Monte Carlo studies on the burnup measurement for the high temperature gas cooling reactor *

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Abstract: Online fuel pebble burnup measurement in a future high temperature gas cooling reactor is proposed for implementation through a high purity germanium (HPGe) gamma spectrometer. By using KORIGEN software and MCNP Monte Carlo simulations, the single pebble gamma radiations to be recorded in the detector are simulated under different irradiation histories. A specially developed algorithm is applied to analyze the generated spectra to reconstruct the gamma activity of the ^{137}Cs monitoring nuclide. It is demonstrated that by taking into account the intense interfering peaks, the ^{137}Cs activity in the spent pebbles can be derived with a standard deviation of 3.0% (1σ). The results support the feasibility of utilizing the HPGe spectrometry in the online determination of the pebble burnup in future modular pebble bed reactors.

Key words: high temperature gas cooling reactor, burnup, γ activity, Monte Carlo

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1 Introduction

The high temperature gas cooling reactor (HTGR) is drawing great attention from scientists because of its intrinsic safety [1–3]. In the type of the modular pebble bed reactors (MPBR), the fuel ball, consisting of a core of uranium compound fuel mixed with phenolic resin and a coat of graphite, undergoes a multi circulation during the operation of the reactor. The burnup of each pebble is online assessed non-destructively via the activities of the monitoring nuclide ^{137}Cs before the central control system makes a circulation/discharge judgment of the pebble [4–7]. The precision of the online determination of ^{137}Cs activity is thus of importance.

The circulation of fuel pebbles of MBPR is frequent [8]. The typical cooling time and measuring time for each pebble are about 50 hours and 20 seconds, respectively. As a consequence, the radiation background from short-lived nuclides is very high, so a high purity germanium (HPGe) detector with excellent energy resolution is preferred to see the ^{137}Cs peak clearly [9–11]. By conditioning the HPGe detector with proper running parameters like the shaping time and the flat-top constant, it has been shown that less than 3% deviation is achievable by a single source test [12, 13]. However, with the presence of all other background radiations, it is essential to check whether the activity of ^{137}Cs monitoring nuclide can be accurately derived through the gamma spectrometry. Since there is no burnt fuel pebble available for experimental investigation yet, Monte Carlo simulation is the method that we are mainly to rely on to make the assessment. In this paper, we first generate the yield of all the fission products from KORIGEN software [14, 15] for all burnup histories of a single pebble. Then a Monte Carlo procedure (MCNP) [16, 17] is applied to simulate the gamma spectra to be recorded by the HPGe detector with inclusion of all the absorption materials. Finally, a specially developed algorithm is adopted to analyze all the generated gamma spectra and to calculate the activities of ^{137}Cs nuclide. By comparing with the KORIGEN output, the feasibility of utilizing the HPGe gamma spectrometry in the determination of the pebble burnup is demonstrated. The paper is arranged as follows: Section 2 presents the simulation packages and the data flow, the simulation results are presented in Section 3. Section 4 is a summary.

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2 Simulation packages

The activity of a fuel pebble in the reactor depends on the burning history which includes the burnup and the neutron flux. Since there is no existing pebble reactor running so far, KORIGEN software is applied to obtain the radioactivity of the fuel pebble with different burnup under different neutron fluxes. In the calculation, three situations are considered. (1) The pebble is flowing down in the center of the reactor core, corresponding to the maximum neutron flux (dubbed N1); (2) The pebble is flowing down along the inner wall of the reactor core, corresponding to the minimum neutron flux (dubbed N2) and (3) The pebble is flowing down between the wall and the center and the average neutron spectral parameter is used, corresponding to the average neutron flux (dubbed N3). Fig. 1 presents the activity of various isotopes in a single pebble as a function of burnup for different neutron fluxes. It is shown that the correlation between the single pebble activity and the burnup is evident. The activities of the selected isotopes exhibit a monotonous increasing trend, although the total radiation of the pebble decreases with the burnup being larger than about 20000 MWd/tU. The activity of ^{137}Cs as a long-lived isotope is proportional to the amount of the burnt fuel and hence exhibits an increasing trend. On the other hand, the decreasing trend of the total activity is due to the competition of the γ emission between the short-lived fission products and the long-lived nuclides [11]. Interestingly, except for the ^{137}Cs isotope, the single pebble activities depend also on the neutron flux. Therefore, even though the slope of the curve for ^{137}Cs is smaller, which means less sensitivity of the activity on the burnup, it is proposed as the burnup monitoring nuclide for its resistance to the neutron flux.

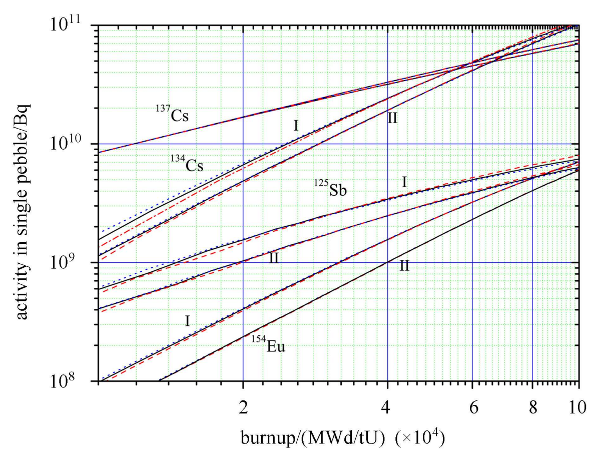

Fig. 1. (color online) Activities of various isotopes in a single pebble as a function of burnup for pebbles with low (I) and high (II) enrichment of ^{235}U under different neutron flux N1 (dash-dotted), N2 (dashed) and N3 (solid).

As long as the yield of all the fission products are predicted by KORIGEN at given irradiations, the geometry acceptance, material absorption of the burnup measuring system and the response of the detector are simulated with MCNP package based on a full size prototype of the whole system. The fuel pebble consists of a spherical core with 50 mm in diameter and a coating of graphite 5 mm in thickness (the total diameter is then 60 mm). The core is the mixture of uranium compound, graphite (about 80% in mass) and phenolic resin (C_7H_6O, about 20% in mass). It is assumed in the simulation that the radiation of ^{137}Cs originates homogeneously. The pebble is then accommodated in a steel container with the same dimension of the real one designed for the reactor. The HPGe spectrometer, being about 480 cm to the source, is shielded by a cylindrical lead chamber with 6 cm in thickness. A tungsten collimator of 540 mm length is installed between the spectrometer and the fuel pebble, with the throat being approximately 40 cm to the detector center. The throat diameter of the collimator is 3.2 mm. The geometrical parameters of the HPGe crystal and the aluminum shielding are set according to the instrument manual provided by the manufactory.

Figure 2 presents exemplarily one gamma spectrum expected in the HPGe detector for an undepleted pebble after 50 hours cooling time. The energy resolution (FWHM in keV) is incorporated in the simulation with the formula FWHM$=0.55+\sqrt{E+0.5E^2}$ where E is the gamma energy in MeV, as experimentally measured [12]. It is shown that due to the short cooling time (about 50 hours), all the isotopes with short life-time contribute to the whole spectrum. Particularly in the vicinity of

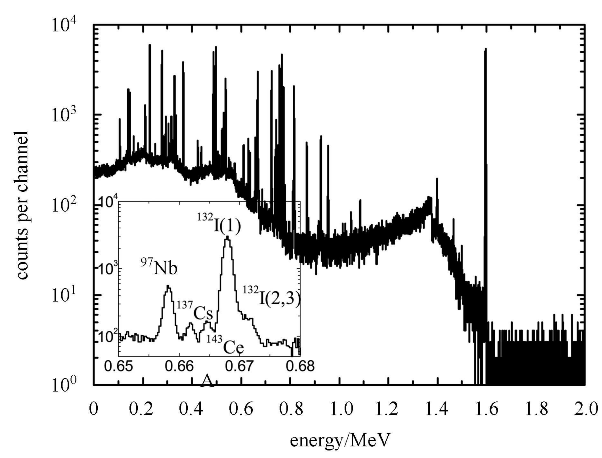

Fig. 2. A gamma spectrum expected in the HPGe detector for a single pebble with deep burnup under the averaged neutron irradiation flux. The insert shows the energy peaks in the vicinity of 662 keV of ^{137}Cs nuclide. The initial intensities are generated with KORIGEN and the material absorption, the geometrical acceptance and the detector response are simulated with MCNP.

662 keV of ^{137}Cs , the energy peaks of ^{97}Nb and ^{143}Ce are presented and exhibit higher intensity. This is totally different from the case of an experimental reactor, for which the cooling time is several months and the energy spectrum is very clean, and hence calls for careful treatment in the spectrum analysis. Because of this, in order to discriminate the ^{137}Cs out from the interfering peaks with the least uncertainty, the HPGe detector is needed with the running parameters well optimized. In addition, a sophisticated gamma spectrum analysis algorithm is called for.

3 Gamma spectra analysis and results

An algorithm for analyzing the gamma spectra is developed and tested experimentally with three sources [12, 13]. Here it will be applied in the gamma spectra simulated with KORIGEN and MCNP, in which all the fission products are taken into account, as shown in Fig. 2. After the spectrometer is calibrated, the program first searches the peak in the vicinity of 662 keV of ^{137}Cs nuclide. If the peak is found and confirmed by the prescribed criteria including a signal-to-noise ratio and the peak shape matching, the valley corresponding to the background level is searched within $\pm 5\sigma$ range on both sides of the peak. The width of the range is an adjustable parameter. Near the valleys on both sides five channels (adjustable parameter again) are used to evaluate the background. If the peak of 662 keV is not found, on the contrary, the activity of ^{137}Cs is set to be zero. The treatment is trivial because this situation indicates a pebble with a very low burnup which will be refilled to the reactor core. If other peaks are recognized in the given range, on the other hand, these peaks will be included and the searching for background level is extended to the outmost peak in the range. After the background is subtracted, single or multi Gaussian fit is conducted to derive the area of the corresponding peaks in the range. Once optimized, all the analysis parameters are fixed in the analysis to minimize the statistical fluctuation [12]. It has been demonstrated in a single source test [12] that the uncertainty of the ^{137}Cs activity can be determined within 2.8% uncertainty (1σ) in the case which corresponds to a spent fuel pebble. Here we investigate the correlation between the burnup and the activity of ^{137}Cs for a pebble with all radiation backgrounds under different irradiation histories.

Figure 3(a) presents the reconstructed ^{137}Cs activity $A_{\rm rec}$ as a function of burnup for the maximum neutron flux N1 (see Section 2). The error bars denote the statistical uncertainty of the activity determination following the calculation in [12]. Two sets of the measuring time, 10 and 25 seconds, are considered. If the reconstructed ^{137}Cs activity is below a threshold value in the first 10 seconds' measurement, the pebble is recharged to the reactor as an undepleted pebble. Otherwise the measurement is extended to 25 seconds to derive more accurately the activity with higher statistics in order to make a recirculation/discharge judgment on a pebble-by-pebble basis. It is shown that in both cases, the activities of ^{137}Cs increase with the burnup in accordance with the

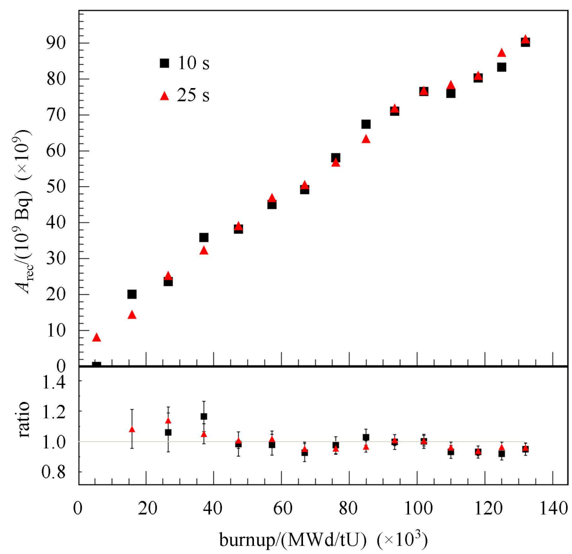

Fig. 3. (color online) (a) Activity of the burnup indicator ^{137}Cs reconstructed from the simulated gamma spectra $A_{\rm rec}$, (b) The ratio of the reconstructed activity and the initial output of KORIGEN as a function of the burnup in measuring time of 10 (solid squares) and 25 (solid triangles) seconds, respectively, at the maximum neutron flux N1.

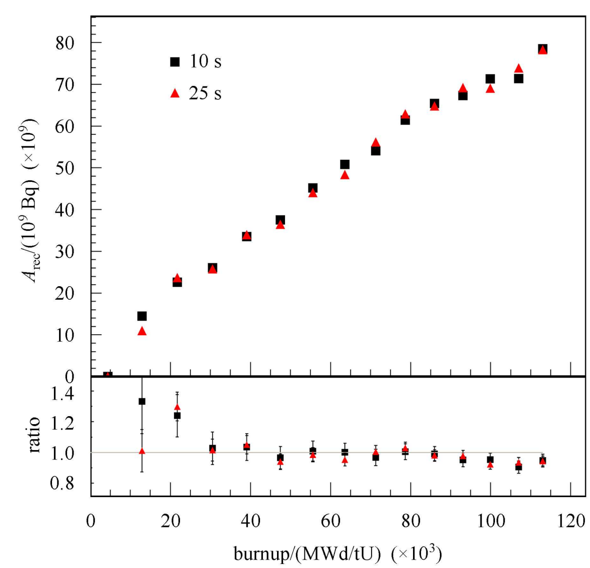

Fig. 4. (color online) The same as Fig. 3 for the minimum neutron flux N2.

initial tendency shown in Fig. 1. The ratio of the reconstructed ^{137}Cs activity and the initial value from KORIGEN output $A_{\rm ini}$ is presented in Fig. 3(b). It is clearly shown that for the lower burnup pebbles, because the ^{137}Cs counting rate is low while the total radiation background is intense, the ratio exhibits large uncertainty and deviates considerably from unity, indicating a significant error occurs in the reconstruction of the gamma activity. While in the deep burnup case, the net counting rate of ^{137}Cs is increased and the background is relatively lower, the ratio is near unity with a few percent deviation and demonstrates the reliability of the gamma activity reconstruction. Figs. 4 and 5 present the same observables for the minimum (N2) and averaged (N3) neutron flux , respectively. The conclusion holds for these two situations.

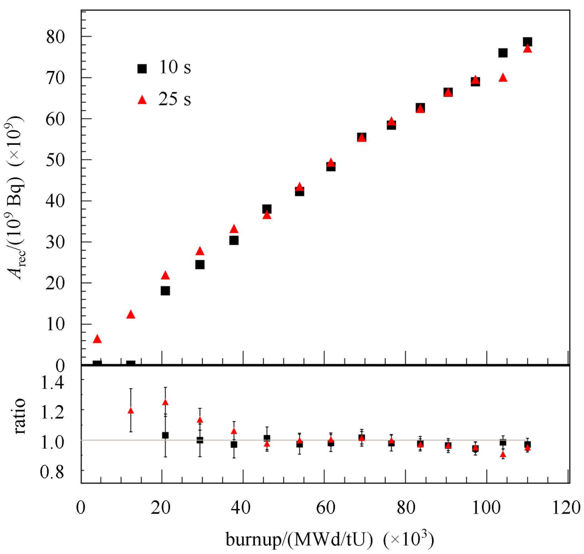

Fig. 5. (color online) The same as Figs. 3 and 4 for the averaged neutron flux N3.

In order to quantitatively evaluate the deviation of the analysis, in Fig. 6 we present the distribution of the ratio for the simulations corresponding to the high prescribed burnups between 40 and 100 GWD/tU for 10 and 25 seconds measurement respectively for all three neutron fluxes. This range is investigated because the pebble will be discharged if the burnup is approximately above 80 GWD/tU. The standard deviation of these analyses is a 3.0% (1σ)in agreement with the Gaussian fit which yields σ=2.8%. The reconstruction of the ^{137}Cs activity for the pebbles with lower burnup is not essential since they will be refilled to the reactor core as undepleted ones. It suggests that by taking into account the intense interfering background, the ^{137}Cs activity can still be determined reasonably well (within 3.0% (1σ)) for the nearly depleted pebbles to be recirculated or discharged in the real application of the future modular pebble bed reactors.

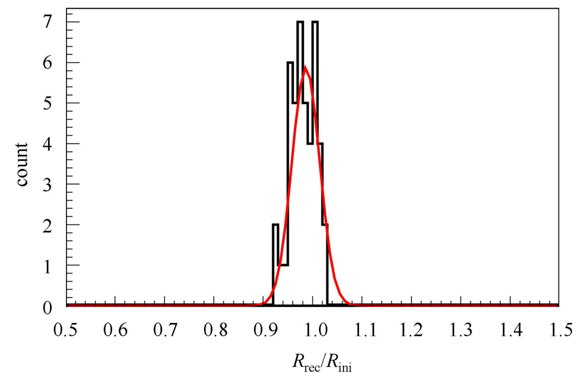

Fig. 6. (color online) The ratio of the reconstructed activity $A_{\rm rec}$ and the initial output of KORIGEN $A_{\rm ini}$ of ^{137}Cs for the pebbles with prescribed burnup between 40 and 100 GWD/tU in 25 seconds' measurement. The histogram is fit by a Gaussian function.

4 Summary

In summary, we have studied the burnup measurement via KORIGEN and MCNP Monte Carlo simulation. The initial radiations of all the fission products were calculated with KORIGEN software for different burnups at different neutron radiation fluxes. The geometrical acceptance, material absorption and the detector responses are simulated with MCNP. With a specially developed gamma analysis procedure, the activities of the monitoring nuclide ^{137}Cs are derived for all gamma spectra. It is shown that for the nearly depleted pebbles, the reconstruction of the gamma activity is achieved with a standard deviation of less than 3.0% (1σ) in comparison with the initial values. The results demonstrate the feasibility of utilizing an HPGe gamma spectrometer in the online determination of the pebble burnup in future modular pebble bed reactors.

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