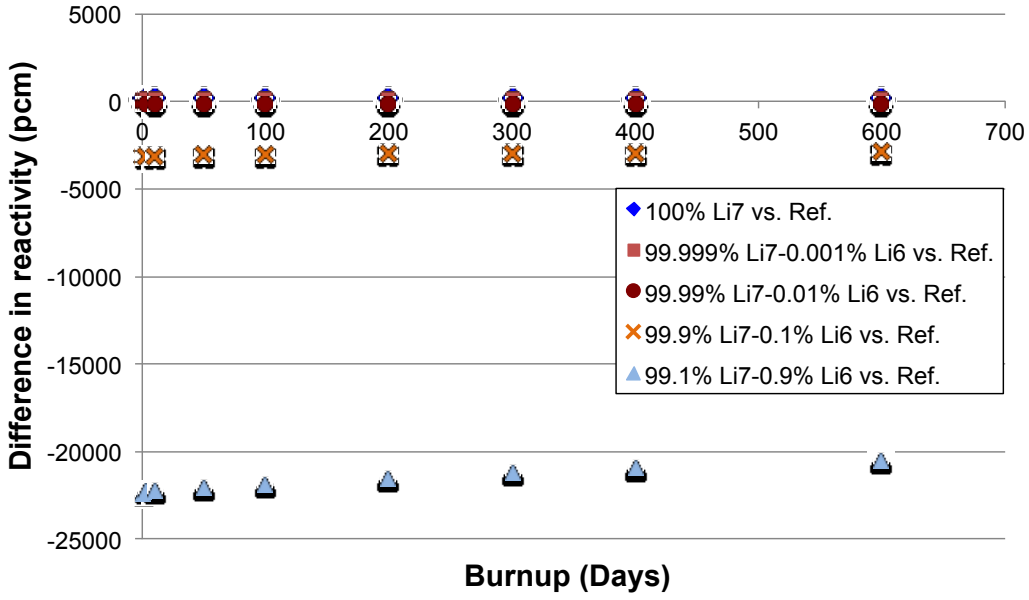

Figure 6. Difference in reactivity for the 7 wt.% UO_2 case between various ^{7}Li enrichments and the baseline enrichment in FLiBe

In addition, the results show that the change in k_{eff} for every percentage change of ^{7}Li enrichment is different between the 19.75 wt.% (Figure 5) and 7 wt.% (Figure 6) ^{235}U cases. At higher UO_2 enrichment (19.75 wt.%), every 0.005% increase in ^{7}Li enrichment will add about 55 pcm reactivity (every 0.001% increase will add ~11 pcm). In comparison, at lower fuel enrichment (7 wt.%), every 0.005% (0.001%) increase in ^{7}Li enrichment will add approximately 173 (35) pcm. The reactivity drops significantly by 1051 pcm (for 19.75 wt.% ^{235}U) or 2941 pcm (for 7 wt.% ^{235}U) when the ^{7}Li enrichment is reduced from 99.995% to 99.9% (0.095% change). In general, it can be concluded that the neutronic performance is highly sensitive to changes in the enrichment of ^{7}Li in FLiBe at much lower UO_2 fuel enrichment. No previous studies have observed and reported this coupled effect of fuel and ^{7}Li enrichments on the neutronic performance of FHR systems.

Other factors, such as the fuel assembly composition (graphite, coolant and fuel) and configuration (pitch and fuel pin dimensions), may also influence the size of the effects arising from changes in ^{7}Li enrichment in a core. For example, for the Liquid Salt Very High-Temperature Reactor (LS-VHTR) design, ORNL reported that an increase of ^{7}Li enrichment from 99.99% to 99.995% results in an increase of the cycle length by 12 days [4]. In our analysis, however, with such an enrichment change, the cycle length is increased either by 35 (19.75 wt.% ^{235}U) or 29 (7 wt.% ^{235}U) days, as shown in Figure 7. In regard to discharge burnup, an increase in ^{7}Li enrichment of 0.005% results in either 0.24% (19.75 wt.% ^{235}U) or 6.08% (7 wt.% ^{235}U) increase, as shown in Figure 8.

In another different example, Lewis [12] reported that the AHTR fuel plank design with 19.75 wt.% ^{235}U TRISO fuels cannot achieve supercriticality at 99.5% ^{7}Li. In contrast, the fuel pin-type assembly from our analysis can still achieve supercriticality even at 99% ^{7}Li when the fuel enrichment is 19.75 wt.% ^{235}U.

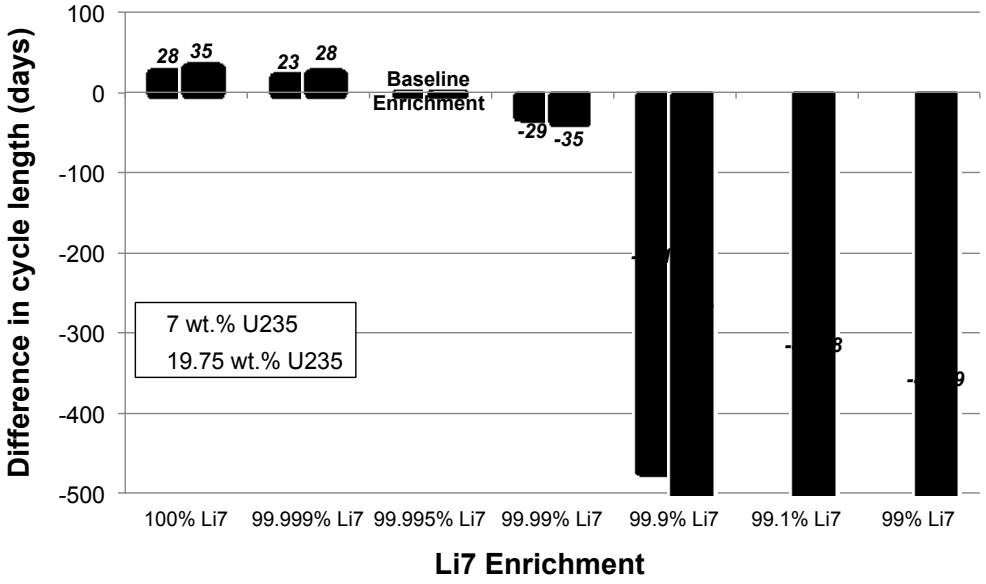

Figure 7. Difference in cycle length (days) between various ^{7}Li enrichments and the baseline enrichment in FLiBe.

The different compositions of ^{6}Li and ^{7}Li also affect the coolant TCR of the core. Figures 9 and 10 show the coolant TCR for both the 19.75 wt.% and 7 wt.% ^{235}U cases, respectively. Higher ^{7}Li enrichments give slightly better and desirable negative coolant TCRs. For example, for every 0.001% increase in ^{7}Li enrichment, the coolant TCR is improved and incremented by approximately -0.013 to -0.014 pcm/K for both ^{235}U enrichment cases. The coolant TCR for 7 wt.% ^{235}U, however, is more negative compared to that of the 19.75 wt.% ^{235}U case by on average, -0.41 pcm/K.

The improvement in coolant TCR (more negative values) at higher ^{7}Li enrichment is due to the associated decrease in the ^{6}Li isotope. During a perturbation to calculate the TCR, the coolant temperature is perturbed (increased by 30 K), while the coolant density will drop slightly (by 0.77%) from its density at the normal operating temperature, reducing its ability to moderate and absorb neutrons. For FLiBe coolant with a higher ^{6}Li composition, the total cross-section of lithium (^{6}Li + ^{7}Li) becomes more dependent on the relatively large ^{6}Li cross-section (Figure 1). Hence, if the composition of ^{6}Li is higher, the reactivity of the system when being perturbed will change more from the reactivity of normal operation, resulting in a large magnitude of coolant TCR.

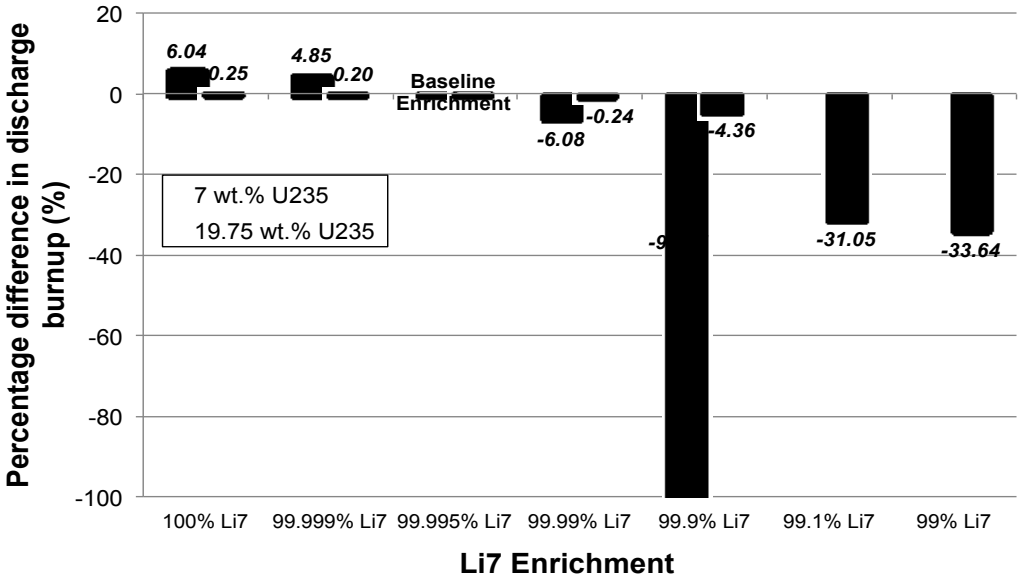

Figure 8. Percentage difference in discharge burnup between various ^{7}Li enrichments and the baseline enrichment in FLiBe.

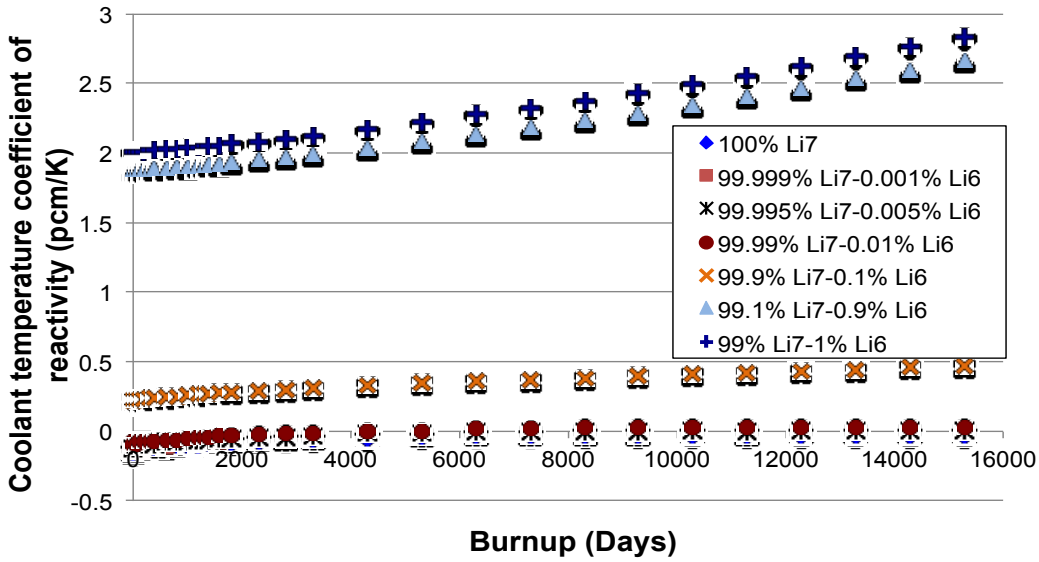

Figure 9. Coolant temperature coefficient of reactivity Case 1: 19.75 wt.% UO_2 at various ^{7}Li enrichments in FLiBe.

From the coolant TCR plots, it can also be observed that, for a long period of depletion time, the coolant TCR steadily and gradually moves towards more positive values. This pattern can be attributed to the hardening of the flux spectrum as the fuel depletes. When the spectrum hardens, fast neutrons can more easily stream through the FLiBe coolant, due to FLiBe having a small neutron cross-section, especially when the temperature is slightly increased (perturbed) and simultaneously the density is slightly decreased. These streaming neutrons later are quickly moderated by the graphite pins and block in the core, adding positive reactivity. The change of coolant TCR towards positive values is not very obvious for the 7 wt.% ^{235}U case, however, because the cycle length is relatively shorter, and the extent of spectrum hardening commensurately smaller.

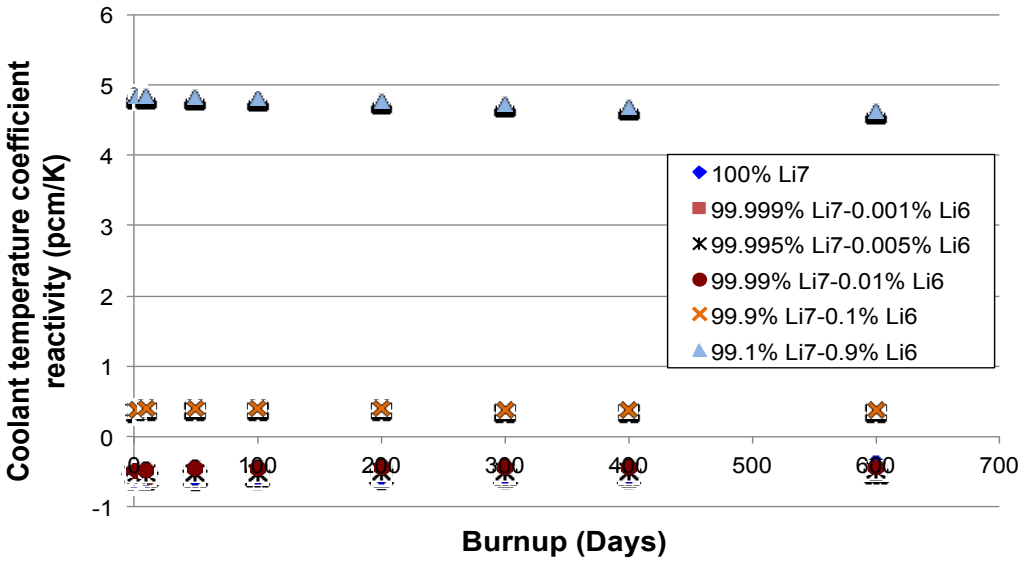

Figure 10. Coolant temperature coefficient of reactivity Case 2: 7 wt.% UO_2 at various ^{7}Li enrichments in FLiBe.

CONCLUSIONS

In summary, the clear advantages of using high ^{7}Li enrichments in FLiBe are a longer cycle length, higher discharge burnup and more negative coolant temperature coefficient of reactivity. Furthermore, high purity of ^{7}Li in FLiBe is favourable because it is one of the best ways to reduce the amount of tritium produced through the ^{6}Li(n,α)^{3}H reaction. In Stempien's study [13], it is found that by increasing the ^{7}Li enrichment in FLiBe by 0.004%, the tritium production rate and the necessary sizes of tritium capture systems are reduced by a factor of 4. The FHR White paper [6] reports that at higher ^{6}Li concentrations, approximately above 0.01%, the effect of coolant neutron absorption might outweigh its moderating capacity. Additionally, it is reported that the coolant temperature reactivity coefficient becomes positive at higher ^{6}Li concentrations, similar to results obtained in this study.

In general, higher ^{7}Li enrichments like 99.995% are commonly used in the baseline FLiBe coolant in various FHR designs such as SmAHTR, Mk1 PB-FHR and LS-VHTR. More importantly, this baseline enrichment has been implemented in the Molten-Salt Reactor Experiment (MSRE), as explained in Ref. 14. Additionally, as shown earlier, increasing the ^{7}Li enrichment by 0.005% will only increase reactivity and discharge burnup by small amounts, approximately less than 180 pcm positive reactivity and less than 6% discharge burnup. Moreover, even if pure ^{7}Li (100%) is used in the coolant, a small amount of ^{6}Li is still being produced over the lifetime of the reactor via the ^{6}Be(n,α)^{6}He reaction with subsequent beta decay to ^{6}Li (Ref. 15). ^{6}Li is never

fully depleted out as an equilibrium ^{6}Li concentration is preserved via ^{6}Li production from beryllium and its destruction by neutron absorption and following tritium production [16]. Thus, it might not be worthwhile to use the highest possible purity of ^{7}Li (100%), considering the enrichment process will likely be more difficult and expensive as suggested by Stempien [13] and Ingersoll [15].

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