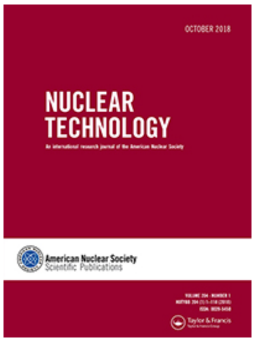

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Influence of Coolant ^{6}Li Concentration on the Neutronic Parameters of Lithium-Cooled Reactor

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Abstract — *Lithium is an attractive coolant for space nuclear reactors due to its good thermal properties and low density. The main objective of this study is to investigate the effect of coolant ^{6}Li concentration on the neutronic parameters of the lithium-cooled space reactor, with the aim to provide an appropriate reference for the purification of lithium coolant and the safety of the lithium-cooled space reactor. The neutronic calculations based on the lithium-cooled reactor are performed using the SuperMC code with the ENDF/B-VII cross-section database. The effects of coolant ^{6}Li concentration were studied over the range of 0 to 8.5 at. % as well as the corresponding effects on the depletion, helium production, neutron spectrum, and temperature reactivity coefficient. The results show that the ^{6}Li concentration of 0.01 at. % in the lithium coolant is appropriate in the lithium-cooled reactor. In this case, the neutronic parameters including the depletion, helium production, and neutron spectrum showed no obvious change compared to that of the pure ^{7}Li, and the coolant reactivity coefficient has a negative effect on reactivity.*

Keywords — *Lithium coolant, ^{6}Li concentration, reactivity coefficient, helium production.*

Note — *Some figures may be in color only in the electronic version.*

I. INTRODUCTION

Space nuclear reactors, which are compact and lightweight, highly reliable, and have a long operating lifetime, are expected to play an important role in the exploration of space, including interplanetary exploration, lunar and Martian surface missions, deep-space science missions, etc.[1] Compared with the solar option, they can operate without light conditions and provide high power. Usually, space reactors are cooled with liquid metals, liquid-heat pipes, or gas.[2] Among these, the liquid-metals–cooled space nuclear reactors have low mass and can be operated with high power densities and surface heat fluxes, thus they are suitable options for space nuclear reactor. The options available for the working fluid of liquid-metals coolant are potassium, sodium, sodium-potassium eutectic, and lithium. Lithium is an attractive coolant for space nuclear reactors due to its good thermal properties and low density. By now, several lithium-cooled space reactor concepts have been developed by different laboratories. For instance, the Atomic Energy Commission in the United States developed the first concept of the lithium-cooled space reactor, SNAP-50 (Ref. 3). The National Aeronautics and Space Administration, U.S. Department of Defense, and U.S. Department of Energy conducted the SP-100 program,[4] which developed a lithium-cooled space reactor for use as a lunar or Martian surface power station, an orbital power supply, and a power supply for nuclear electric propulsion. Currently, the lithium-cooled space reactor concept of the sectored compact space reactor for small power[5] (SCoRe-S), developed by the University of New Mexico, was designed for avoiding single-point failure. Besides, the Central Research of Electric Power Industry in Japan also developed a lithium-cooled reactor for a lunar surface power station, RAPID-L (Ref. 6).

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It is known that lithium has two stable isotopes ^{6}Li and ^{7}Li in nature. The isotope ^{6}Li with a percent natural abundance of 7.5% has a high neutron absorption cross section. The neutron absorption of the lithium coolant is enhanced with the increase of coolant ^{6}Li concentration. However, the change of neutron absorption within the coolant present in the reactor will affect the characteristics of the reactor, such as the mass, dimensions, safety margin, and operation lifetime. In order to spend the minimum cost and effort required to launch a spacecraft, the nuclear reactor should be compact and lightweight. For any reactor, negative reactivity coefficients are required for safety. In addition, gases in the form of ^{4}He, hydrogen, deuterium, and tritium produced in the lithium coolant resulting from nuclear reactions with ^{6}Li and ^{7}Li are a matter of concern. While the hydrogen and its isotopes can readily dissolve in the lithium coolant and diffuse out of the structural material with high temperatures, the helium gas cumulates in the lithium coolant. These gases can affect the strain of the structural materials and the thermal-hydraulic properties and restrict the operation lifetime of the lithium-cooled reactor. For instance, gas bubbles in lithium coolant could blanket fuel pins, leading to the reduction of the fuel pin heat transfer coefficient. Unfortunately, the helium gases are primarily produced by neutron reaction with ^{6}Li (Refs. 7 and 8) and the lithium purity is restricted owing to this reason. However, the high purification is a challenge to purification technology and the economic cost.

Currently, there is no comprehensive parameter investigation to describe the effects of varying the coolant ^{6}Li concentration on a baseline lithium-cooled space reactor design with respect to the excess reactivity, helium gas production, neutron energy spectrum, and temperature reactivity coefficients. This study uses the SuperMC version 3.10 Monte Carlo computer code with the ENDF/B-VII cross-section database to investigate the neutronic effects of varying the ^{6}Li concentration in the range of 0 to 8.5 at. % on the design concept of the SCoRe-S (Ref. 5), which is a lithium-cooled, fast neuron spectrum reactor with a mean core temperature of 1130 K. The corresponding effects of the ^{6}Li concentration on the lithium-cooled reactor are analyzed, from which an appropriate reference for the purification of the lithium coolant can be obtained.

II. REACTOR DESCRIPTION

In order to assess the effects of varying concentrations of ^{6}Li within the lithium coolant, a standard model of the lithium-cooled reactor was developed based on input from the design concept of SCoRe-S. The basic geometry of the concept design of the SCoRe-S_7 space reactor[5,9] was adopted as the baseline in this study. The cross and vertical sections of the reference reactor are shown in Fig. 1 and the main parameters are presented in Table I. The fuel pin used in the reference reactor is loaded with pellets of highly enriched (95 wt%) uranium mononitride (UN) mixed with 1.52 at. % ^{157}GdN. A 4.0-cm BeO reflector as an axial neutron reflector in the fuel pin is placed in the bottom of the UN fuel stack. In order to decrease the axial transfer of heat from the fuel to the axial neutron reflector, a 2.0-mm-thick rhenium disk is placed between the fuel stack and the BeO neutron reflector in the fuel pins. A 9.15-cm-long gas plenum in the top of the UN fuel stack and the 0.05-mm radial gap between the fuel pellets and the cladding is used for accommodating the fission gases released from the UN fuel and reducing the stresses

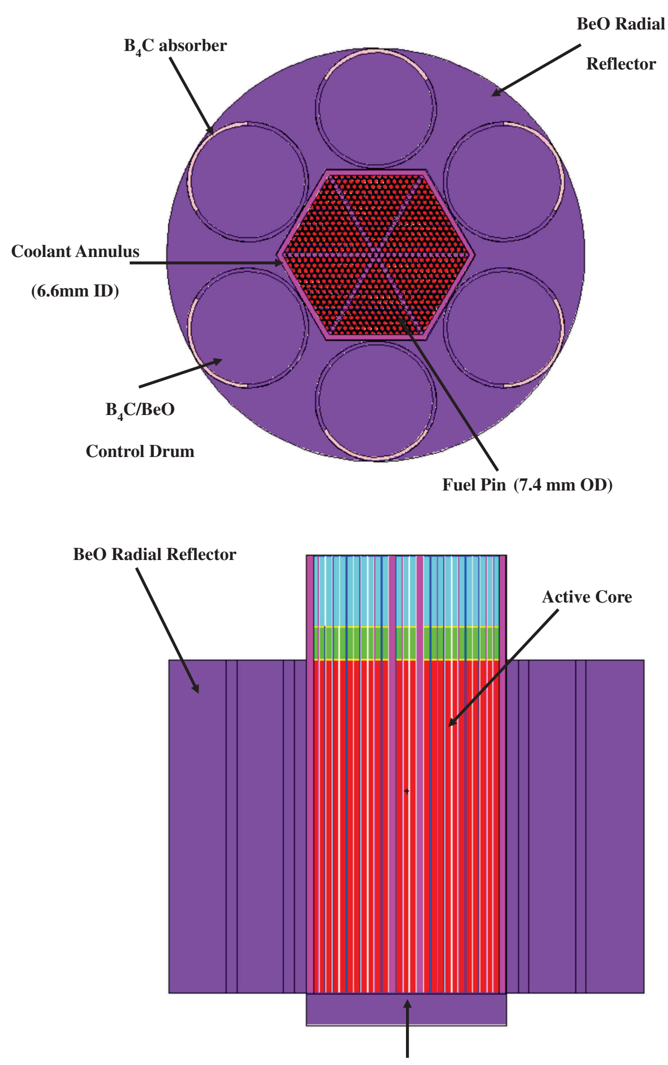

Fig. 1. Cross section of the reference reactor core.

TABLE I

Reference Reactor Core Design Parameters

Parameter	Value
Thermal power	1.6 MW(thermal)
Fuel pins	630
Fuel composition	UN
Fuel density	12.710 g/cm^3
Fuel enrichment	95 wt%
^{157}GdN added to UN fuel	1.52 at. %
Cladding	Mo-14%Re
Active core height/fuel pin height	42.5/56.0 cm
Core height/flat-to-flat ratio	2.0
Reactivity control	BeO/B_4C control drum
Radial BeO reflector thickness	16 cm
Axial BeO reflector thickness	4 cm

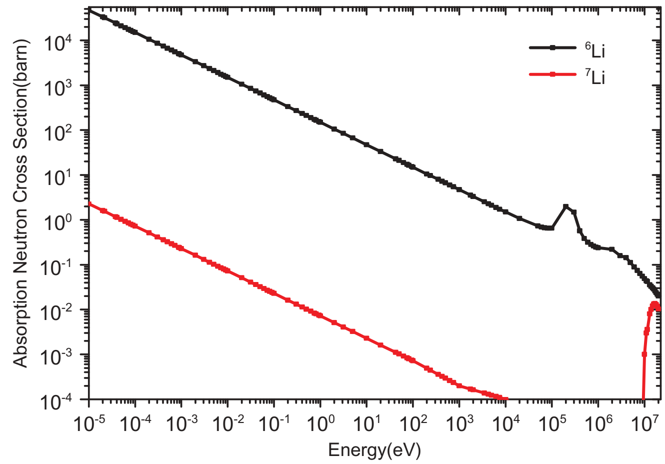

Fig. 2. Micro-absorption cross section of ^{6}Li and ^{7}Li.

in the cladding. The clad of the fuel pin is 0.5-mm thickness of Mo-14%Re. The dimension of each fuel pin is 42.5 cm high and 0.74 cm diameter. The 630 fuel pins, which are arranged in a triangular lattice at a pitch distance of 0.8 cm, are loaded in a hexagonal 0.25-cm-thick Mo-14%Re pressure vessel. The hexagonal pressure vessel is surrounded by the 16-cm thickness of the radial BeO reflector and a 4.0-cm-thick axial BeO reflector is placed in the bottom of the pressure vessel. The coating of multifoil insulation is used for thermal insulation between the pressure vessel and the BeO reflector. The six BeO rotating control drums with 5-mm-thick, 120-deg segments of B_4C neutron absorber in the radial BeO reflector are used to control the reactivity. In order to ensure the subcriticality requirement following a launch abort accident, a 0.1-mm coating of $^{157}Gd_2O_3$ is coated in the outer surface of the reactor vessel.

III. METHOD AND TOOLS

III.A. Physics Analysis

Figure 2 shows the energy-dependent cross sections for ^{6}Li and ^{7}Li over the energy range of 0.00001 eV to 20 MeV, which are extracted from the ENDF/B-VII cross-section database. Owing to the high absorption cross section of ^{6}Li, the variation of ^{6}Li concentration in the lithium coolant directly changes the neutron absorption of the coolant, which then can affect the reactor physics parameters, such as the depletion, neutron energy spectrum, and temperature reactivity coefficient.

The temperature reactivity coefficient α_T is one of the major safety parameters. It is defined as the change in reactivity with respect to the temperature. The temperature reactivity can be determined by the following formula:

$$\alpha_T = \frac{d\rho}{dT} = \frac{Keff_i - Keff_{i-1}}{(T_i - T_{i-1})Keff_i Keff_{i-1}} \quad , \tag{1}$$

where

$d\rho$ = change in reactivity

d_T = change in temperature

$Keff_i$ = effective multiplication coefficient.

The temperature reactivity coefficient is mainly induced by the coolant temperature coefficient and fuel temperature coefficient (FTC). The direct perturbation methodology is usually used to calculate the mentioned coefficients. In calculation, we assumed the temperature change is uniform throughout the core, and the temperature effect on the reactivity is expressed only by a simple temperature coefficient, called the isothermal temperature coefficient. The hot-clean temperature of the reactor core is 1130 K, while the temperature of the reflectors is constant at 300 K. Due to the multifoil insulation separating the core vessel from the BeO reflectors, the reflectors can keep low temperature.[10] The thermal expansion in the reactor core is neglected due to the compactness of the core.

The helium gas in the coolant produced by way of the neutron reaction with lithium can affect the operation lifetime and safety of the lithium-cooled reactor. Its production is highly dependent on the neutron energy spectrum and the cross section of nuclear reactions. The cross-section data from the ENDF/B-VII cross-section database indicate that the helium gas is primarily produced by the ^{6}Li(n, t), ^{6}Li(n, nd), ^{6}Li($n, n\alpha$), ^{7}Li(n, γ), ^{7}Li($n, n\alpha$), and ^{7}Li(n, nt) reactions (see in Table II).

III.B. SuperMC Calculations

The neutronic calculations are performed by using the Super Monte Carlo Simulation Program[10,11] (SuperMC 3.10 professional edition), and the cross-section data used by the SuperMC program are based on ENDF-VII evaluation files. The software has been developed by the FDS Team at the Institute of Nuclear Energy Safety Technology, China. It can perform the Monte Carlo simulation of neutron, photon, and coupled neutron-photon transport, activation calculation, depletion calculation, and medical physics analysis, and it can also be applied for critically and shielding design of reactors, etc. The SuperMC model is constructed according to the reactor geometry and used to calculate the effective multiplication factor, the temperature reactivity coefficient, neutron flux distributions in the reactor core, fuel depletion, and gas production. The calculations of the fuel depletion are based on using 10-k source particles per cycle with 50 inactive cycles and 500 active cycles, providing a computational uncertainty of less than 80 pcm. The calculated temperature reactivity coefficient for reactor cores are based on at least 10-k source particles per cycle with 50 inactive cycles and 1000 active cycles, providing a computational uncertainty of less than 30 pcm. The computational uncertainty of other parameter values is less than 1%. In the process of temperature reactivity coefficient calculation, the effect of temperature on the cross section of materials can be dealt with the on-the-fly Doppler broaden function of the SuperMC program. In addition, the tally and tally multiplier card in the SuperMC code are used for recording some parameters in the reactor core, such as the neutron spectrum and the helium production rate.

TABLE II

Nuclear Reactions for Helium Production

Reaction	Threshold Energy
$^{6}Li + n \rightarrow 2n + {}^{4}He + {}^{1}H$	$Q = -3.7$ MeV
$^{6}Li + n \rightarrow n' + {}^{4}He + {}^{2}H$	$Q = -1.5$ MeV
$^{6}Li + n \rightarrow {}^{4}He + {}^{3}H$	$Q = 4.8$ MeV
$^{7}Li + n \rightarrow 2n + {}^{4}He + {}^{2}H$	$Q = -8.7$ MeV
$^{7}Li + n \rightarrow n' + {}^{4}He + {}^{3}H$	$Q = -2.5$ MeV
$^{7}Li + n \rightarrow {}^{8}Li + \gamma \rightarrow \beta^{-} + 2\,{}^{4}He$	$Q = 2$ MeV

IV. RESULTS AND DISCUSSION

IV.A. Effects on Depletion

The effect of the coolant ^{6}Li concentration on depletion is investigated in this section. The hot-clean excess reactivity is calculated at the nominal operation thermal power of 1.6 MW and the full-power time of 10 years. The calculated values of the hot-clean beginning-of-life (BOL) excess reactivity are used to determine the full-power operational life using the fuel depletion capability in the SuperMC code. The SuperMC code calculates the changes in the isotopic composition of the fuel and the lithium coolant during operation. The reactivity depletion analysis is performed using 1-year time steps. The effect of the coolant ^{6}Li concentration on the depletion is presented in Fig. 3. From the graph we know that when the ^{6}Li concentration is above 0.5 at. %, the BOL hot-clean excess reactivity decreased significantly with increasing ^{6}Li concentration. The reactor cannot achieve 10 full power years when the coolant ^{6}Li concentration varies in the range of 5 to 8.5 at. %. When the concentration of ^{6}Li is less than 1 at. %, the change of reactivity with full power year shows no obvious change with respect to the ^{6}Li concentration of 0%. These results are mainly attributed to the change of the coolant ^{6}Li concentration in the reactor core. As the ^{6}Li concentration increases in the lithium coolant, the neutron absorption of the coolant increases, leading to the reduction of the neutron parasitic absorption in fuel. In addition, the inventory of ^{6}Li in the coolant is consumed as the evolution of the reactor, to a certain extent, which has played a role in reactivity compensation.

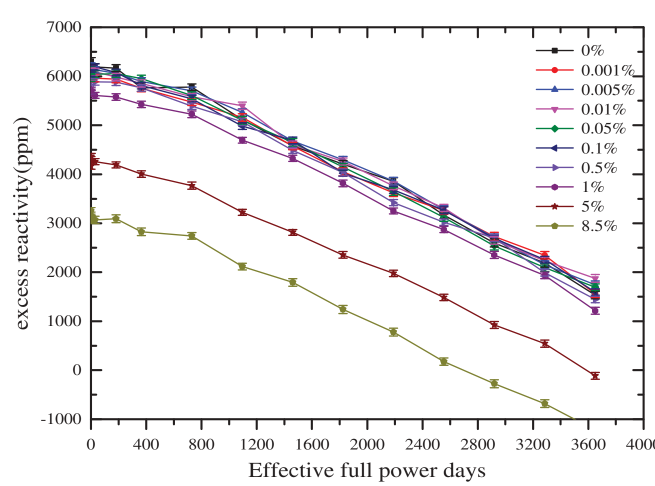

Fig. 3. Variation of excess reactivity with different ^{6}Li concentrations.

Figure 4 gives the relative neutron absorption of the coolant and fuel versus the concentration of ^{6}Li at the startup time. As can be seen from Fig. 4, the neutron absorption of the coolant increases as the ^{6}Li concentration increases. Meanwhile, when the coolant ^{6}Li concentration is less than 0.1 at. %, there is no obvious change in the neutron absorption of fuel when the neutron absorption of the lithium coolant increases. Because there is little inventory of ^{6}Li in the core, it has almost no effect on ^{235}U neutron capture. However, when the coolant ^{6}Li concentration is in excess of 0.1 at. %, the mass of ^{6}Li in the core is so high that a certain number of neutrons are wasted, which significantly leads to the reduction of ^{235}U neutron capture.

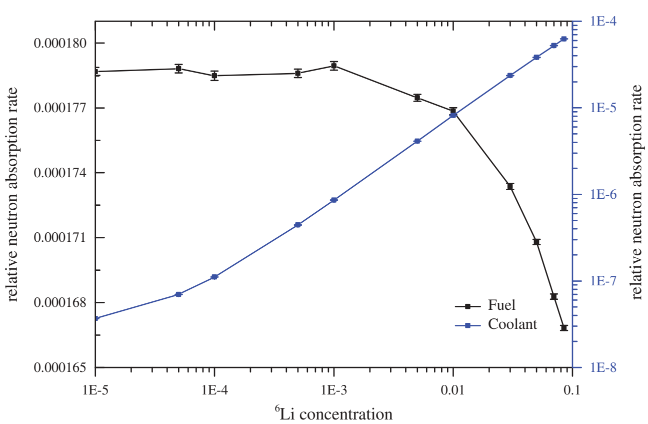

Fig. 4. Variation of relative neutron absorption rate in initial time with different ^{6}Li concentrations.

IV.B. Effects on Helium Production

The helium gas in the primary coolant of the reactor has an adverse impact on system operations because it is undissolved in the lithium coolant. The gas separator/accumulators are usually used to remove the helium gases generated in the primary coolant over the mission life. So the helium production should be as low as possible. In this section, the production of helium gas is calculated and the calculations are presented in Fig. 5. As shown in Fig. 5, after a 10-year operation, the helium production in the lithium coolant is 0.0149 g when the concentration of ^{6}Li is 0.01 at. %. The helium production shows no significant change when the concentration of ^{6}Li is below 0.01 at. %. However, when the concentration of ^{6}Li is 8.5 at. %, the production of helium is more than 7.8 g, and the inventory of helium is more than 1800 times the inventory of the helium for ^{6}Li concentration of 0%. So the ^{6}Li concentration of 0.01 at. % is recommended from the aspect of helium production.

IV.C. Neutron Energy Spectrum

The neutron energy spectrum in the reactor core in the initial time is calculated using SuperMC with 175 logarithmically spaced energy groups. Figure 6 shows the neutron energy distribution over the range of 0.001 keV to 20 MeV. The change of ^{6}Li concentration has almost no effect on the fast-spectrum neutron energy distribution. However, a change in the low-energy distribution from 0.01 eV to 1 keV is obvious when

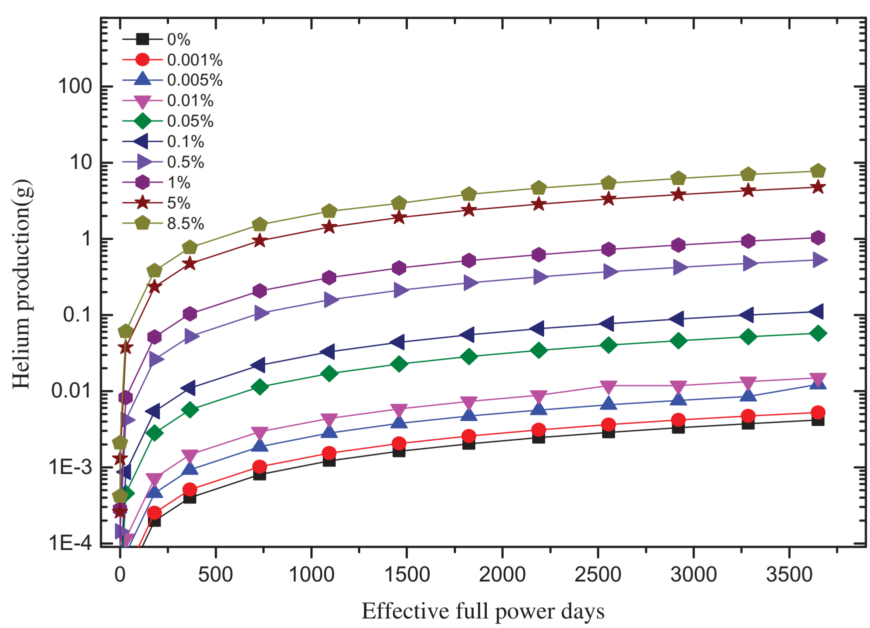

Fig. 5. Variation of helium production with different ^{6}Li concentrations.

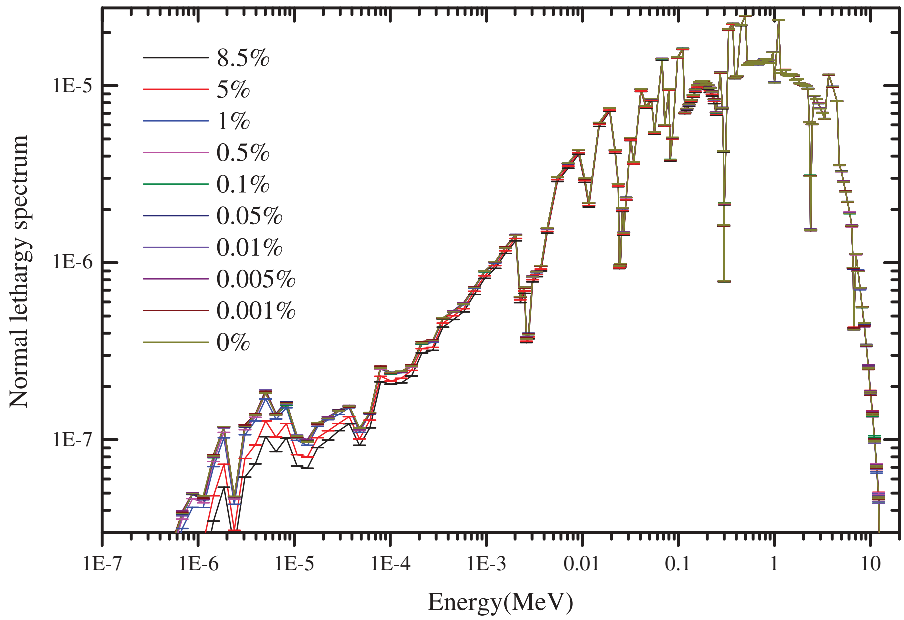

Fig. 6. Normalized lethargy spectrum of reactor core at initial time with different ^{6}Li concentrations.

the ^{6}Li concentration is more than 0.5 at. %. The reason is that the ^{6}Li has higher thermal neutron absorption cross section. However, the ^{6}Li concentration within the coolant was found to have a negligible effect on the neutron energy spectrum over the range of 0 to 0.5 at. %.

IV.D. Effects on Temperature Reactivity Coefficients

IV.D.1. Coolant Temperature Coefficient

The coolant temperature coefficient accounts for the change of reactivity when the coolant temperature varies. The variation of coolant temperature results in the following effects:

1. The density of the coolant decreases with the increase of coolant temperature. The density of lithium from 200°C to 1600°C is calculated using Eq. (2) (Ref. 12):

$$\rho(T) = 0.515 - 1.01 \times 10^{-4}(T - 200)\,. \qquad (2)$$

2. The changes in the neutron cross section of lithium are caused by Doppler effect.

In calculation, the change in the neutron cross section of the lithium coolant is calculated by the on-the-fly Doppler broaden function of the SuperMC program. Meanwhile, the coolant density is an input parameter so it can be manually modified with temperature range from 500 to 1300 K. The lithium-coolant density at different temperatures is calculated by Eq. (2). The effective multiplication coefficient for each temperature case is calculated using the SuperMC code and the results are presented in Fig. 7. As can be seen when the ^{6}Li concentration is less than 1 at. %, the coolant temperature coefficients are negative value (~1 pcm/K). However, when the concentration of ^{6}Li is above 1 at. %, changes in the excess reactivity become unremarkable as the coolant temperature rises and the coolant temperature coefficients have almost no effect on the reactivity. The main reason for this result is that the neutron absorption in the coolant increases as the ^{6}Li concentration rises. The neutron utilization in fuel increases when the coolant temperature rises because of the decrease of the neutron absorption of the coolant. In order to maintain the inherent safety features of nuclear reactor, the ^{6}Li concentration should be less than 1 at. %.

IV.D.2. Fuel Temperature Coefficient

The change of FTC is due to the Doppler broadening of the resonance region of the ^{238}U in the fuel and neutron spectrum hardening. To calculate the FTC, the fuel temperature is modified only while keeping the temperatures of other materials constant. The Doppler-broadened cross sections of materials are calculated in the temperature range from 700 to 1800 K and the results are depicted in Fig. 8. It can be seen that the FTC in the reactor core has an insignificant effect on the reactivity. The effect of the ^{6}Li concentration on the FTC is very small. Because the 95% high-enriched fuel is used in the nuclear reactor, the effect of Doppler broadening on the FTC is expected to be insignificant due to the low amount of ^{238}U in the fuel and a fast

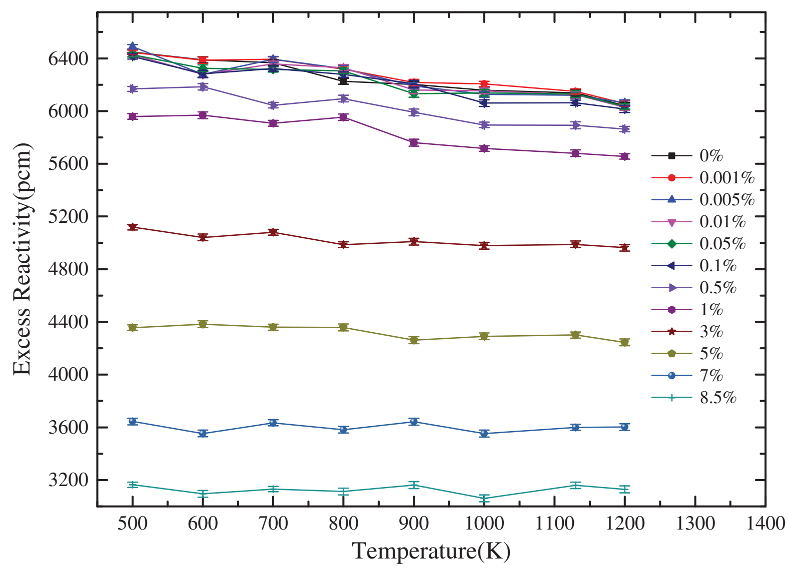

Fig. 7. Coolant reactivity coefficient estimates with different ^{6}Li concentrations.

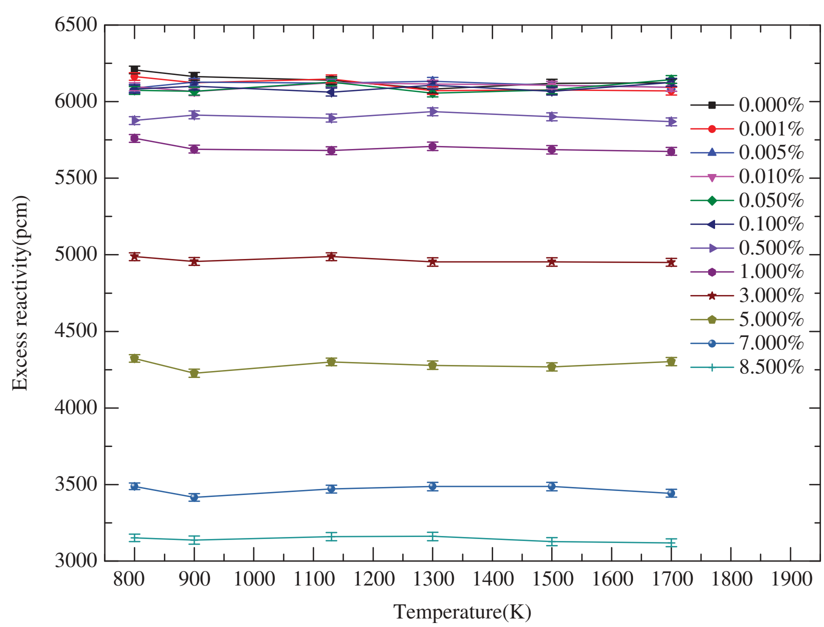

Fig. 8. Fuel reactivity coefficient estimates with different ^{6}Li concentrations.

neutron spectrum with most neutron energies above the resonance region in the reactor core (see in Fig. 6).

V. CONCLUSION

In this paper, the neutronic effect of the coolant ^{6}Li concentration on a lithium-cooled reactor is investigated by SuperMC code. The depletion calculations showed that it has an insignificant effect on excess reactivity when the concentration of ^{6}Li in the lithium coolant is less than 1 at. %, and the excess reactivity of the reactor cannot ensure the safety of the operation for 10 full power years when the coolant ^{6}Li concentration varies in the range of 5 to 8.5 at. %. The main reason is that the high neutron absorption of ^{6}Li leads to the decrease of ^{235}U neutron capture. Meanwhile, the calculations of helium production were performed and the results showed that the helium production is almost the same when the coolant ^{6}Li concentration is less than 0.01 at. %. In addition, other neutronic parameters such as the neutron spectrum, coolant temperature coefficient, and FTC are also calculated. The results show that the change in the coolant ^{6}Li concentration mainly causes a slight change in the thermal neutron region of the neutron spectrum. The temperature reactivity coefficient is an important safety parameter; the study reveals that the change in the concentration of coolant ^{6}Li has no noticeable effect on the FTC and the effect of the FTC on reactivity can be neglected. The main reason is that there is a low amount of ^{238}U in the fuel and a fast neutron spectrum in the nuclear reactor. The coolant temperature coefficient has a negative effect on the reactivity when the concentration of ^{6}Li is below 1 at. %. However, when the concentration of ^{6}Li is at or above 5 at. %, the coolant temperature coefficient has unremarkable feedback to the reactivity. It is concluded that the ^{6}Li concentration of 0.01 at. % is an appropriate reference for the base lithium-cooled reactor. In this case, the effect of the coolant ^{6}Li concentration on the neutronic parameters in a lithium-cooled reactor can be neglected, and this lithium concentration is a suitable value for a lithium-cooled reactor that satisfies a minimum of helium production and a relatively low technology requirement for lithium purification.

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