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Development and validation of the code COUPLE3.0 for the coupled analysis of neutron transport and burnup in ADS

Lu Zhang[1,2] · Yong-Wei Yang[1] · Yuan-Guang Fu[3,4] · De-Liang Fan[1] · Yu-Cui Gao[1]

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Abstract The analysis of the fuel depletion behavior is critical for maintaining the safety of accelerator-driven subcritical systems (ADSs). The code COUPLE2.0 coupling 3-D neutron transport and point burnup calculation was developed by Tsinghua University. A Monte Carlo method is used for the neutron transport analysis, and the burnup calculation is based on a deterministic method. The code can be used for the analysis of targets coupled with a reactor in ADSs. In response to additional ADS analysis requirements at the Institute of Modern Physics at the Chinese Academy of Sciences, the COUPLE3.0 version was developed to include the new functions of (1) a module for the calculation of proton irradiation for the analysis of cumulative behavior using the residual radionuclide operating history, (2) a fixed-flux radiation module for hazard assessment and analysis of the burnable poison, and (3) a module for multi-kernel parallel calculation, which improves the radionuclide replacement for the burnup analysis to balance the precision level and computational efficiency of the program. This paper introduces the validation of the COUPLE3.0 code using a fast reactor benchmark and ADS benchmark calculations. Moreover, the proton irradiation module was verified by a comparison with the analytic method of calculating the ^{210}Po accumulation results. The results demonstrate that COUPLE3.0 is suitable for the analysis of neutron transport and the burnup of nuclides for ADSs.

Keywords COUPLE3.0 · Neutron transport · Burnup · Accelerator-driven subcritical system

1 Introduction

An accelerator-driven subcritical system (ADS) is considered suitable for the transmutation of minor actinides (MA), and it also achieves energy production with good safety characteristics due to the subcriticality [1, 2]. Therefore, numerous research studies on ADSs have been conducted worldwide. Fuel depletion analysis is an important part of the evaluation of a subcritical reactor core. Domestic and foreign research institutions have developed several different programs [3–5] for the analysis of ADSs. Verified by benchmark calculations, these programs can be used to analyze a subcritical reactor of ADSs.

During the continuous operation of ADSs, large amounts of spallation products are produced in the target zone by protons and neutrons. Most of them are toxic with high levels of radioactivity, and the level of the external exposure dose rate is one of the important issues of radiation protection during operation and maintenance. For the lead–bismuth–eutectic (LBE) target of the ADS, the ^{210}Po is of high concern [6]; it is produced by a (n, γ) reaction of ^{209}Bi together with the β-decay of ^{210}Bi. When tungsten is

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✉ Yong-Wei Yang
yangyongwei@impcas.ac.cn

[1] Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China

[2] University of Chinese Academy of Sciences, Beijing 100049, China

[3] Institute of Applied Physics and Computed Mathematics, Beijing 100088, China

[4] CAEP Software Center for High Performance Numerical Simulation, Beijing 100088, China

used as the spallation target material, we should be concerned about Hf because it has a large cross section for neutron absorption [7], which has serious impacts on the target–reactor coupling behavior. Therefore, it is important to consider the accumulation behavior of the spallation products in the target.

The code COUPLE2.0 [8–10], developed by the Institute of Nuclear and New Energy Technology (INET) at Tsinghua University, couples the neutron transport program, MCNPX, based on the Monte Carlo method with the nuclide depletion calculation code, ORIGEN2.1. It can be used for the analysis of a subcritical reactor in an ADS [11]. In response to additional ADS research requirements at the Institute of Modern Physics at the Chinese Academy of Sciences, the COUPLE3.0 version was developed in collaboration with the High Performance Computation Center at the China Academy of Engineering Physics.

For the COUPLE3.0 code, the following functions were added:

(a) A proton radiation calculation module was developed that can be used to analyze the accumulation behavior of residual nuclides in the spallation target based on the running history.
(b) A fixed-flux neutron irradiation module was added for the analysis of burnable poisons.
(c) According to the depletion of some important nuclides and considering the contribution of the reaction of some minor actinides (n, absorption) to the overall neutron absorption in the reactor, the user can set an importance cutoff value (ICV) to balance the precision level and the computation time. The transport process studies were achieved with a parallel message passing interface (MPI) to improve the calculation efficiency. The addition of the post-processing function of the nuclide information is convenient for the extraction of the nuclide information. COUPLE3.0 consists of two modules, the neutron irradiation module and the proton irradiation module. In the first module, a constant fission power is used for the burnup analysis of the fuel zone; a constant flux model is also used for the burnup analysis for the burnable poison zones that do not produce fission power. The second module allows for determining the effects of the bare proton target and the target–reactor coupled system on the accumulation behavior of the residual nuclides.

Using a thorium-based ADS benchmark, we verified the COUPLE3.0 code with regard to the burnup behavior in the ADS system that was induced by a fixed neutron source. A validation was conducted for the applicability of the code for the MA transmutation analysis using a fast reactor benchmark. In order to verify the accuracy of the proton irradiation module, the accumulation of ^{210}Po in the target was calculated using COUPLE3.0, and the results were compared with the estimated values from an analytical method; the results were in good agreement.

2 Neutron irradiation module

The neutron irradiation burnup calculation is one of the functions of the COUPLE3.0 code, which is based on the COUPLE2.0 code; in the new version, many aspects of the physical model were improved to enhance the accuracy of the expansion method for solving the equation of the burnup chains. Because of the existence of a spatial difference in the neutron flux distribution, the nuclear fuel depletion is different for each location in actual systems. Generally, an approximation can be obtained by dividing the fuel depletion area. The internal neutron flux in the fuel depletion area does not change significantly, and there is no significant difference in the nucleon density in this region; therefore, the region can be simulated as a point. In a relatively short period of time, the fuel depletion is not very obvious, the change in the neutron spectrum is not very significant, and the change in a single-group cross section is not obvious; therefore, the solution of the linear depletion equation can be obtained. In COUPLE3.0, MCNPX2.7.0 [12] is used for the transport calculation and the single-group section for the fuel burnup calculation based on the neutron flux and ORIGEN2.1 [13] is used for the burnup calculation to obtain the nuclide compositions at the end of the time step. The main part of COUPLE3.0 is responsible for delivering the necessary data for the neutron transport calculation by transferring the merged single-group cross sections from MCNPX to ORIGEN2.1, calculating the power level of the depletion areas, and transferring them to ORIGEN2.1; at the end of the time step of the burnup calculation, ORIGEN2.1 transfers the material compositions to MCNPX to perform the neutron transport calculation in the next step. The two programs run alternately until the fuel depletion time reaches the end. The process of the neutron irradiation calculation is shown in Fig. 1.

COUPLE3.0 can run in a parallel MPI environment. The data used for MCNPX come from the ENDF/B-VII.0 library, which is currently suitable for fuel depletion calculations. However, it should be pointed out that, because of the large dataset of ENDF/B-VII.0 [14], the calculations are computationally intensive. In COUPLE3.0, all nuclides are assumed to be able to change over time, which allows the program to deal with practical problems, such as fuel containing neutron absorption or the depletion of poisonous materials. In order to improve the calculation accuracy, a

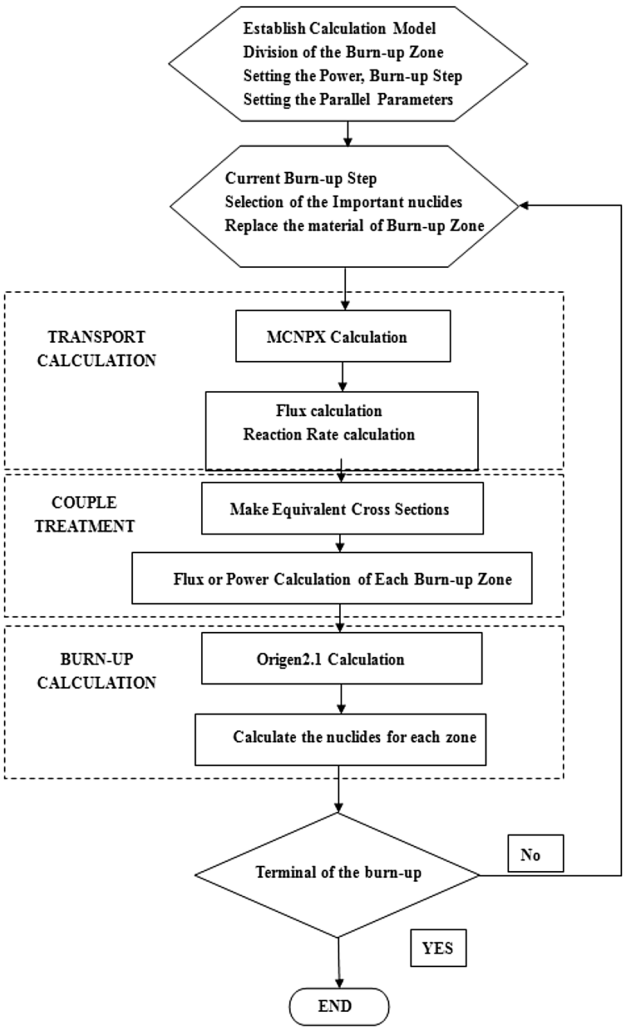

Fig. 1 Calculation process of neutron irradiation in COUPLE3.0

branching ratio is introduced for the excited states of the minor actinides.

3 Proton radiation calculation module

A proton irradiation function was added to COUPLE3.0 and can simulate the high-energy proton bombardment on the spallation target, the spallation reaction of the residual nuclides with depletion, and the accumulation of heavy metals. The spallation reaction generates neutrons and large amounts of residual nuclides, which accumulate and disappear by decay or other types of nuclear reactions such as absorption. The above process is complex, and for an accurate simulation, the cross section of the proton reaction and the residual nuclear yield of the different nuclides have to be known. Using these parameters, an equation of the proton irradiation burnup can be written similarly to the neutron irradiation equation; however, because some data are not available, it is impractical to develop an equation of the proton irradiation burnup. COUPLE3.0 coupled with MCNPX2.7.0 makes the simulation possible, not because of the proton reaction data of MCNPX2.7.0, but because COUPLE3.0 can calculate the residual nuclear yield after the different incident particles have bombarded the nuclides.

The meaning of the yield of the residual nuclides is the same as that of the recharge rate of the F_i in the burnup Eq. (1) used in ORIGEN2.1, and it can be used for the simulation of the burnup process of the proton irradiation.

$$\frac{\mathrm{d}X_i}{\mathrm{d}t} = \sum_{j=1}^{N} l_{ij}\lambda_j X_j + \phi \sum_{k=1}^{N} f_{ik}\sigma_k X_k - (\lambda_i + \phi\sigma_i + r_i)X_i + F_i \quad i = 1\ldots N, \tag{1}$$

where X_i is the nucleon density of the Ith nuclides; N is the upper limit of the type of nuclide; L_{ij} is the jth nuclide acting as the parent of the Ith nucleus, which produces a fraction of the Ith nuclide after decay; λ_j is the decay constant of the jth species; ϕ is the neutron flux averaged over the energy zone and the burnup area; f_{ik} is the share of the Ith nuclide resulting from the nuclear reaction of the kth nuclide; σ_k is the single-group reaction cross section of the kth nuclide averaged over the neutron energy spectrum; r_i is the removal ratio of the ith nuclide; F_i is the recharge rate of the ith nuclide.

When COUPLE3.0 is used in the calculation of proton irradiation, it is similar to the neutron irradiation calculation method and the concept of a transport step is also introduced.

In the transport step, the accumulation of the spallation product is not obvious and it has little effect on the neutron yield, the neutron energy spectrum, and the residual nuclear recharge rate. Thus, the burnup calculation is performed with a constant recharge rate. In the next step, it is considered that the accumulation of the spallation products is considerable and that the influence on the transport cannot be ignored. At the same time, the materials in the burnup zone are renewed and the recharge rate is calculated for the burnup calculation. MCNPX2.7.0 is used for the transport calculation to obtain the distribution of the spallation neutron flux, the single-group cross section, and the remaining nuclear recharge rate. ORIGEN2.1 is used for the burnup calculation in order to obtain the nuclide content of the burnup zones at the end of the transport step. The main portion of the code is responsible for transferring data between MCNPX2.7.0 and ORIGEN2.1 to achieve the coupling calculation of transport and burnup. It calculates the neutron flux and the recharge rate at a given proton irradiation level that are transferred to ORIGEN2.1 for the burnup calculation. Subsequently, COUPLE3.0 extracts the

calculation results of ORIGEN2.1 at the end of the time step, selects the important nuclides, and updates the fuel compositions of the materials in the burnup zones for the transport calculation in the next step. It alternately and repeatedly calls the MCNPX2.7.0 and ORIGEN2.1 codes until the end of the proton irradiation cycle is reached. The flowchart for the proton irradiation calculation in COUPLE3.0 is shown in Fig. 2.

The proton irradiation burnup calculation in COUPLE3.0 is divided into two modes. The first one considers the transmutation of the residual nuclides and the second one only considers the decay of the residual nuclides. The first method is closer to the actual physical process. Because the neutron flux in the spallation neutron target area is high, the transmutation reactions of the residual nuclides should not be neglected; in addition, the neutrons from the subcritical reactor core result in the transmutation of the residual nuclides.

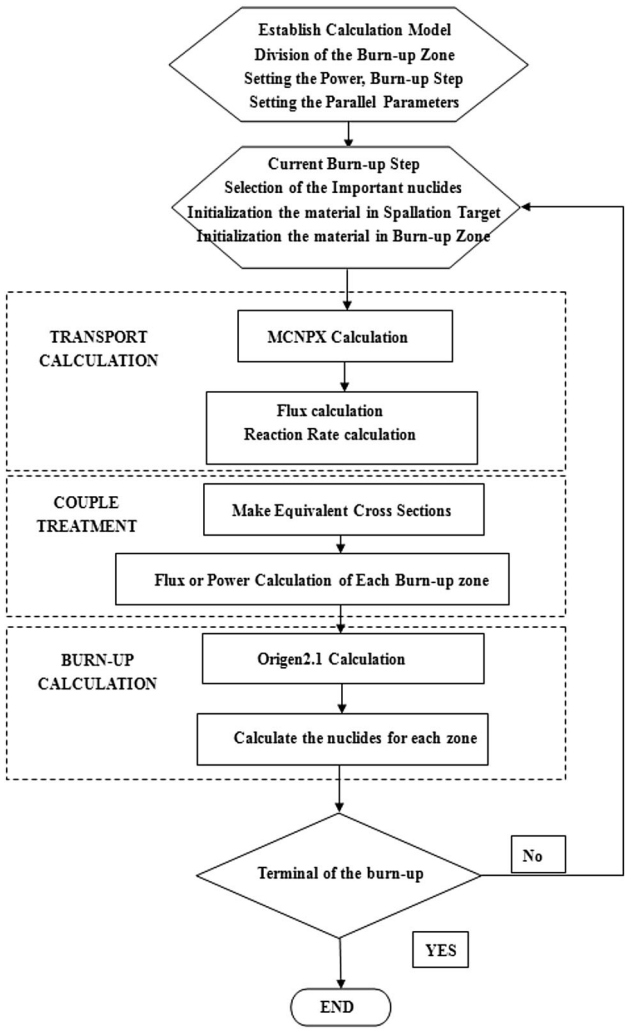

Fig. 2 Proton irradiation calculation in COUPLE3.0

It is necessary to point out that, due to the lack of a cross section in high-energy neutron data, the reaction rates are underestimated. For the second kind of treatment, because of the neglect of the disappearance of the residual nuclides due to the nuclear reaction induced by the neutrons, the accumulation of the residual nuclides is overestimated. In view of the safety analysis, the effect of the specific nuclides is often overestimated. Therefore, this method is conservative.

For a liquid or a flowing target, the residual nucleus will spread to other areas due to the flow and the actual cumulative amount of the residual nuclei in the target area is less than the amount in the stationary target. To solve this problem, COUPLE3.0 uses a rough approximation and considers that the migration rate of the residual nucleus is due to the flow of the fluid and that each spallation zone corresponds to a migration rate; this is represented by a coefficient between 0 and 1.

At first, COUPLE3.0 calculates the recharge rate of the remaining nuclides without flow by calling MCNPX2.7.0; subsequently, the recharge rate is multiplied by the rate of fuel consumption and then transferred to ORIGEN2.1 for the burnup calculation. The migration rate needs to be manually set by the user. When the migration rate is 1, all the nuclides are lost. When the migration rate is 0, there is no loss of nuclides.

4 Preliminary verification of COUPLE3.0

4.1 Fast reactor benchmark verification

In order to verify the applicability and reliability of COUPLE3.0 when used in the calculation of a reactor with a fast neutron spectrum, we performed the calculation and analysis of the benchmark of the fast spectrum plutonium-burning reactor proposed by the Organization for Economic Cooperation and Development Nuclear Energy Agency (OECD/NEA). Figure 3 shows the geometric model of the benchmark problem. In the calculation, the overall burnup time is set as 625 effective full power days (EFPD) and the thermal power is set as 1200 MW. Table 1 provides the results from different research institutions and programs [15]. It can be seen that, in terms of the change in the amount of actinides or the change in reactivity between the beginning of the circle (BOC) and ending of the circle (EOC), the calculation results using COUPLE3.0 are in agreement with the results of all the research institutions. This indicates that the program is reliable for the burnup analysis of a fast neutron spectral device.

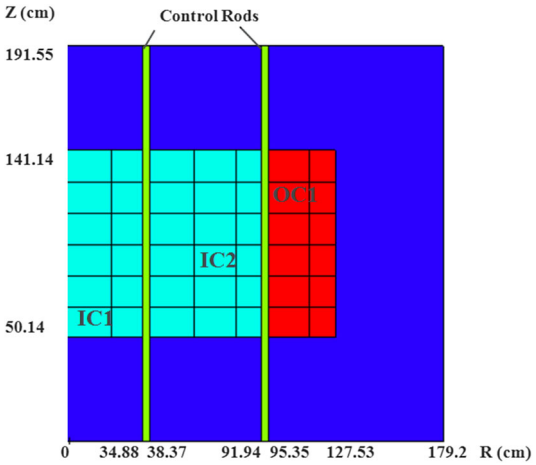

Fig. 3 (Color online) Schematic diagram of the OECD/NEA fast spectrum incinerator

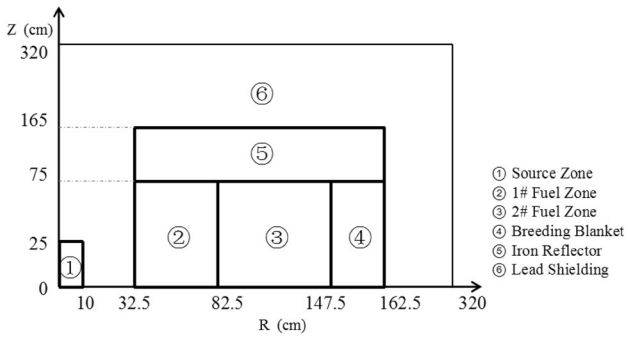

Fig. 4 IAEA-ADS burnup benchmark model

4.2 ADS burnup benchmark test

In order to verify the use of COUPLE3.0 for ADS research, we analyzed the ADS benchmark problem of the International Atomic Energy Agency (IAEA) [16]. The benchmark relates to an ADS device, including a cylindrical subcritical ^{233}U-fueled reactor with ^{232}Th as a fertile material and natural lead as a reflector. The neutron source is placed in the center of the subcritical reactor to simulate the spallation neutron source. The benchmark problem is to test the burnup characteristics of the ADS under different subcritical conditions. In this study, we focus on the behavior changes of k_{eff} and the intensity of the beam efficiency with an initial k_{eff} value of 0.94. The results are compared with those of other institutions [17, 18]. Figure 4 shows the geometry of the benchmark problem. The core is divided into three zones: the first and second zones are for fuel with different enrichments of ^{233}U and the third zone is the fertile zone with ^{232}Th. The initial k_{eff} value of 0.94 is obtained by adjusting the enrichment of ^{233}U.

Figure 5a shows the changes in the k_{eff} value during burnup. The comparison shows that the change in k_{eff} calculated by COUPLE3.0 is close to the results of the Japanese institution. Figure 5b shows the changes in the beam intensity during the burnup in order to maintain the operating power at the level of 1500 MW_{th}. The results show that the required beam intensity is largest for COUPLE3.0, and this is determined by the characteristics of the changes in k_{eff}. A preliminary analysis indicates that this difference may be attributed to the use of the ENDF/B-VII.0 library in our calculations; the other institutions used the ENDF/B-VI library. Further studies should be conducted to determine the selection of a cross section library.

In COUPLE3.0, the selection of the important nuclides is one approach for increasing the computational

Table 1 Reactivity changes and nuclide changes of the baseline problem of a fast plutonium incinerator

Institute or code	ANL (USA)	CEA (France)	PNC (Jeff2)	PNC (Jeff3.2)	PSI (Swiss)	MOCUP	COUPLE3.0
Reactivity changes between the BOC and EOC [$\%\Delta k/(k_1k_2)$]							
	12.85	13.27	13.60	13.39	13.06	12.44	13.11
Nuclides changes between the BOC and EOC [Δkg]							
^{235}U	− 5.6	− 5.9	− 5.8	− 5.7	− 5.5	− 5.61	− 5.78
^{238}U	− 420	− 411	− 392	− 390	− 384	− 394.5	− 404.9
^{238}Pu	− 50	− 45	− 50	− 47	− 43.4	− 49.0	− 49.4
^{239}Pu	− 149	− 174	− 170	− 131	− 173	− 160.9	− 165.5
^{240}Pu	− 38	− 21	− 32	− 13	− 34	− 34.7	− 29.4
^{241}Pu	− 133	− 139	− 133	− 122	− 137	− 136.5	− 136.5
^{242}Pu	− 29	− 42	− 31	− 20	− 25	− 30.5	− 29.5
^{241}Am	9.1	7.5	8.8	9.5	8.6	9.58	8.43
^{243}Am	31	44	33	34	33	31.2	31.7
^{242}Cm	3.7	5.2	4.8	4.5	4.1	4.63	4.54
^{244}Cm	4.1	7.4	5.3	5.3	5.4	4.48	4.95

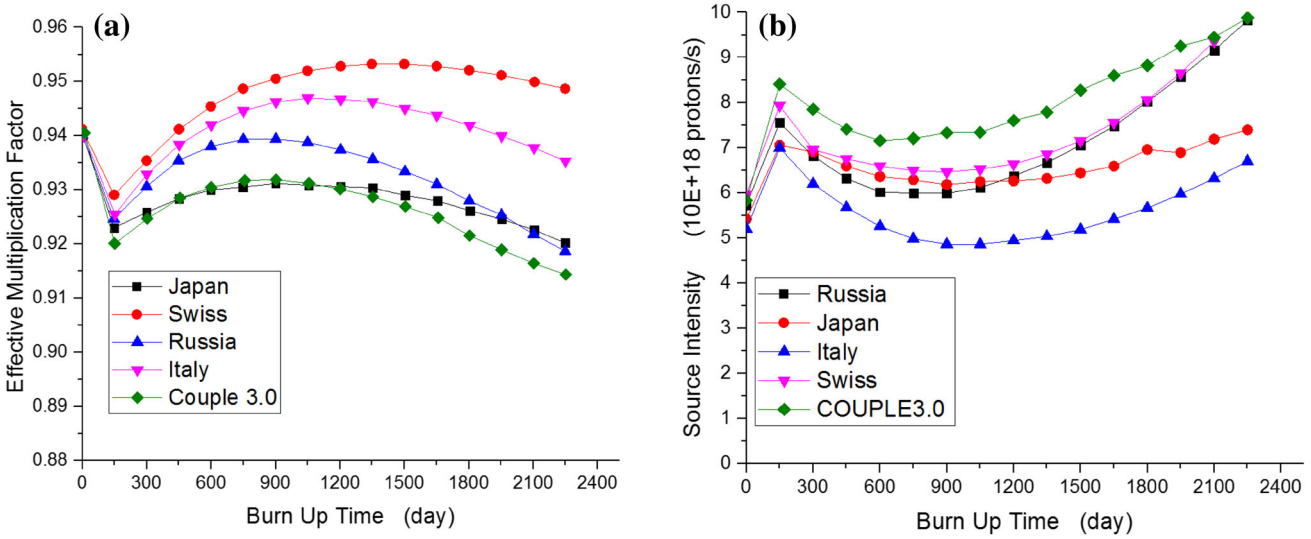

Fig. 5 (Color online) Changes of effective multiplication factors (**a**) and the beam intensity (**b**) during burnup

efficiency. Based on the IAEA-benchmark results, we performed several calculations to compare the computation times for different simulation results. These calculations were carried out using 64 CPUs with 1×10^6 source particles for each burnup step. In order to balance the precision level and the computation time, we set importance cutoff values (ICV) which represents the impacts on neutron absorption for all the nuclides. For each nuclide, its influence on the neutron transport is not considered in the MCNPX when its contribution fraction to the total value of neutron absorption in the specific zone is less than the ICV. This reduces the computation time and results in an acceptable precision level.

Table 2 shows that, when the ICV increases, the number of nuclides for the selected burnup zone decreases and the computation time decreases too. However, when the ICV is less than 1×10^{-8}, there is no significant difference in the effective multiplication factors during the burnup (Fig. 6). When we set the cutoff value as 1×10^{-8}, we can reduce the computation time by almost 40% when compared to an ICV of 1×10^{-13}.

Table 2 The influence of different ICV (importance cutoff values) on the computation time

ICV	Number of nuclides in selected burnup zone	Computation time (min)
1×10^{-13}	309	3701.7
1×10^{-8}	185	2269.8
1×10^{-7}	152	1900.4
1×10^{-5}	69	930.3
1×10^{-4}	22	164

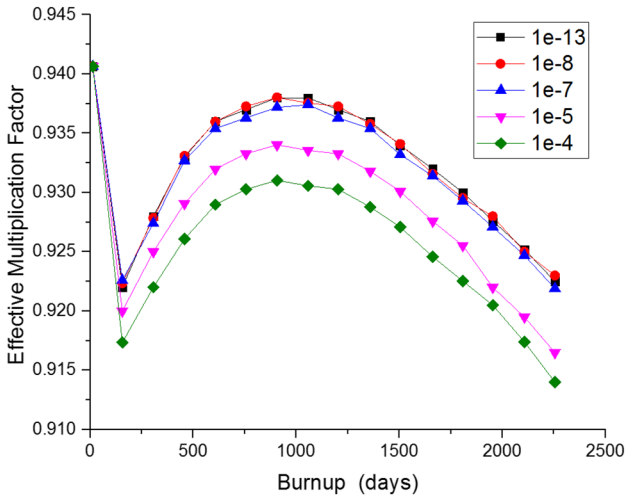

Fig. 6 (Color online) Calculation results with different ICV (importance cutoff values)

4.3 Preliminary verification of proton irradiation

In order to verify the reliability of the newly added proton irradiation function in COUPLE3.0, we chose the target nuclide ^{210}Po as an example due to the lack of benchmark examples for the accumulation of radionuclides in the target area. Our analysis is based on the study of lead bismuth with a window target [19], which is part of the project "Future advanced fission energy—ADS transmutation system" launched by the Chinese Academy of Sciences.

The relevant parameters are as follows: the proton beam energy is 250 MeV and the proton beam intensity is 0.725 mA. Two methods were used for the analysis: method 1 is an analysis of the target nuclide accumulation

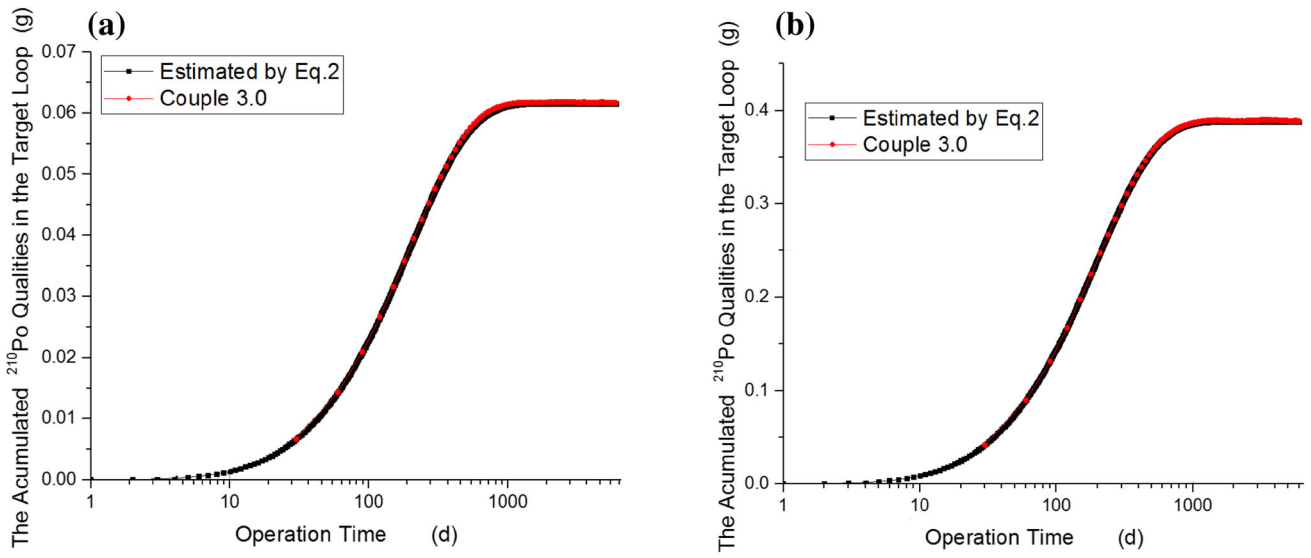

Fig. 7 (Color online) Accumulation of ^{210}Po for the LBE spallation target operated independently (**a**) and coupled with reactor (**b**)

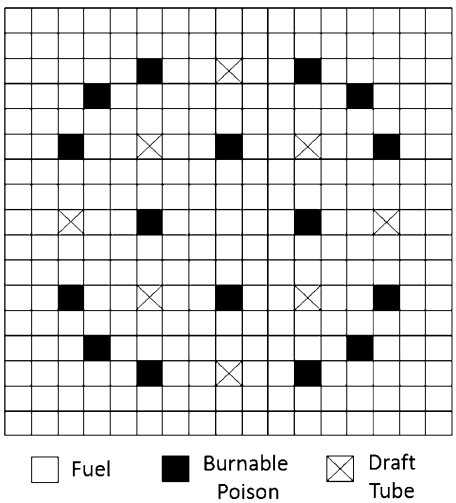

Fig. 8 Calculation model of the burnable poison

behavior using the newly developed COUPLE3.0; method 2 is an analytical approach for the analysis of the ^{210}Po accumulation characteristics. The analysis and comparison are carried out for the bare target and the coupled condition of the target and the reactor, respectively; the k_{eff} for the coupled condition is about 0.70. The analytical method is used to calculate the production of ^{210}Bi and ^{210}Po, which can be obtained by substituting MCNPX to study the evolution of the nuclides in the target.

$$\begin{cases} N_{\mathrm{Bi}}(t) = \dfrac{I_{\mathrm{p}} Y_{\mathrm{Bi}}}{\lambda_{\mathrm{Bi}}}\left(1 - \mathrm{e}^{-\lambda_{\mathrm{Bi}} \cdot t}\right) \\ N_{\mathrm{Po}}(t) = \dfrac{I_{\mathrm{p}} Y_{\mathrm{Bi}} + I_{\mathrm{p}} Y_{\mathrm{Po}}}{\lambda_{\mathrm{Po}}}\left(1 - \mathrm{e}^{-\lambda_{\mathrm{Po}} \cdot t}\right) + \dfrac{I_{\mathrm{p}} Y_{\mathrm{Bi}}}{\lambda_{\mathrm{Po}} - \lambda_{\mathrm{Bi}}}\left(\mathrm{e}^{-\lambda_{\mathrm{Po}} \cdot t} - \mathrm{e}^{-\lambda_{\mathrm{Bi}} \cdot t}\right) \end{cases} \quad (2)$$

where I_{p} is the proton beam intensity, the unit is mA; Y_{Po} and Y_{Bi} are the yields of ^{210}Po and ^{210}Bi, respectively, normalized to per mA; λ_{Po} and λ_{Bi} are the decay constants of ^{210}Po and 210Bi, respectively, normalized to each day.

Figure 7a shows the accumulation of the ^{210}Po nuclides in the target area for the bare target operation, and Fig. 7b shows the accumulation of the target ^{210}Po for the target–reactor coupled condition. It can be seen that the results of the COUPLE3.0 simulation are in good agreement with the results obtained by the analytical method. This is because the cumulative production of the ^{210}Po nuclides is relatively low and the neutron nuclear reaction cross section is low for the analysis situation; this means that the changes in the nuclides induced by the neutron can be neglected. In addition, the burnup of the core is not considered for the verification calculation. Therefore, the core flux is constant.

The preliminary verification calculation shows that COUPLE3.0 is suitable for the simulation of proton irradiation. If the appropriate physical models of the nuclear reaction for medium- and high-energy protons and neutrons are chosen, we can accurately analyze the accumulation of the spallation products. At present, we use the CEM03 model [20] in MCNPX, and the calculated spallation products data fit well with the experimental data [13].

4.4 Verification of burnable poison

The pressurized water reactor (PWR) fuel assembly of the Qinshan's phase II nuclear power plant, which contains 16 poisonous boron rods [21], is used for the verification of the fixed-flux method for the burnable poison calculation function; the enrichment of the ^{235}U is 2.6% (Fig. 8). The main parameters are listed in Table 3.

Table 3 Reference parameters used in the assembly of Qinshan's second nuclear power plant

Parameters	Value
Assembly type	17 × 17
Subtend distance (cm)	21.504
Cell pitch (cm)	1.26
Moderator temperature (K)	583.85
Fuel temperature (K)	924.15
Boron concentration (ppm)	0
Radius of fuel rods (cm)	0.4095/0.418/0.475
Radius of burnable poison (cm)	0.21375/0.23075/0.2413/0.4267/0.4369/0.48385/0.5625/0.6025
Composition of the burnable poison	0.74%^{10}B, 55.16%O, 1.11%Al, 39.694%Si

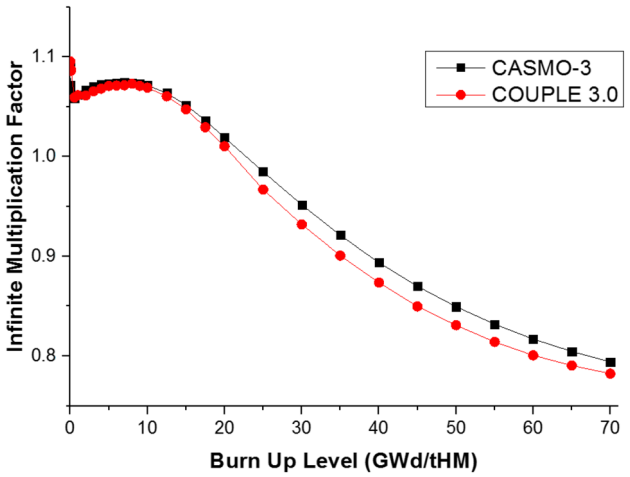

Fig. 9 (Color online) Burnup behavior of the infinite multiplication factor

A comparison of the results of COUPLE3.0 and CASMO-3 (Fig. 9) indicates that the trend is similar for the two methods. In the beginning, the two curves fit well and at values greater than 15 GWd/tHM and the COUPLE3.0 calculation yields lower values. These differences may be caused by the differences in the nuclear database because we used ENDF/B-VII.0 in COUPLE3.0.

5 Conclusion

COUPLE3.0 can run the transport module in parallel in the MPI environment, which greatly improves the calculation accuracy and efficiency. Two types of neutron irradiation modes with constant power and a constant flux radiation model are included in the program and can be used for the calculation of the burnup of fuel and burnable poison.

The proton irradiation calculation module can be used to analyze the accumulation behavior of the residual nuclides in the spallation target based on the running history and the influence of the accumulated nuclides in the target area on the neutron behavior of the subcritical system. The improvements in COUPLE3.0 also include the selection of important radionuclides and the calculation of the branching ratios for the excited states of the radionuclides, which makes the calculation more accurate. The program has a user-friendly interface and is convenient for post-processing, extracting information, and determining activity information for the nuclides.

The calculation and analysis of a fast spectrum plutonium benchmark for incineration indicate that COUPLE3.0 is applicable and reliable for the analysis of the fuel burnup. The analysis of the IAEA-ADS burnup benchmark problem and the comparison with the results from other research institutes show that the k_{eff} is lower than the average and the beam intensity is larger. We need to perform further analysis on the database selection and the calculation model. COUPLE3.0 cannot only be used for burnup analysis in ADS research, but also for the nuclide cumulative analysis of the target, the evaluation of the radiation characteristics of the spallation products, and their influences on neutronic characteristics, which can provide important reference data for the design and construction of ADS systems.

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