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Activation calculations for multiple recycling of breeder ceramics by melt processing

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ABSTRACT

Multiple tritium breeder recycling is an attractive option to make a future fusion power reactor ecologically and economically sustainable. This research project aims at exploring the feasibility of multiple breeder ceramics recycling by remelting with regard to the activation behavior. The average activation level of multiple reused breeder pebbles is calculated for a DEMO reactor with HCPB (Helium Cooled Pebble Bed) blankets on the assumption that all spent pebbles are mixed and reprocessed by remelting. The calculation loop verifies the feasibility of a multiple reprocessing scheme with an acceptable waiting period of approximately 22 y for the remote handling dose rate level—even after 15 times recycling. This paper also describes the limitation of the current approach for the average activation property and provides some recommendations for future improvements.

1. Introduction

Multiple recycling of spent ceramic breeder is a challenge to minimize radioactive waste formed in a fusion power reactor. Also, reuse of lithium resource from the spent breeder is economically important because of the high cost of lithium-enriched materials. To allow a multiple reuse, a breeder material needs to have low activation property as well as good tritium breeding capability and high chemical stability. A modified melt process [1] has been developed for the fabrication of breeder pebbles of lithium orthosilicate, Li_4SiO_4, which is a promising candidate material meeting the demands mentioned above. In this process, breeder pebbles are fabricated by melting the raw materials at a high temperature in a noble metal crucible and then quenching melt droplets by a jet spray of liquid nitrogen. A melt-based process in principal enables a facile recycling without any additional process step such as wet-chemical treatment known as a costly and time-consuming step [2]. The reprocessing experiment showed the feasibility to reproduce well qualified pebbles by re-melting the fabricated pebble and by adding $LiOH{\cdot}H_2O$ to compensate the simulated Li-burn up [2]. More recently, two-phase pebbles, composed of Li_4SiO_4 and 10–30 mol% of Li_2TiO_3, have been proposed as an advanced breeder material in order to enhance the mechanical properties [3].

A former activation assessment [4] investigated the activation property of Li_4SiO_4 pebbles after a single-use of 1 full power year (fpy), where the material was considered to be subjected to the highest radiation level at a thin front layer in the central outboard blanket module. It is reported that the waiting period for the remote handling level (10 mSv/h) is approximately 50 y, which is dominantly determined by the assumed Co impurity even if its concentration is at the detection limit of inductively coupled plasma optical emission spectrometry (ICP-OES) (i.e. 1 mg/kg). It also estimated that a waiting period of approximately 400 y is required for the hands-on level (10 μSv/h) due to a Pt contamination released from the noble metal crucible alloy during the melt fabrication process. These waiting times are able to apply for the highest activated breeder pebbles in the reactor. For the assessment of the multiple reprocessing scheme, it is must be consider, however, that all spent pebbles are mixed and reprocessed for the next use in the reactor. To this end, it is required to calculate the average activity inventories of breeder pebble in the reactor.

To realize the multiple breeder recycling scheme with acceptable waiting periods, it is important not only to minimize impurity concentrations in the initial batch driving from the raw materials but also to investigate compositional change over multiple reuse. Recent experiments simulating a five-time recycling indicated that the impurity concentrations of the noble metals (i.e. Pt, Rh, and Au) gradually increase over the recycling steps by the release from the crucible alloy [5]. These impurity contaminations with cycle have risks of prolonging the waiting periods. Besides, isotopes formed by

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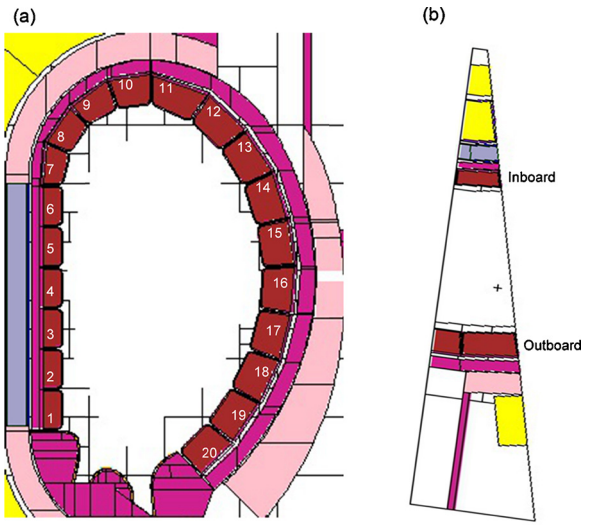

Fig. 1. Vertical cut (a) and horizontal cut (b) of the HCPB DEMO model, which contains 20 homogenized blanket modules (10 inboard and 10 outboard blankets).

neutron induced reaction could be critical, e.g. the accumulation of long life nuclides may result in a negative impact on the waiting periods. In spite of these risks, an activation property of a multiply reused pebble has been little investigated up to now.

The objectives of this work are to assess the average activation behavior of multiple reused breeder ceramics material in a DEMO reactor based on the current experimental data of the melt process and to investigate the effect of impurity accumulations during the multiple recycling on the waiting period. The following calculations steps are carried out by employing the Monte Carlo n-particle (MCNP) and the FISPACT inventory code; neutron transport calculation to obtain the neutron flux distribution (space and energy), activation calculations to obtain the nuclide and activity inventories for single use and for multiple reuses.

2. Methodology

2.1. Neutronics analysis

Neutron flux spectra were calculated in the standard VITAMIN-J 175 group structure by employing the MCNP5-1.60 code [6] and the nuclear data from FENDL-2.1 library [7]. The recently developed helium cooled pebble bed (HCPB) DEMO reactor model (CAD 11.25° torus sector model) [8] with an assumed fusion power of 2119 MW was employed (Fig. 1). The major and minor radii are 9.0 and 2.25 m, respectively. The model contains 10 inboard and 10 outboard blanket modules filled with the homogenized mixture of Eurofer (23.0 vol%), Be (21.0 vol%), breeder (10.2 vol%), and void (45.8 vol%). The homogenized blanket modules are indicated as BM and numbered from 1 to 20 as seen in Fig. 1a (e.g. the blanket module no. 5 is indicated as BM5). In accordance with the symmetry of the reactor, the 11.25° model includes one full inboard and 3/2 sized outboard blankets in toroidal direction as seen in Fig. 1b. Li_4SiO_4 is used as breeder ceramics with a ^6Li enrichment of 60 at% and no impurity. The resulting tritium breeding ratio (TBR) is at 1.04 for the assumed HCPB blanket design [8].

In the first step, the neutron fluxes averaged over each homogenized blanket module were calculated. This accordingly provided 20 neutron flux values and the associated spectra. Next, a weighted average neutron flux was calculated for the 20 blanket modules by using the volume of breeder pebbles in each blanket as a weighting function. In addition, neutron flux densities were also calculated along radial direction by subdividing a blanket into 5 small subsegments (see Section 3.2 for details).

Table 1
The initial compositions (wt%) of 100LOS and 70LOS used for the activation calculations.

Element	Initial composition (wt%)	
	100LOS	70LOS
Li	21.71667	18.89619
O	54.29365	51.40784
Si	23.83618	17.08041
Ti	0.00455	12.46679
Al	0.00156	0.00177
Au	0.00180	0.00180
Ba	0.00400	0.00400
C	0.11510	0.10795
Ca	0.00803	0.01020
Co	0.000012	0.000012
Cr	0.00047	0.00049
Cu	0.00061	0.00059
Fe	0.00560	0.00446
K	0.00067	0.00521
Na	0.00380	0.00380
Ni	0.00024	0.00026
Mg	0.00088	0.00126
Mn	0.00043	0.00039
Pt	0.00320	0.00320
Rh	0.00200	0.00200
Sr	0.00015	0.00017
Zn	0.00019	0.00020
Zr	0.00019	0.00099

2.2. Activation calculation

2.2.1. Conditions

Activation behavior of breeder material was investigated by using the FISPACT inventory code [9] and the EAF-2007 activation data [10] by involving neutron fluxes obtained by the MCNP calculations as described previously. The assumption is that 1 kg of 60 at% ^6Li enriched ceramic breeder pebbles with the sintering density of 95% T.D. are packed in a blanket with a packing factor of 64% and are used in the reactor for 3 fpy. Two kinds of breeder materials were used for the initial compositions, namely Li_4SiO_4 (100LOS) and Li_4SiO_4 with 30 mol% Li_2TiO_3 (70LOS). Given that the theoretical densities of Li_4SiO_4 ($P2_1/m$) and Li_2TiO_3 ($2\,C/c$) are 2.42 g/cm^3 [11] and 3.38 g/cm^3 [12], the effective densities of 100LOS and 70LOS are 1.44 and 1.66 g/cm^3, respectively. The determined impurity concentrations listed in Table 1 were used for the activation calculations as the initial compositions. The impurity concentrations in the raw materials ($LiOH{\cdot}H_2O$, SiO_2, and TiO_2) were investigated by ICP-OES. Only the Co concentration (0.12 ± 0.05 ppm) was determined by inductively coupled plasma mass spectrometry (ICP-MS) because it was previously specified as the most critical impurity dominating t_R even with 1 ppm [4].

2.2.2. Calculation loop

The calculation loop shown in Fig. 2 was constructed under a FORTRAN 90 environment based on a recycling scheme, namely, fabrication/reprocessing by the melt process, use in the reactor for a continuous irradiation over 3 fpy, and storage until the contact dose rate (CDR) achieves the remote level. This calculation loop was sequentially continued for 15 calculation cycles. The weighted average neutron flux is used for all of the FISPACT calculations during the loop. This is based on the assumption that the compositional change of breeder material during the calculation step has a negligible effect on the neutron flux. To ensure its validity, the averaged neutron fluxes over all blanket modulus were recalculated again with the MCNP code by involving major impurities contained in the 15th batch and were then compared with the initial ones.

2.2.2.1. FISPACT calculations. The first FISPACT (FISPACT1 in Fig. 2) step calculates the change of CDR up to 10^7 y after shutdown. From