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Neutronics Performance Analysis of Cesium Transmutation in Pressurised Water Reactors

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Abstract. The rapid development of nuclear reactors has provided us with efficient and clean energy, but at the same time the disposal of spent fuel after fission reaction is facing many problems. In particular, some of the long-lived and highly radioactive fission products (LLFP) in the spent fuel have half-lives of tens of thousands of years or even hundreds of thousands of years, which pose a great threat to the natural environment of human beings. The international community has proposed the 'separation-transmutation' method to meet this challenge. This method refers to the extraction of LLFP from fission products, and the treatment of LLFP so that it can be converted into short-lived or low radioactivity nuclides. In this paper, the long-lived fission product Cs-135 was added to a pressurised water reactor fuel assembly for transmutation, and the effect of different Cs loading on the reactivity control of the fuel assembly was investigated. The results show that the introduction of Cs-135 reduces the Keff value of the fuel assembly, and the transient neutron lifetime of the core gradually decreases and the share of slow-generating neutrons increases as the CsF molar ratio becomes larger. In addition, it is found that after the introduction of Cs, the number of thermal neutrons in the fuel assembly decreases because Cs-133 captures and consumes a part of the thermal neutrons to be converted into Cs-135.

Keywords: Transmutation; Monte Carlo; fuel combustion; LLFP.

1. Introduction

As an important clean and efficient new energy source, nuclear energy plays an important role in power production and life. The rapid development of nuclear power has provided us with strong energy support and guarantee, but the nuclear reactors running all the time are also emitting radioactive substances constantly, especially some of the long-lived fission products (LLFP), which still have high radioactivity after a hundred years and will cause serious damage to the environment. Exploring effective treatment of LLFP has been an important issue for nuclear researchers at home and abroad. As of the end of June 2022, there are 660 nuclear power units in operation worldwide, with a total installed capacity of about 394 million kilowatts. For the commonly used commercial light water reactors, the general refuelling cycle is in the range of 1 to 1.5 years, and with an average fuel consumption of 50 GW-d/tU, the annual spent fuel unloaded from a nuclear power unit with an installed capacity of 1,000 MWe is estimated to be about 20 tU. Domestically, China's spent fuel stockpile exceeds 80 million tonnes as of January 2022, and the new spent fuel added is about 1,000 tHM per year [1].

Long-lived fission products such as Tc-99, I-129, Zr-93, Cs-135, which have half-lives up to 105-106 years, are radiotoxic and highly geochemically mobile, and may still have an impact on the environment in a repository. The Cs separated from reactor spent fuel mainly consists of three isotopes, Cs-133, Cs-135 and Cs-137, of which only Cs-135 is a long-lived nuclide with a half-life of 2.3 million years. Whereas Cs-133 is a stable nuclide, Cs-137 has a shorter half-life of 30.07 years.The transmutation of Cs-135 in a reactor is mainly through a neutron capture reaction to produce short-lived Cs-136, which then releases an electron decay to produce stable Ba-136.Previously scientists have learnt that the ratio of Cs-135 to Cs-137 identifies the source of cesium whether it comes from nuclear weapons or nuclear reactors in nuclear power plants [2].

Cs-135 and Cs-137 are fission products of uranium or plutonium, and Cs-135 is produced by the decay of Xe-135, a fission product of the uranium nucleus, without absorbing neutrons. After a

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nuclear reactor has been damaged, U-235, when irradiated by neutrons, will produce a series of radionuclides through nuclear fission reactions, including Cs-135. In addition, the cooling water after a nuclear accident contains a large amount of radionuclides such as Cs-135. These radionuclides will be discharged into the sea along with the nuclear effluent, causing continuous, long-term and potential hazards to the environment and human beings. Currently, one of the most promising methods for treating high-level radioactive waste is separation transmutation [3]. Separation is the extraction of the usable U and Pu elements, minor actinides (MA) and long-lived fission products (LLFP) from the spent fuel, and transmutation is the treatment of MA and LLFP elements to obtain short-lived or low radioactivity nuclides.

In this paper, a 17×17 pressurised water reactor (PWR) fuel assembly model is established, and the MCNP software is used to complete the establishment of the baseline fuel assembly model, and then Cs-135 is introduced for transmutation, and the effects of different Cs-135 introductions on the reactivity control of the fuel assembly are studied and calculated.

2. Introduction to the Calculation Procedure

In this paper, MCNP software is used to calculate and solve the related data. Monte Carlo (MC) is a solution method born in the process of studying the Boltzmann neutron transport equation, which uses random numbers to conduct statistical experiments to obtain statistical eigenvalues as the numerical solution of the problem to be solved. The Monte Carlo method has played an important role in the scientific research of atomic bomb engineering, and has been increasingly widely used in many aspects of physical engineering. Based on Monte Carlo, members of the Los Alamos National Laboratory in the United States have developed a large-scale multifunctional Monte Carlo ion transport program, MCNP (Monte Carlo Neturon and Photo Transport Code). safety, radiation shielding, medical physics radiotherapy, etc. MCNP is the main programme module which calls the main modules IMCN/MCRUN/XACT and PLOT respectively as required by the problem being run. To process a problem with the MCNP programme, an input file needs to be written in the format specified by the MCNP, in which information about the emitted particles (i.e., shape and position of the source, type of particles, and energy spectrum), the medium through which they pass (including chemical composition, density, and geometry), and the counts to be detected (detection positions, energy intervals) should be given in accordance with the problem to be solved, and then submitted to MCNP. Then submit it to the MCNP programme for processing to get the result, and then extract the information you need from the output file according to your needs.When receiving the paper, we assume that the corresponding authors grant us the copyright to use the paper for the book or journal in question.

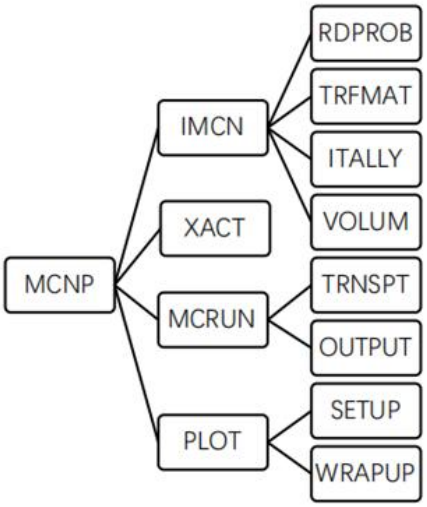

Fig. 1 Schematic diagram of MCNP programme module structure

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3. Modelling

3.1 Fuel Assembly Modelling

In this paper, a 17×17 pressurised water reactor fuel assembly model is proposed. In this paper, the pressurised water reactor fuel core block consists of a ZIRLO casing tube and a uranium dioxide ceramic core block encapsulated inside the tube. The two ends of the casing are sealed and welded with end races and the fuel casing is pre-filled with helium at a certain pressure in order to delay the occurrence of mechanical interaction between the core block and the casing as well as to avoid deformation and damage of the casing under high pressure environment. The outer shell is surrounded by water, the outer boundary of the water grid element is square, and the inner shell is the source region. In this paper, the fuel element is simplified into a grid element model, which is used as the most basic structural unit of the reactor. Its structural dimensions are shown in the table below. The fuel rods are made of UO2 with a density of 10.4668 g/cm3 , the zirconium alloy cladding material ZIRLO, including Zr (zirconium), Sn (tin), Fe (iron), Cr (chromium), O (oxygen), and the moderator material is boron water.

Table 1. Reactor grid element dimensions

parameter	Value（unit：cm）
Grid distance	1.26
axial height	2
outer diameter of fuel rod	0.81915
inner diameter of zirconium alloy shell	0.835660
outer diameter of zirconium alloy shell	0.949960

Fuel assembly geometry dimensions and material component composition, can be through the Universe set card, FILL filling card and LAT grid card, the structure of the same raster elements can be repeated structure and grid nesting to complete the modelling of the fuel assembly to build the fuel assembly cross-section shown in the figure below.

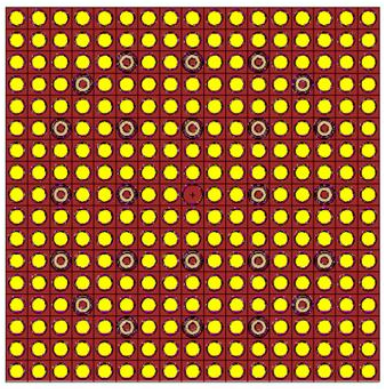

Figure 2.Fuel assembly modelling diagram

3.2 Neutronics Reactivity Modelling

In the reactivity control of pressurised water reactors (PWRs), downstream neutrons and slow onset neutrons have a large impact on the reactivity and power of the core. Instantaneous neutrons are released within 10-14 seconds after fission and directly affect the instantaneous power change of the reactor, and are produced in a very short time, which leads to an increase in neutron flux density and an increase in reactor power [4]. Retarded neutrons are usually produced within a few milliseconds or minutes after fission and account for less than 1% of the total fission neutrons, but the production of retarded neutrons prolongs the time response of the reactor, makes the reactor power slower and is critical for reactor control. Therefore, the slow-generating neutron share and related parameters should be statistically evaluated.

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The effective slow-burning neutron fraction (β_{eff}) is calculated using the following equation.

$$\beta_{eff} = 1 - \left(K_p / K_{eff} \right) \quad (1)$$

where Keff is the effective proliferation factor for the MCNP5 software cycle run and Kp is the transient neutron proliferation factor.

The effective neutron generation time (Λ_{eff}) is calculated as.

$$\Lambda_{eff} = l_p / K_{eff} \quad (2)$$

where Λ_{eff} is the effective neutron generation time and lp is the software-calculated instantaneous neutron lifetime.

4. Experiments and Analysis of Results

In the spent fuel of light water reactors, the mass shares of Cs-133, Cs-135 and Cs-137 are 76%, 17% and 7%, respectively, and these shares are used as the different proportions of Cs elements introduced into the core pressurised water reactor [5]. Since there is no solubility limitation of CsF in pressurised water reactors and the addition of CsF to the core has a relatively small effect on the chemical composition of the pressurised water reactors, the choice was made to use Cs-135 and its isotopes added to pressurised water reactors in the form of CsF loading for the study of neutronic properties. The following table shows the change in the effective multiplication factor of the fuel assembly after introducing different Cs-135 loadings.

The introduction of LLFPs nuclides into the reactor may cause a decrease in the effective core multiplication factor due to their large neutron capture cross section. As can be seen from the table, the introduction of Cs-135 reduced the Keff value of the fuel assembly. In Table 2, the βeff has been showing an increasing trend from 0 mol% to 1 mol% for CsF loading. This may be due to the fact that after the introduction of Cs into the core, there will be a large amount of Cs-133 to generate Cs-135 by capturing neutrons, and this process will continue to exist for a limited operating time, with a decrease in the number of fissionable neutrons, and thus an increase in the share of slow-generating neutrons.

Table 2. Table of effective proliferation factors and retarded neutron fractions of fuel assemblies at different CsF device sizes

CsF loadings/mol %	Keff	Standard Error	Kp	Standard Error	βeff
0	1.00774	0.00022	1.00502	0.00023	0.26991%
0.3	0.90718	0.00025	0.90450	0.00022	0.29542%
0.5	0.86247	0.00023	0.85972	0.00021	0.31885%
0.7	0.82589	0.00022	0.82321	0.00021	0.32450%
1	0.78055	0.00024	0.77795	0.00022	0.33310%

Cs-135 was added to the fuel, and the corresponding neutron flux density was output using MCNP software, and the results were recorded in Fig. 3 and Fig. 4, from which it can be seen that the neutron shares of different energy segments are different, with a lower share of high-energy neutrons and a higher share of low-energy neutrons. Under the initial loading condition of the core, the share of fast neutrons with larger energy than 0.1 Mev is 20.1884%, and the share of thermal neutrons with smaller energy than 0.1 Mev is 79.8116%. Calculations based on the data in Fig. 4 show that the proportion of fast neutrons larger than 0.1 MeV increases gradually with the increase of the molar ratio of CsF introduced into the core, which are 20.7925%, 21.0636%, 21.3024% and 21.6099%, respectively, probably due to the fact that after the core is introduced into the Cs, the Cs-133 will capture and consume a part of the thermal neutron to be converted into the Cs-135, resulting in the decrease in the number of thermal neutrons.

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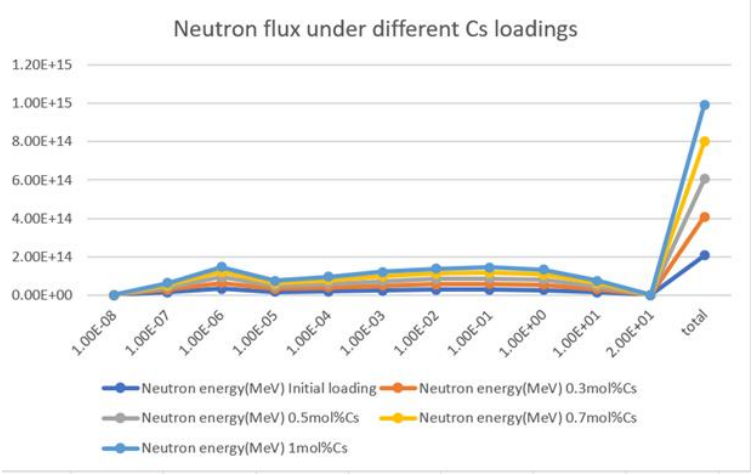

Fig. 3 Neutron flux density distribution with different CsF loading

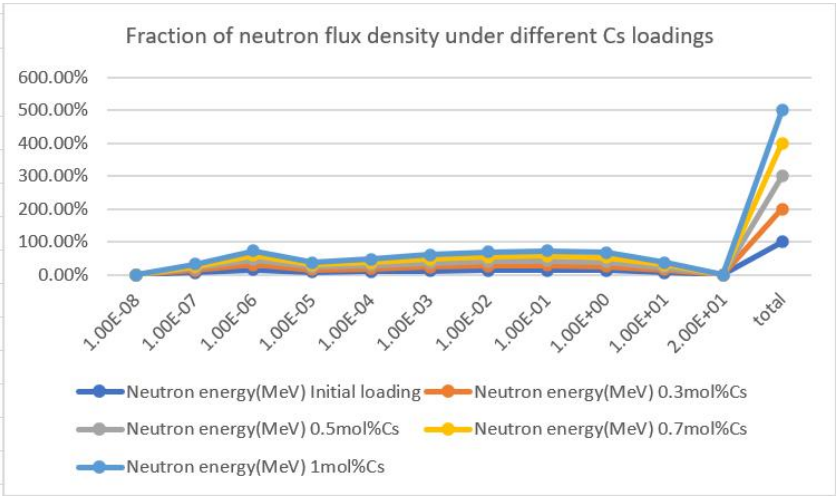

Fig. 4 Distribution of neutron flux density shares with different CsF loading

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