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Effect of isotope enrichment on performance of lead-lithium blanket of inertial fusion reactor

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Abstract. Simulation studies on neutron transport and material activation for lead-lithium (Pb-17Li) blanket of the inertial fusion reactor "KOYO-FAST" were performed with the PHITS code and the EASY 2005 code. Tritium breeding ratio (TBR) and productions of harmful radioactive isotopes²¹⁰Po and ²⁰³Hg in the blanket are made clear. The blanket performances improved by enrichment of stable isotope (⁶Li, ²⁰⁴Pb, ²⁰⁶Pb or ²⁰⁷Pb) are also made clear. The TBR is 1.28 for the blanket with a natural Pb-17Li alloy. The large TBR is allowed because of large blanket space in the reactor chamber. The TBR reaches 1.64 when ⁶Li is enriched to 90%. In this case, intermediate and thermal neutrons, that are necessary to produce ²¹⁰Po and ²⁰³Hg, are preferentially captured by the ⁶Li (n, α) T reaction. Then, the productions of ²¹⁰Po and ²⁰³Hg are mitigated. The enrichment of ²⁰⁴Pb leads to a small TBR, because neutrons are captured by 204 Pb due to its large cross section for (n. γ) reaction. The TBR of 206 Pb or 207 Pb enriched Pb-17Li is more or less the same with that of the natural Pb-17Li. In these cases, the productions of ²¹⁰Po and ²⁰³Hg are slightly mitigated because of the removal of ²⁰⁸Pb and ²⁰⁴Pb from the blanket.

1. Introduction

Conceptual design of a fast-ignition laser fusion reactor has been performed in Japan. According to the staged development of the reactor [1], the laser inertial fusion test experimental reactor (LIFT) [1] and the fast ignition laser fusion power plant (KOYO-FAST) [2] are being designed. In the design of the KOYO-FAST, lead lithium alloy (Pb-17Li) is selected as tritium (T) breeder in the liquid blanket system. The first wall of the chamber is protected by the film flow of Pb-17Li from high-energy neutrons, a particles and high-temperature debris. These liquid metal technologies prolong the life time of the reactor chamber and mitigate the difficulties on the maintenance scenario. Some key issues such as film flow stability [3], material compatibility [4], T monitoring [5], T recovery technology [6, 7] and other safety related phenomena [8] are being studied.

Some harmful radioactive nuclides are produced by the activation of Pb-17Li under neutron irradiation. The activation affects on the reactor safety during the reactor operation and maintenance as well as the

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processing of the activated alloy for re-use or final disposal. Some articles (see, for example, L. Petrizzi et al. [9] and G. Casini et al. [10]) analysed the activation of a Pb-Li blanket in magnet confinement fusion reactors, and reported the associated safety effects. Polonium-210 (²¹⁰Po) and mercury-203 (²⁰³Hg) are widely recognized as the major radioactive nuclides produced in Pb-17Li and are of special concern. ²¹⁰Po is a radiotoxic alpha-ray-emitting nuclide with a half-life of 138.4 days and is highly mobile due to its high vapour pressure. ²⁰³Hg is a radiotoxic gamma-ray-emitting nuclide with a half-life of 46.6 days. Their production must be quantified to plan the maintenance scenario and to prepare contingency plans for the accidental release of radioactive nuclides from the blanket system. Simultaneously, ways to suppress the production of harmful radioactive nuclides must also be investigated.

Natural lithium (Li) consists of two stable isotopes: Li-6 (⁶Li) and Li-7 (⁷Li). The T breeding ratio (TBR) is known to increase for high concentrations of ⁶Li in Li. The technology of ⁶Li enrichment has been investigated [11]. Lead (Pb) has a large cross section for the (n, 2n) reaction for fast neutrons. Natural Pb consists of the four isotopes ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb. Enriching the Pb isotopes by laser separation and gas centrifugation is possible. It is known that Pb-isotope-enriched alloys have distinctive nuclear characteristics. For example, ²⁰⁸Pb-enriched liquid Pb has been proposed as coolant for fast reactors and acceleration driven systems instead of natural Pb because of its small neutron absorption and moderation [12]. The beneficial effect of the Pb isotope enrichment on the performance of fusion blanket systems has rarely been discussed, and the information is limited.

Simulation studies on neutron transport and material activation of the Pb-17Li blanket of the "KOYO-FAST" were performed. The purpose of the present study is to make clear the tritium breeding performance and the productivity of ²¹⁰Po and ²⁰³Hg of the blanket. The effect of the isotope enrichment on these performances is clarified.

2. Method and model for neutron transport analysis and activation calculation

The KOYO- FAST has four reactor chambers, and the fusion output of each chamber is 800 MW. The output is corresponding to the generation of 2.84×10^{20} neutrons/s with the energy of 14.1MeV. The desired lifetime of the reactor chamber is 20 years. However, the final optical systems have a life time of approximately 2 years, and must be replaced during the maintenance procedure while the reactor is stopped. The availability of the reactor is assumed to be 50% due to the required high frequency of maintenance. In this case, the total operation period of the blanket under neutron irradiation is roughly estimated to be 10 years.

Figure 1 (a) shows the geometry of the reactor chamber. The structure of the reactor chamber is simplified by the removal of the final optics etc. due to its small volume ratio. A point neutron source is situated at the position of the fast ignition. The vessel is made of JIS-STBA26. The structural material of the blanket is the reduced-activation ferritic martensitic steel JLF-1 and SiC/SiC. In the neutron transport calculation, the uniformly mixed breeder layer is simulated. The compositions of the blanket is 90vol.% Pb-17Li, 5vol.% JLF-1 and 5vol% SiC/SiC. The chemical composition of JIS-STBA26 and JLF-1 steels are Fe-9Cr-1Mo-0.45Mn-0.15C-0.07Ta -0.5Si -0.05N (typical contents, wt%) and Fe-9Cr- 2W- 0.5Mn- 0.1C- 0.07Ta- 0.05Si- 0.05N (typical contents, wt%), respectively. The average neutron-wall loading is 4.2 MW/m².



Fig.1 (a) Geometry of reactor chamber (b) neutron flux distributions in reactor chamber by natural Pb-17Li and ⁶Li-enriched Pb-17Li

The neutron spectra at various positions in the reactor chamber are obtained by the PHITS Monte Carlo code [13] with the JENDL 4.0 nuclear data library [14] for neutrons with energy between 1.0×10^{-5} MeV and 14.918

MeV. The neutron spectrum for each region of the reactor chamber is used for the following activation analysis. The activation analysis is performed with the EASY2005 code package containing the FISPACT-2005 code [15]. The group structure is GAMII. The period of the neutron irradiation is the total operation period of the blanket, i.e. 10 years. Then, the activation conditions are clarified for the various periods as 1 second, 1 min, 1 hour, 1 day, 30 days, and 1–10 years (with one-year increments). The dilution of the radioactive nuclides in the blanket loop by the circulation of the liquid breeder is not taken into account.

The natural isotopic abundance ratios of ⁶Li and ⁷Li are 7.5% and 92.5%, respectively. Those of ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb, and ²⁰⁸Pb are 1.4 atom%, 24.1 atom%, 22.1 atom%, and 52.4 atom%, respectively. These isotopes have their own microscopic cross sections for nuclear reactions. Fig. 2 shows the microscopic cross section of these isotopes by the JENDLE 4.0 nuclear library. Pb has larger cross section for the (n, 2n) reaction for fast neutrons than Be as shown in Fig. 2 (a). The cross section for the (n, 2n) reaction of ²⁰⁴Pb is the steepest and that of ²⁰⁷Pb is the widest in the isotopes. ²⁰⁴Pb has the largest cross section for (n, γ) reaction. The ²⁰⁴Pb cross section for the (n, γ) reaction is three orders magnitude larger than ²⁰⁸Pb cross section is located in the lower energy region than other isotopes.

Table 1 shows the conditions of isotope concentrations in the Pb-17Li in the current work. Each Pb isotope is enriched to 90% in the Pb isotopes to clarify the effect of the each isotope on the blanket performance. Then, the ²⁰⁸Pb, which is the heaviest and initially included at the highest concentration in the natural Pb, is 8.3 mol%. These compositions of Pb-17Li are held fixed during the reactor operation and do not change by burn up or nuclear transformation and. The concentrations of impurities in the Pb-17Li are not taken into account in the current work to feature the effect of the isotope enrichment on the blanket performance.



Fig. 2 Microscopic cross sections of ⁶Li, ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb

	Natural	⁶ Li enrich	²⁰⁴ Pb enrich	²⁰⁶ Pb enrich	²⁰⁷ Pb enrich
⁶ Li	1.29	15.3	1.29	1.29	1.29
⁷ Li	15.7	1.7	15.7	15.7	15.7
²⁰⁴ Pb	1.16	1.16	74.7	0	0
²⁰⁶ Pb	20.0	20.0	0	74.7	0
²⁰⁷ Pb	18.3	18.3	0	0	74.7
²⁰⁸ Pb	43.5	43.5	8.3	8.3	8.3
Total	100	100	100	100	100

Table 1 Isotope conditions of Pb-17Li (unit: %)

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3. Results and discussions

3.1. Performance of natural Pb-17Li blanket

The TBR is 1.28 as presented in Table 2. This value is greater than that obtained for magnetic- confinement demonstration reactors [15, 16], because the blanket space is not encroached upon by a magnet and the covering ratio of the plasma is nearly 100 %. T is generated mainly in the blanket by the ⁶Li (n, α) T reaction with the neutron- multiplication (n, 2n) reaction by Pb (Table 2). When the initial T inventory and the T recovery ratio are assumed as 20 kg and 90%, respectively, the doubling time of tritium is estimated approximately 1 year.

The neutron spectra in the liquid wall and the blanket in the region indicated in Fig. 1 (a) are shown with solid blue lines in Fig. 3 (a). The drop in the neutron flux near the neutron energy of 0.24 MeV is due to the large peak in the cross section of ⁶Li (n, α) T as shown in Fig. 2 (b). Fig. 1 (b) shows neutron flux distributions for high and low energy neutrons in reactor chamber. These figures indicates that neutrons are uniformly diffused in the blanket. The radial profiles of T, 210 Po and 203 Hg productions in this region are shown in Figs. 4 (a), (b) and (c), respectively. The typical processes to produce 210 Po and 203 Hg from Pb are given as follows;

$${}^{208}\text{Pb}(n,\gamma) \xrightarrow{209}\text{Pb} \xrightarrow{\beta} {}^{209}\text{Bi} \xrightarrow{209}\text{Bi}(n,\gamma) \xrightarrow{210}\text{Bi} \xrightarrow{210}\text{Bi} \xrightarrow{\beta} {}^{210}\text{Po}, \tag{1}$$

204
Pb $(n, 2n)^{203}$ Pb $\rightarrow {}^{203}$ Pb $\xrightarrow{\beta-203}$ Tl $\rightarrow {}^{203}$ Tl $(n, p)^{203}$ Hg, (2)

206
Pb $(n, \alpha)^{203}$ Hg. (3)

Table 2 TBR and production of ²¹⁰Po and ²⁰³Hg in reactor chamber after continuous operation for 10 years

		Natural	⁶ Li enriched	²⁰⁴ Pb enriched	²⁰⁶ Pb enriched	²⁰⁷ Pb enriched
		Pb-17Li	Pb-17Li	Pb-17Li	Pb-17Li	Pb-17Li
TBR		1.28	1.63	0.521	1.31	1.33
(T_6, T_7)		(1.259, 0.024)	(1.635, 0.03)	(0.498, 0.023)	(1.285, 0.024)	(1.306, 0.024)
Production	²¹⁰ Po	9.17x10 ⁻⁶	7.54×10^{-7}	5.19x10 ⁻⁷	1.72×10^{-7}	1.81x10 ⁻⁶
[g per Pb-17Li kg]	²⁰³ Hg	5.02x10 ⁻⁶	3.88x10 ⁻⁶	6.41x10 ⁻⁵	1.00x10 ⁻⁵	3.01x10 ⁻⁶



Fig. 3 Blanket neutron spectra in the horizontal cross section at level of ignition point in chamber (a) Natural Pb-17Li and ⁶Li enriched case, (b) ²⁰⁴Pb- or ²⁰⁷Pb- enriched case

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Fig. 4 Radial profiles of nuclide productions, (a) T, (b) ²¹⁰Po and (c) ²⁰³Hg

3.2. Effect of ⁶Li enrichment

The TBR is 1.64. The neutron spectra in the ⁶Li-enriched blanket are harder than those in the blanket without enrichment as shown in Fig. 3 (a). The radial profile of tritium production in liquid wall and blanket region (Fig. 4 (a)) indicates that its slope is steeper than that with natural Pb-17Li. The thermal neutron flux in this blanket is much smaller than that without isotope enrichment as shown in Fig. 1 (b). These features are attributed to the reaction rate of the ⁶Li (n, α) T reaction is dominant in the blanket.

The productions of ²¹⁰Po and ²⁰³Hg are reduced by 91.8% and 22.7%, respectively rather than that without enrichment as presented in Table 2. The reduction attributes to that the reaction rate for the (n, γ) reaction of the Pb isotopes in ⁶Li-enriched Pb-17Li blanket becomes smaller than that in natural Pb-17Li blanket because the reaction rates of ⁶Li (n, α) T becomes larger by its enrichment.

3.3. ²⁰⁴*Pb-enriched case*

The TBR of ²⁰⁴Pb enriched blanket is 0.52 and is much less than that without the enrichment (Table 2). The reason is the large cross section of the (n, γ) reaction. The ²⁰⁴Pb-enriched blanket has lower neutron flux for the energy less than 0.01 MeV as shown in Fig. 3 (b) and the neutron spectrum had a noticeable drop in the neutron flux around 10^{-3} MeV. The drop is attributed to the cross section for the (n, γ) reaction, which has the resonance region in the energy range between 10^{-1} and 10^{-3} MeV. The resonance region overlaps the cross section of ⁶Li (*n*, *a*) T. The production of ²¹⁰Po was reduced by 94.3%. However, the production of ²⁰³Hg is 12 times larger than the natural Pb-17Li case. Fig. 5 shows the trend of isotope productions based on the results of FISPACT calculation. The trend indicates that ²⁰³Hg is produced from ²⁰⁴Pb via the formation of ²⁰³Pb and ²⁰³Tl as presented in Eq. (2).

3.4. ²⁰⁶*Pb- or* ²⁰⁷*Pb-enriched case* ²⁰⁶*Pb-* or ²⁰⁷*Pb-* enrichment leads to more or less the same TBR vis-à-vis natural Pb-17Li as presented in Table 2. The neutron spectra in these blankets are close to those in natural Pb-17Li blanket (Fig. 2 (b)). Fig. 4 (a) indicated that the radial profile of the T production in the ²⁰⁶Pb- or ²⁰⁷Pb- enriched blanket is close to that of

Indicated that the radial profile of the 1 production in the ²⁰⁷Pb- or ²⁰⁷Pb- enriched blanket is close to that of natural Pb-17Li blanket because the ⁶Li (n, α) T reaction is dominant for T production in these cases. The enrichment of ²⁰⁶Pb or ²⁰⁷Pb leads to the suppression of ²¹⁰Po production. The production of ²¹⁰Po is reduced by 98.1% and 80.3% in ²⁰⁶Pb and ²⁰⁷Pb enriched cases, respectively, rather than that without the isotope enrichment. The suppression is due to the removal ²⁰⁸Pb, which produces ²¹⁰Po by Eq. (1), and the concomitant decrease in relative content of ²⁰⁸Pb. In the ²⁰⁶Pb enriched case, the production of ²⁰³Hg becomes 2 times larger than the natural Pb-17Li case due to the reaction given by Eq. (3). The production of ²⁰³Hg is reduced by 40.0% in the ²⁰⁷Pb enriched case due to the removal of ²⁰⁴Pb and ²⁰⁶Pb, which produces ²⁰³Hg as expressed by Eq. (2) and (3).



Fig. 5 (a) Production of various nuclides in ²⁰⁴Pb-enriched Pb-17Li blanket (b) Production pathways of Hg isotopes from Pb isotopes

4. Conclusion

Simulation studies on neutron transport and material activation for lead-lithium (Pb-17Li) blanket of the inertial fusion reactor "KOYO-FAST" were performed. The blanket performances improved by enrichment of some stable isotopes (⁶Li, ²⁰⁴Pb, ²⁰⁶Pb and ²⁰⁷Pb) were investigated. Major conclusions are follows;

- (1) The TBR is 1.28 for natural Pb-17Li blanket. The large TBR is allowed because the blanket space is not limited in the design of inertial fusion reactor, in which superconducting magnet coils are not necessary for plasma confinement.
- (2) The TBR is 1.64 for ⁶Li-enriched blanket. The productions of ²¹⁰Po and ²⁰³Hg are reduced by 91.8% and 22%, respectively. The productions of these isotopes are mitigated because intermediate and thermal neutrons, that are necessary to produce ²¹⁰Po and ²⁰³Hg, are preferentially captured by the ⁶Li (n, α) T reaction.
- (3) TBR is 0.521 for ²⁰⁴Pb-enriched blanket. The small TBR is caused because neutrons are captured by ²⁰⁴Pb due to its large cross section for (n. γ) reaction. The production of ²⁰³Hg is promoted by 128% because of the production of 203 Ti, which transmutes to 203 Hg by (n. γ) reaction.
- (4) The enrichment of ²⁰⁶Pb or ²⁰⁷Pb leads to more or less the same TBR with natural Pb-17Li. The production of ²¹⁰Po is reduced by 98.1% and 80.3% for ²⁰⁶Pb- and ²⁰⁷Pb-enriched cases, respectively. The production of ²⁰³Hg is reduced by 40% in the ²⁰⁷Pb-enriched case because of the removal of ²⁰⁴Pb and ²⁰³Pb from the blanket.

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