M&C 2023 - The International Conference on Mathematics and Computational Methods Applied to Nuclear Science and Engineering · Niagara Falls, Ontario, Canada · August 13 – 17, 2023

Burnup and Analytical Analysis of Neutron Poison Accumulation in the Beryllium Reflector of the Jules Horowitz Reactor

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ABSTRACT

The Jules Horowitz Reactor (JHR) is a Materials Testing Reactor (MTR) under construction at CEA Cadarache in southern France. Its primary function is to provide materials and fuel irradiation testing capabilities in its adjustable experimental positions, while also being able to produce medical radio-isotopes, for an international consortium of industry and research partners.[1]

Like many other MTRs, JHR has a beryllium reflector to reduce neutron leakage from the core which not only reduces core size and the amount of fuel necessary for criticality, but also provides a more uniform power distribution. This last point is particularly of interest for running experiments in order to assure homogeneity of conditions across tests. Beryllium has many excellent properties such as a high neutron multiplication, low absorption cross section, and high scattering cross section of neutrons. It is expensive and toxic, however, making replacing beryllium during a MTR's lifetime unappealing.

The focus of this paper is how neutron poison accumulation affects the reflector's performance over time. Two different approaches were used with Serpent 2 Monte Carlo neutronics code[2] , focusing on ^{6}Li, ^{3}He and ^{3}H, which are all part of the same beryllium transmutation chain that could affect the primary function of the beryllium reflector. The results indicate that after ten years, the accumulation of ^{6}Li and ^{3}He would result in reductions in the thermal neutron flux of up to 2% and 5%, respectively, in the reflector experimental sites, while the effect of ^{3}H was negligible. Furthermore, the decrease in reactivity induced by poisons during this period was also minimal. Therefore, this neutronic assessment suggests that it will be unnecessary to replace beryllium components during the first ten years of operation.

KEYWORDS: JHR, Monte Carlo neutronics, MTR, beryllium, reactor poisons.

1. REACTOR POISONING OF BERYLLIUM

Starting with a fresh core and reflector, the atomic concentration of ^{6}Li increases as fast neutrons interact with ^{9}Be producing ^{6}He and an α particle. ^{6}He has a half-life smaller than a second, so

it quickly decays into ^{6}Li releasing a β particle. The atomic concentration of ^{6}Li decreases as it absorbs thermal neutrons producing tritium and α particles.

$$^{9}\text{Be} + \text{n} \longrightarrow {}^{1}_{0}\text{Be}^{*} \longrightarrow {}^{6}\text{He} + \alpha - 0.64\text{MeV}$$
$$^{6}\text{He} \longrightarrow {}^{6}\text{Li} + \beta^{-}$$
$$^{6}\text{Li} + \text{n} \longrightarrow {}^{3}\text{H} + \alpha + 4.7\text{MeV}$$

Tritium accumulates gradually throughout reactor lifetime produced by ^{6}Li, as seen above. It decays into ^{3}He emitting a β particle. ^{3}He, in turn, undergoes a (n,p) reaction to produce tritium creating a feedback loop between the two isotopes.

$$^{3}\text{H} \longrightarrow {}^{3}\text{He} + \beta^{-}$$
$$^{3}\text{He} + \text{n} \longrightarrow {}^{3}\text{H} + \text{p}$$

Tritium has a low solubility in beryllium and builds up in micro-voids in the matrix as well as in helium bubbles and oxide inclusions.[3] It is produced during reactor operation and constantly decays into ^{3}He. ^{3}He is depleted during reactor operation. As the reactions of ^{9}Be and ^{6}Li depend on neutrons while the reactions of ^{6}He and ^{3}H are decay processes, the former are predominant during reactor operation and the latter increases in importance during reactor outage.

The Bateman Equation describes the rate of change in concentration of an isotope over time. It balances the production of a species with its removal, both due to a variety of nuclear reactions. By applying this, one can obtain the change in atomic concentration of ^{6}Li, $N_{^{6}\text{Li}}$, which is described by:

$$\frac{dN_{^{6}\text{Li}}}{dt} = N_{^{9}\text{Be}}\sigma^{\text{Be}}_{(n,\alpha)}\phi - N_{^{6}\text{Li}}\sigma^{\text{Li}}_{(n,\alpha)}\phi, \tag{1}$$

where $\sigma^{\text{Be}}_{(n,\alpha)}$ is the cross section of ^{9}Be for neutron absorption, $\sigma^{\text{Li}}_{(n,\alpha)}$ is the cross section of ^{6}Li for neutron absorption, and ϕ is the neutron flux. This equation assumes that the ^{6}Li absorption cross section is the dominant reaction, that the atomic concentration of ^{9}Be remains constant throughout the reactor's lifetime, and that neutron flux is constant. Thus, it is expected that the amount of ^{6}Li would eventually plateau, reaching a maximum concentration.

The atomic concentration of ^{3}He is given by:

$$\frac{dN_{^{3}\text{He}}}{dt} = \lambda_{^{3}\text{H}}N_{^{3}\text{H}} - N_{^{3}\text{He}}\sigma^{\text{He}}_{(n,p)}\phi, \tag{2}$$

where λ is the decay constant of tritium, N is isotope atomic density, $\sigma^{\text{He}}_{(n,p)}$ represents the cross section of helium neutron absorption, and ϕ is neutron flux. Tritium atomic concentration, in turn, is given by:

$$\frac{dN_{^{3}\text{H}}}{dt} = N_{^{6}\text{Li}}\sigma^{\text{Li}}_{(n,\alpha)}\phi - \frac{dN_{^{3}\text{He}}}{dt}, \tag{3}$$

where the terms are as previously described.

Table 1: JHR parameters.

Power (MW)	100
Fuel type	U_3Si_2
Fuel enrichment (%)	27.5
On cycle (days)	25
Cycles (per year)	6
Annual stop (days)	65

Table 2: Reflector material composition.

Element	%
Be-9	97
H-1	1.7
O-16	1.4
C	0.29
Al-27	0.033
Mg-24	0.023
Fe-56	0.019
Si-28	0.017
Mg-25	0.003
Mg-26	0.003
Si-29	0.001
Si-30	0.001
Fe-54	0.001
Fe-57	<0.001
Fe-58	<0.001

2. MONTE CARLO NEUTRONICS ANALYSES

2.1. Assumptions and simulation conditions

Several analytical studies have been carried out simulating the accumulation of poisons in reactors such as ATR, SAFARI, BR2, and MARIA.[3,4,5,6] All of these have retroactively benefited from a detailed operational history as well as experimental results to verify simulations. The aim of this paper is to make a credible estimate of the future accumulation of neutron poisons in the JHR reflector and simulate the effect this has on reactivity and irradiation performance. The analytical method benefits from being fast. However, in the absence of experimental data, a burnup calculation is necessary to validate results. Several reactors have required the replacement (or rotation) of beryllium components. Hence, simulations in this paper were limited to 10 years. Reactor properties are found in Table 1.

This work applied a Serpent 2 model of JHR generated by CEA's python tool GADGET, which has been benchmarked in the past.[7] Serpent 2 calculations were performed with code version 3.1.32 using JEFF3.1.1 nuclear data libraries. Burnup calculations were calculated simulating $5 \cdot 10^7$ neutrons on each depletion step ranging from 20 to 50 days. Analytical solutions were obtained from criticality calculations with $1 \cdot 10^9$ simulated neutrons. Each of these methods is detailed further in the following sections.

Due to the asymmetric nature of the reflector, radial and axial discretisation were performed to separate beryllium into multiple rings with individual material definitions. As such, the impact of radial poison distribution can be better quantified due to the fact that the vast majority of the poison accumulation happens in the regions of the reflector closest to the core. The geometry used in both calculations is shown in the Figure 1. Details of the material composition of the reflector, including

impurities, are found in Table 2. Predicting the exact amount of neutron poisoning accumulated in the beryllium reflector of the JHR over its lifetime is a fools errand. The biggest source of uncertainty is knowing exactly how many effective full power days the reactor will be used for in the future.

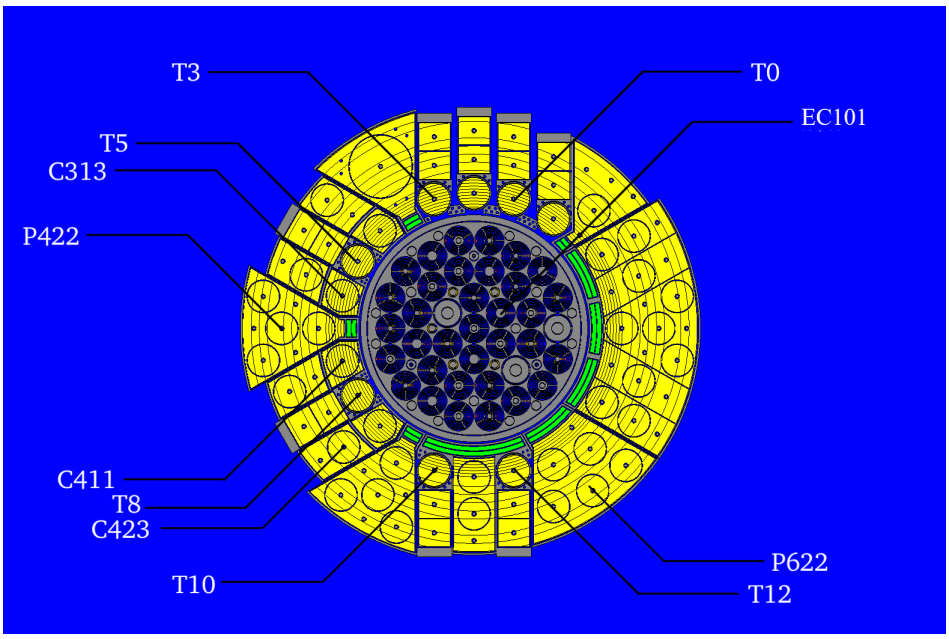

Figure 1: Serpent constructive solid geometry model of the JHR with the beryllium reflector (yellow) split in radial sectors. Experimental sites are labelled.

2.2. Burnup method

The burnup calculation in Serpent is based on solving N Bateman equations corresponding to N nuclides in the system in order to find the concentration of each isotope at any time. Serpent uses a two-step approach for burnup calculations. In the first step, neutron transport using Monte Carlo is used to produce a detailed distribution of neutron fluxes and reaction rates throughout the system. Then, in the second step, the Bateman equations are solved using the neutron flux and reaction rates from the first step to calculate the evolution of the isotopes in the system over time. The accumulation of poisons is a non-linear effect as they affect the neutron flux spectrum; reaction rates need to be updated at every step. Thus, small burnup steps need to be used. A simplified refuelling scheme with a fresh core every 600 days was used. A more precise refuelling and following a complex pattern of on and off cycles would increase computational cost. This would be an unnecessary level of detail for the purposes of providing a baseline estimate with which to compare analytical results.

2.3. Analytical method

The analytical method used in this paper is based on a method established by Wróblewska to quantify the accumulation of poisons in the MARIA reactor.[3] It applies the Bretscher and Snelgrove general solution to Equations 1, 2 & 3, solving the accumulation of reactor poisons in a homogeneous beryllium block. The method starts off by obtaining a 281-energy group flux distribution

from a Serpent calculation, after which the poison concentrations can be solved analytically by integrating the Bateman equations over discrete time steps with a MATLAB script. The off-periods of the reactor are accounted for by integrating over time steps with zero flux, during which the concentrations of the poisons evolve via decay. With this approach, time evolution of ^{3}H, ^{3}He and ^{6}Li concentrations can be solved.

This approach assumes constant flux during on-cycle operation and ignores the feedback effects of the poison accumulation, unlike the burnup method. As such, it is a transmutation calculation specific to studying the changes in beryllium, offering significantly lower computational cost in comparison to the burnup calculation. A core in middle of cycle configuration was used as a fixed and constant neutron source, as this is considered representative of the average neutronic conditions of the reactor over the span of its lifetime. Figure 2 illustrates the on-cycle variation of JHR thermal and fast flux distribution in the reflector region, from which can be seen that the middle of cycle flux is a good average, and that the fast-to-thermal flux ratio remains near constant throughout the cycle.

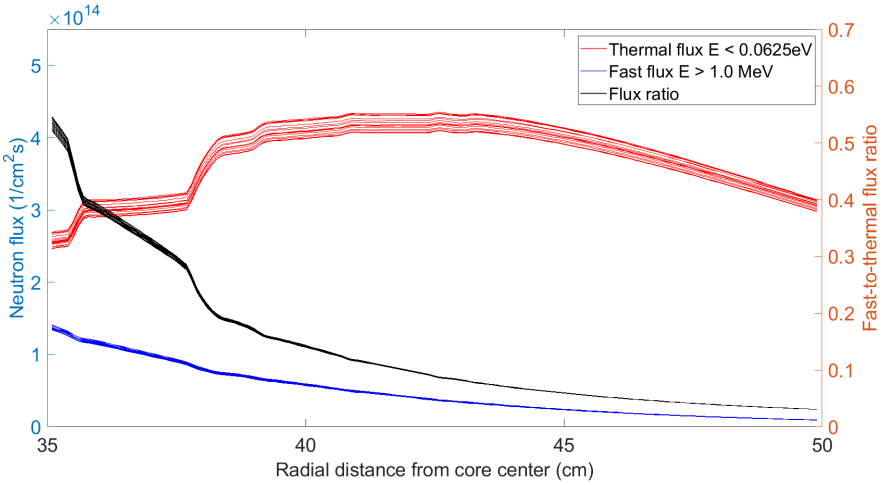

Figure 2: On-cycle variation of fast and thermal flux distribution in the JHR reflector as a function of radial distance from the core center.

Although the same method is used in both studies, the MARIA reactor uses beryllium as a moderator in the core, whereas in JHR it constitutes the reflector. Furthermore, instead of a homogeneous block this study used bespoke neutron flux spectra for each discretised beryllium segment. This provides an opportunity to solve the radial and axial distribution of the reactor poisons. Lastly, the impacts of these poisons on the neutronic conditions were analysed with another set of Serpent calculations. A total of 10^9 neutrons were simulated for both the initial calculation, as well as each of the following calculations studying the impacts of poisoning.

2.4. Measuring the effects of neutron poisoning

Once the estimates for accumulation of poisons were completed, the concentrations obtained for ten years of predicted JHR operation were inserted back into the discretised material definitions of the beryllium blocks. With these in place, the calculations were run again with otherwise identical geometry and setup. Other forms of irradiation damage and activation were ignored in these

analyses, and the effects of each poison (^{3}H, ^{3}He and ^{6}Li) were studied with a separate Serpent calculation.

Detectors were placed in different sites of the JHR in order to measure changes in neutron flux. These sites are illustrated in Figure 1. Experimental site EC101, in the core, was selected to show that beryllium neutron poisoning has no effect. Sites C313, P422, C411, C423, and P622 represent different experimental positions in the beryllium reflector at different distances from the core. Positions T0, T3, T5, T8, T10, and T12 show locations where Molybdenum-99 is expected to be produced. Emphasis was placed on determining the thermal flux as this is the most important in reflector experimental sites. Furthermore, the effect of the poisoning on the reactivity of the reactor was also considered.

3. RESULTS AND DISCUSSION

3.1. Estimating neutron poison population

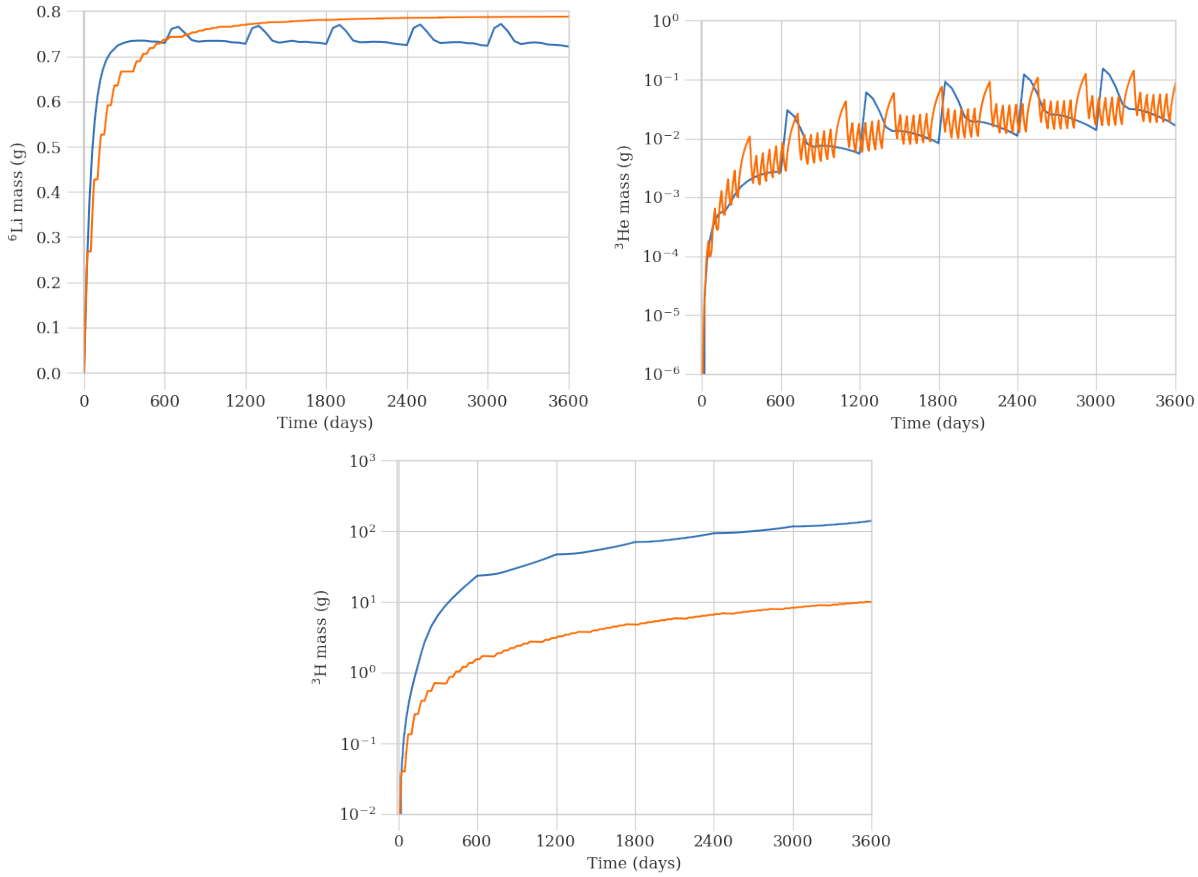

Figure 3: Accumulation of ^{6}Li, ^{3}He, and ^{3}H over time in the JHR beryllium reflector, analytical solution (orange) and burnup calculation (blue).

Figure 3 shows that the burnup and analytical results follow similar trends. Differences in graph shape for ^{6}Li and ^{3}He are attributable to the different assumptions and simplifications made such as the burnup calculation refuelling in 600 day cycles while the analytical solution following a more

detailed cycle described in Table 1. Both ^{6}Li solutions follow a generally asymptotic trend, as expected. Over the ten year period studied, the maximum total amount of ^{6}Li in all of the beryllium reflector blocks predicted by the burnup calculation is 0.771g which is in good agreement with the value predicted by the analytical solution 0.787g. There is also good agreement between the two methods for ^{3}He, with the burnup calculation predicting a maximum amount of 0.153g and the analytical solution estimating 0.157g.

Conversely, the analytical and burnup analyses of tritium accumulation show considerable disagreement, the cause of which remains to be identified. The burnup calculation routine predicted 140g of tritium in the reflector blocks, whereas the analytical solution predicted 9.9g. Since the analytical solution only accounts for the tritium accumulated via one reaction chain, the discrepancy most likely originates from other reaction chains experienced by the elements described in 2.

Figure 4 shows that 80% of ^{6}Li and ^{3}He and 66% of ^{3}H accumulate in the first 10cm of beryllium. It was expected that the majority of poison accumulation would happen closest to the core. Furthermore, the poisons in the innermost sectors of the beryllium reflector have the highest impact on the neutronic conditions of the reactor, since they directly reduce the reflector albedo. Because of this, a finer mesh was used for defining the radial sectors of beryllium for the innermost region.

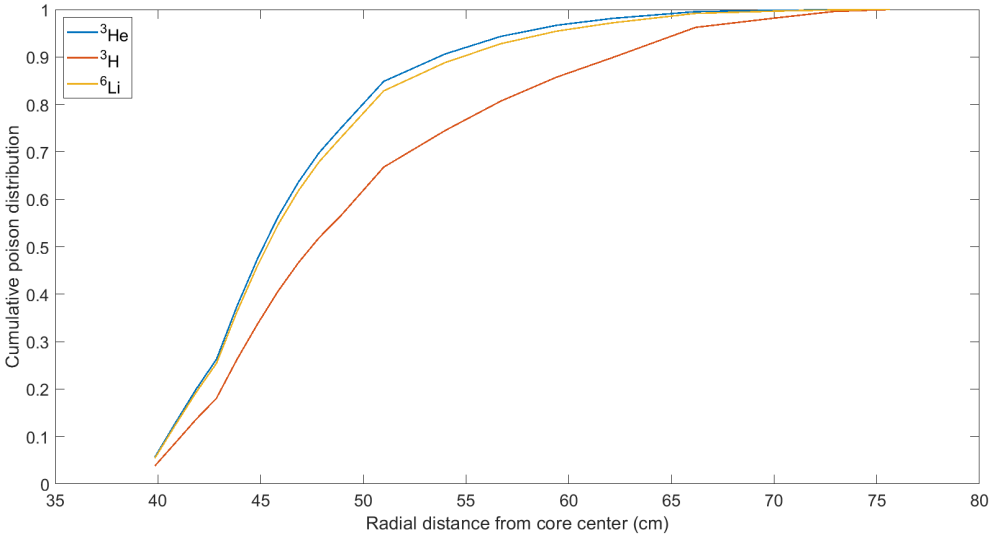

Figure 4: Cumulative poison distribution of the three studied isotopes in relation to the radial distance from JHR core center.

3.2. Estimating the effects of neutron poisoning

This section describes the effects of neutron poisoning on both the flux spectra measured in experimental positions in the reflector and also in core reactivity. For both analyses, the highest value of each neutron poison determined in the previous section was used in order to provide a conservative estimate of neutronic changes incurred. Figure 5 shows that ^{3}He build-up has the largest effect on neutron flux in the inspected experimental positions. The biggest changes measured were up to 5% decreases in flux. In the case of ^{6}Li, thermal fluxes in experimental sites were reduced by up to 2%.

Tritium had no statistically significant impact on flux, despite the large mass of poison being used as input. The graph shows that beryllium poisoning has no effect on the neutron flux spectrum of experimental positions in the core (such as position EC101). These results suggest that there is no need for replacing the beryllium blocks due to changes in neutron fluxes in experimental sites, but that changes in flux over this period of time should be considered as a potential source of uncertainty in experiments.

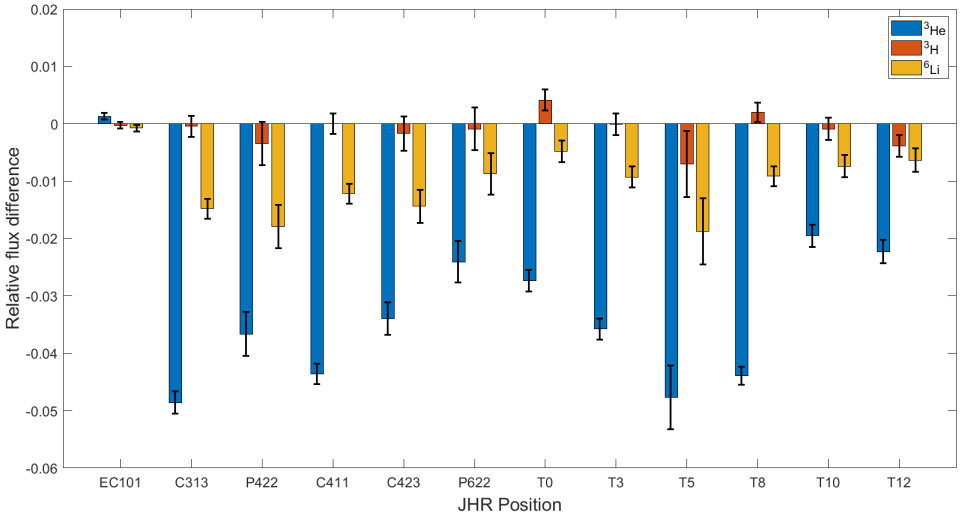

Figure 5: Relative impact of the ^{6}Li, ^{3}He, and ^{3}H poisoning in the JHR on the thermal flux distributions of experimental sites corresponding to Figure 1.

Criticality calculations before and after poisoning show that k-effective decreases in the presence of ^{6}Li and ^{3}He, whereas for ^{3}H the reactivity loss is not statistically significant. The results show that the beryllium reflector becomes less effective due to the build up of ^{6}Li and ^{3}He and that tritium has a negligible effect. These results are summarised in Table 3. The reactivity decrease caused by ^{6}Li and ^{3}He is very small, however. This suggests that if the JHR follows the anticipated on and off cycles, there would be no need to replace beryllium components within the first ten years.

Table 3: Changes in reactivity due to poisoning after 10 years of JHR operation due to different isotopes.

Isotope	Reactivity change (pcm)
^{6}Li	-21 ± 4.0
^{3}He	-58 ± 4.2
^{3}H	-1 ± 4.0

4. CONCLUSIONS

This paper employed two different methods to estimate the accumulation of poisons in the JHR over a ten-year period. While there were several assumptions and simplifications made, the results showed a good level of agreement for ^{6}Li and ^{3}He but not for tritium. However, when assessing the neutronic effects, it was found that tritium had negligible impact on the flux spectra measured at the experimental sites in the reflector and negligible effect on the reactor's reactivity and so the discrepancy was disregarded. ^{6}Li and ^{3}He did cause a measurable change in flux in experimental sites and also reactivity, with ^{3}He having the greatest impact. However, this change was relatively small. Therefore, there is likely no need to replace beryllium components within the first ten years of operation.

Nonetheless, it is essential to note that the duration of on and off cycles remains the biggest source of uncertainty in these estimates. Since the mass of ^{6}Li in the reflector plateaus relatively early, the reactivity loss it causes remains constant, whereas for ^{3}He the reactivity impact shows large fluctuations depending on the length of off-cycle periods. In the event of an unforeseen prolonged shutdown period, this could have significant consequences, potentially necessitating the replacement of beryllium components. The study also revealed that most poisons accumulate within the first 10cm of the reflector from the core's edge. Therefore, only this part of the beryllium would need replacing, rather than the entire reflector. Additional research could investigate the impacts of maximum reactor poisoning on the reactor startup conditions.

While this study focused on the effects of poisons on neutronics, the buildup of poisons can also result in irradiation-induced structural damage such as swelling, crack formation, and spalling. The accumulation of He-4 and tritium is particularly important in these processes, as gaseous species accumulating in beryllium pores can cause a decrease in fracture toughness. Therefore, it is crucial to consider induced swelling and embrittlement alongside neutronic effects when determining when to replace the reflector. For this further study, agreement between tritium accumulation estimates would be crucial.

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