Calculation of the Amounts of Fission Products (^{85}Kr, ^{129}I, ^{137}Cs) in Pressurized Water Reactors (PWR)

Ali. K. Hasan[1], Najim. A. Askouri[2], Haidar. M. Talib[3]

[1]Professor, University of Kufa-Faculty of Education/ for Girls, Iraq
[2]Assistant Professor, University of Kufa-Faculty of Education/ for Girls, Iraq
[3]Master, University of Kufa-Faculty of Education/ for Girls, Iraq

Abstract: ***The fission process of uranium-235 nucleus, produces fission products such as (^{85}Kr, ^{129}I, ^{137}Cs) as the most important products. The risks of these products on the reactor safety and the environment are important the quantities of these products depend on the fission rate inside the nuclear fuel; ie, on their yields. Uranium dioxide ceramics UO_2 are widely used in pressurized water reactors (PWR). The calculations were obtained need by using (MATLB R2011b), through calculating the effects of thermal power, neutrons flux, cooling water temperature. It was found that the quantities of fission products increased with thermal power and neutrons flux. The increase of coolant temperature led to decrease of the amounts of fission products.***

Keywords: fission, products, 85Kr, 129I, 137Cs

1. Introduction

Uranium dioxide is a ceramic refractory uranium compound. In many cases used as a nuclear fuel. The most widely used form is cylindrical, sintered pellets. The pellets are ground to specified dimensions and (for thermal reactors) are loaded into long thin zircaloy-4 tubes which serve as cladding contains helium gas [1]. To make fuel rod (Fig.1). The PWR fuel currently used the fuel assembly types are 17x17. This selection of UO_2 over other potential fuel materials is based on its excellent combination of chemical stability, especially to water reactor coolants; compatibility with potential cladding materials zircaloy-4 [2]; dimensional stability under irradiation; very high melting point; and straightforward, economical production from diffusion plants, The basic properties of uranium dioxide are given in Table 1,[3]. These desirable characteristics were recognized at a very early stage in the development of power reactors [4], pressurized-water-type nuclear power reactor uses light water as the reactor coolant and moderator in the state of high temperature and high pressure not boiling in the reactor core (primary system: reactor coolant system) and sends the high-temperature and high-pressure water to steam generators (primary system) to generate steam through heat exchangers (steam system: secondary coolant system) for a turbine generator to generate electricity[5,6]. The fission process of uranium-235 nucleus in the reactor produce radioactive fission gases among other radio nuclides [7].

Table 1: Basic Properties of Uranium Dioxide at 0.1 MPa, 298 K [2]

Property	Value
Crystal structure	FCC
Theoretical Density, (kg/ m^3)	10960
Melting point, ̊C	2850
Boiling point, ̊C	3542
Thermal Conductivity (W/m. ̊K)	8.68
Molecular mass (amu)	270.3

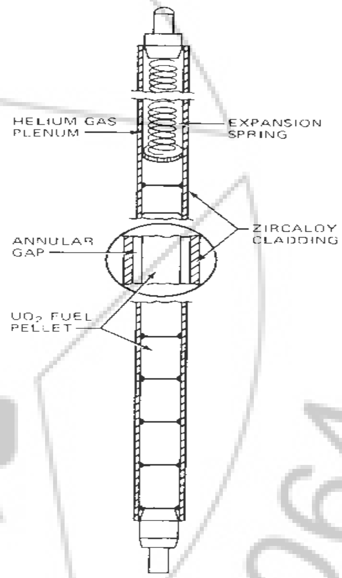

Figure 1: Construction of a uranium dioxide fuel element [1]

2. Theoretical Part

Nuclear fission is a phenomenon where a heavy parent nucleus is divided into two or more daughter nuclei spontaneously or as a result of nuclear reactions [8]. The distribution of fragment masses following a nuclear fission is one of the most basic quantity and has been observed since just after the discovery of fission by Hahn and Strassmann in 1938 [9]. In the fission process, a neutron interacts with the target nucleus creating a compound nucleus that is unstable and splits into smaller nuclei releasing two or more neutrons, energy and fission fragments [10]. The fission fragments are radioactive and decay via β decay, which changes the atomic number but not the atomic mass number. The maximum yield for any one isotope is less than 7% [11]. It also indicates that fission products fall into two broad groups: a group of light nuclei with mass number between 80 and 110, Like (Kr-85) and a heavy group with mass numbers between 125 and 155. Like (I-129,Cs-137). Krypton atoms form bubbles that may alter the structural and mechanical properties of the fuel, Iodine and Cesium have long life isotopes and are known to be highly dissoluble in water and to have a high fission yield. The risks of these products on the reactor safety and the environment are feasible.

3. Calculation

The amounts of fission products depend on the fission rate inside the nuclear fuel **[1].** The fission rate can be roughly estimated the average values for effects of thermal power, neutrons flux, cooling water temperature. Therefore in a reactor of volume V [m3] [1, 5]

$$F = V\, N\, \sigma_f\, \varnothing \tag{1}$$

Where : N is number of fissile nuclei [nuclei/m^3], σ_f is fission cross section of the fissile material [m^2/nucleus], $\varnothing$ is thermal neutron flux [n/cm^2. sec]. The change of coolant temperature affects the fission rate, and the following formula is used to correct microscopic cross sections for temperature: [12]

$$\sigma = \sigma_\circ \left(\frac{t_\circ}{T_b}\right)^{1/2} \tag{2}$$

Where: σ is microscopic cross section corrected for temperature, $\sigma_\circ$ is microscopic cross section at coolant temperature, $t_\circ$ is temperature at 20 $^\circ$C, T_b is coolant temperature.

Let eq (2) in eq (1)

$$F = V\, N\, \sigma_\circ \left(\frac{t_\circ}{T_b}\right)^{1/2} \varnothing \tag{3}$$

Assuming that the reactor has been operating for enough time that nearly all of the radioactive decay energy is being deposited as heat, and that fission rate for ^{235}U required to produce 1 watt of thermal power is [13]:

$$F = \frac{1\ \text{(J/sec)}}{1.6022 \times 10^{-13}\ \text{(J/MeV)} \times 200\ \text{(MeV/fission)}} \tag{4}$$

The required fission rate equal (3.12×10^{10} fission/s), approximate average fission rate through the effects of thermal power is [2,12]

$$F = 3.12 \times 10^{10}\ \text{P (W)} \tag{5}$$

The amounts of fission products can be readily determined from the of fission rate and the fission yield Y_i . Let N_i be the concentration of a particular fission product at a time t following reactor startup, The rate at which the concentration of a nuclear species (Ni) in a reactor core changes with time is given by [2]:

$$\frac{dN_i}{dt} = F\ Y_i - \lambda_i N_i - \sigma_a^i N_i\, \varnothing \tag{6}$$

Where ($\lambda_i N_i$) decay rate, ($\sigma_a^i N_i \varnothing$) destruction rate, For the case where $\sigma_a^i >> 0$, and assuming $N_i = 0$ at $t = 0$, the number of atoms N_i at any time t after operation begins[12] :

$$N_i\,(t) = \frac{F \times Y_i}{\lambda_i}\ (1 - e^{-\lambda_i t}) \tag{7}$$

The concentration of each fission product depends strongly on the time t that the reactor has been operating decay constant λ_i . If $\lambda_i t$ <<1 then the concentration will increase linearly with time [1,12]:

$$N_i\,(t) = F\, Y_i\ t \tag{8}$$

Eq (8) gives the production rate of the fission products in a unit volume of reactor. where F (fission /sec) is the fission rate, Yi (atoms/fission) is the total cumulative yield, Table 2 lists the cumulative yields and t (sec) is any time t after operation begins. By applying fitting property in program (MATLAB R2011b) we got a mathematical equation which proved that, the error ratio in the calculations are very small and used this equation to calculate the quantities of fission products at (1000,2000,3000) MW

$$M_h = a \times t \tag{9}$$

Where: M_h (Mol) is quantities of fission products, t (sec) is time of reactor operation, a (Mol/sec) is constant in Table 3.

Table 2: Lists The Cumulative Yields of (^{85}Kr, ^{129}I, ^{137}Cs) Nuclide [14]

Nuclide	Half-life	Units	Cumulative fission yields (%)
Kr-85	10.752	year	0.286
I-129	1.7×10^7	year	0.706
Cs-137	30.05	year	6.221

Table 3: Lists The Values of Constants in Eq (9).

Nuclide	a^1	RMSE	a^2	RMSE	a^3	RMSE
Kr-85	1.48 e-7	1.27 e-15	2.97 e-7	2.54 e-15	4.46 e-7	3.14 e-15
I-129	3.67 e-7	2.62 e-15	7.34 e-7	5.25 e-15	1.10 e-6	1.01 e-14
Cs-137	3.23 e-6	3.36 e-14	6.47 e-6	6.73 e-14	9.70 e-6	9.19 e-14

a^1: value of constant at thermal power 1000 MW.
a^2: value of constant at thermal power 2000 MW.
a^3: value of constant at thermal power 3000 MW.
RMSE: Root Mean Square Error

4. Results and Discussion

Assuming in this study that pressurized water reactor PWR operational within agiven rate of thermal neutrons flux is (3.3×10^{13} - 5.5×10^{12}) n/cm^2.s, thermal power is (500-3000) MW and operation time starts is (86400 to 31104000) sec. The amounts of fission products (^{85}Kr, ^{129}I, ^{137}Cs) resulted from calculations are shown in figs (2,3,4), It was found that fission products quantities increase within operation time, thermal power and neutrons flux, e.g., for ^{85}Kr, the generated amount at power 3000 MW, flux 3.3×10^{13} n/cm^2.s and operation time 86400 sec, equal (0.038 mol). But the product amount of same power and flux at 31104000 sec, equal (13.977 mol). As noted the increase in reactor power, the fission products amount also increase, ^{129}I at the time 31104000 sec and power 500 MW the generated amount equal (5.709 mol), and at 3000 MW the generated amount equal (34.256 mol). So the effect of coolant temperature on the fission rate into account, the calculations were within two degrees of the coolant temperature are: (383,583) $^\circ$K. The high degree of coolant temperature at the maximum thermal power of the reactor. Amounts with effects coolant temperature through figs (5,6,7). The calculations with increase of coolant temperature led to decrease of radioactive gases amounts, Like ^{137}Cs at power 3000 MW and 383 °K the amount generated equal (365.092 mol), at the same power and (583 °K) equal (294.492 mol).

Therefore, it is essential to maintain the accumulated fission gaseous or evaporated radio nuclide under control without affecting the performances of fuel assembly to avoid environmental potion.

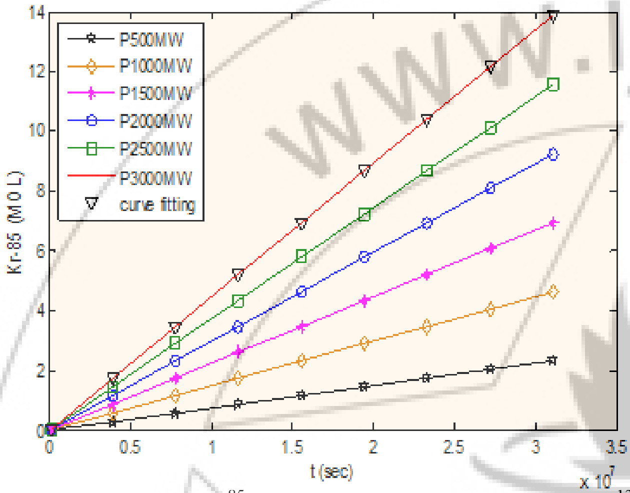

Figure 2: Amounts of ^{85}Kr at thermal neutron flux (3.3×10^{13} - 5.5×10^{12}) n/cm^2.s

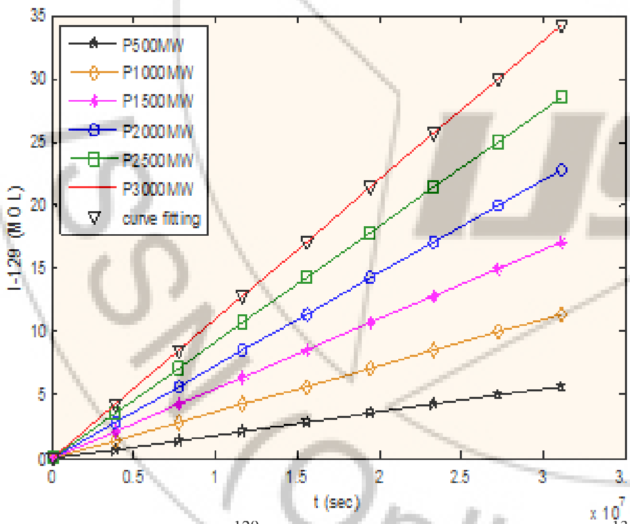

Figure 3: Amounts of ^{129}I at thermal neutron flux (3.3×10^{13} - 5.5×10^{12}) n/cm^2.s

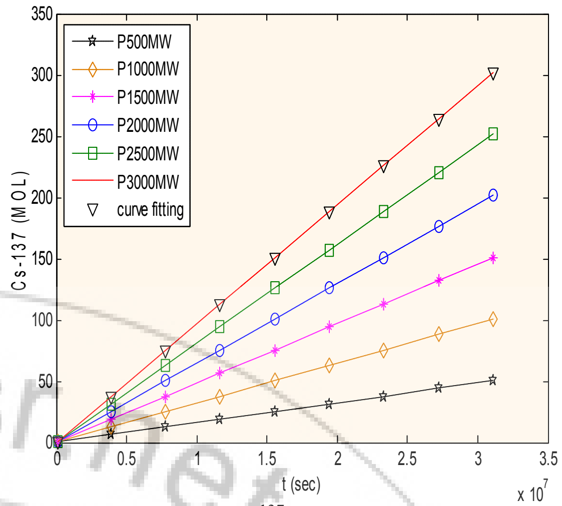

Figure 4: Amounts of ^{137}Cs at thermal neutron flux (3.3×10^{13} - 5.5×10^{12}) n/cm^2.s

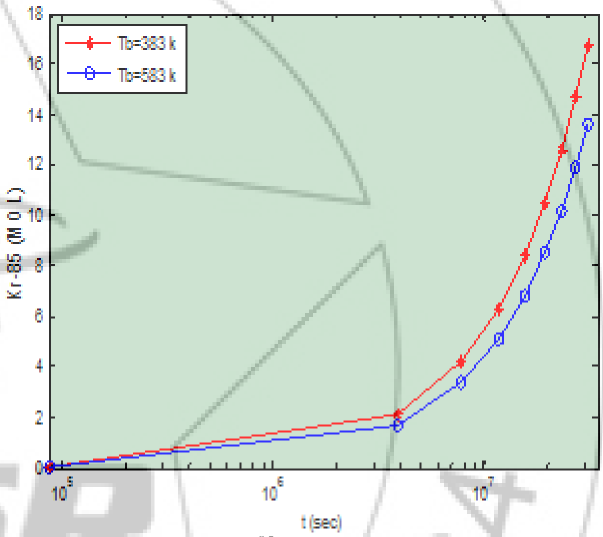

Figure 5: Amounts of ^{85}Kr at coolant temperature two (383,583) °K

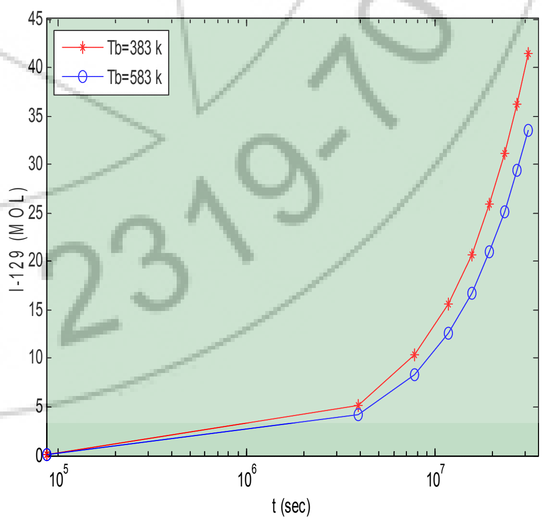

Figure 6: Amounts of ^{129}I at coolant temperature two (383,583) °K

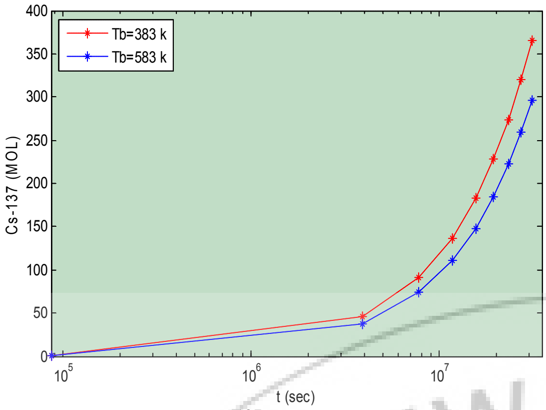

Figure 7: Amounts of ^{137}Cs at coolant temperature two (383,583) $^{\circ}$K

5. Conclusions

In the present work using the computer program (MATLAB R2011b) for calculation the fission products. It focuses on the three existent components in pressurized water reactors are: (^{85}Kr,^{129}I,^{137}Cs). Using the fission rate data and fission yield from INDC-0534 (IAEA, 2008). These study showed several important results are in Figures (2,3,4,5,6,7), the agreements between the present calculation results and measured data say that the present computational algorithm written in the matlab program is qualify for precise calculations of fission product.

References

[1] M. MA. Benjamin, " Nuclear Reactor Materials and Application", Vannostrad Reienhold, 1982

[2] S. Glasstone and A. Sesonske, "Nuclear Reactor Engineering", Third Edition, Van Nostrand Reinhold,1981

[3] IAEA, "Advanced Fuel Pellet Materials and Fuel Rod Design for Water Cooled Reactors", 2010

[4] A. Strasser, P. Rudling, " Fuel Fabrication Process Handbook", A. N. T. International, Sweden, 2005

[5] T. Jevremovic, "Nuclear Principles in Engineering", Springer, 2005

[6] O. Noboru, J. Takagi, J. Nucl. Sci. and Technology, Vol. 2 (4), p.(127-131), 1965

[7] T. fukuda, S. omori and S. Yamagishi, J. Nucl. Sci. and Technology, Vol. 10 (4), p.(242-249), 1973

[8] A. I. El-Shanshoury, Arab J. Nucl. Sci. and Applications, Vol. 45 (4), p.(257-264), 2012

[9] P. Ngoc Son, Asian J. Sci. and Technology, Vol. 5 (5), p.(295-298), 2014

[10] L. S. Tong, J. Weisman, "Thermal Analysis of Pressurized Water Reactors", American Nuclear Society, 1996

[11] JNES, " The Pressurized Water Reactor", Japan Nuclear Energy Safety Organization, 2011

[12] DOE Fundamentals Handbook, "Nuclear Physics and Reactor Theory", U.S.-Department of Energy, Washinghton,1993

[13] M. K. AL-Ghitta, "Solved problems in nuclear physics", Iraq Atomic Energy Commission, 1977

[14] A. L. Nichols, D. L. Aldama and M. Verpelli, " Nuclear Data for Saffguards : Data Base Extensions", IAEA, 2008