Effect of Fission Fragments on the Properties of UO$_2$ Fuel of Pressurized Water Reactors

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ABSTRACT

The effect of Xenon (Xe) and Strontium fission fragments on the properties of UO$_2$ fuel of pressurized water reactors has been evaluated using SRIM-2010 program. The released fission products being highly energetic with different masses, different phase states, and carry different charges cause ionization of the fuel from the surface up to the maximum range with the formation of electron-hole pairs. When the kinetic energy falls below the displacement energy of U and O atoms phonon production takes place. The collision of energetic fission products with the fuel results in the creation of recoil-vacancy pairs. The uranium and oxygen recoils released during the collision process changes the oxygen to uranium ratio of the UO$_2$ matrix. The fission fragments as well as the recoils reside in interstitial positions in the structure of UO$_2$ fuel with the result in increasing the internal stresses. The magnitude of damage introduced in the fuel is calculated on the bases of the fission rate of 4% enriched UO$_2$. The released fission fragments and recoils as well as the increase in the fuel temperatures cause swelling of the fuel, increase fuel-clad interaction, enhance cracking of the pellet and accelerate the release of fission gases. The poisoning effect of xenon fission fragment is also presented.

Key Words: Fission Fragments/ UO$_2$ fuel/ Power Reactors/ Ionization/ Phonons/ Collision/ Crack Formation/ Poison Effect.

INTRODUCTION

Pressurized water reactors (PWRs) comprise a majority of all western nuclear power plants and are one of two types of light water reactor (LWR), the other type being boiling water reactors (BWRs). In a PWR the primary coolant (superheated water) is pumped under high pressure to the reactor core, then the heated water transfers thermal energy to a steam generator. All LWRs use ordinary light water as coolant, neutron moderator and reflector. The most common type of nuclear fuel used in PWRs is heavy fissile elements that can be made to undergo nuclear fission chain reactions in a nuclear fission reactor. The fuel is composed of bundles of fuel rods containing the fuel material. The most common fissile nuclear fuels are $^{235}$U and $^{239}$Pu. A sintered uranium oxide ceramic is formed into pellets and inserted into Zircaloy tubes that are bundled together. The fuel bundles usually are enriched several percent in $^{235}$U (3-5 wt %). Uranium oxide is dried before inserting into the tubes to try to eliminate moisture in the ceramic fuel that can lead to corrosion and hydrogen embrittlement. The Zircaloy tubes are pressurized with helium to try to minimize pellet cladding interaction (PCI) which can lead to fuel rod failure over long periods.

This work presents theoretical study on the effect of fission fragments on the properties of 4% enriched UO$_2$ fuel of pressurized water reactors. It includes the damage resulting from fission products, swelling and cracking of pellet, fuel-clad interaction as well as fission gas problems. The
defects resulting in fuel material is simulated by using SRIM-2010 computer code \(^{(3,4)}\). The other problems were reviewed with the help of our previous work and published results.

**RESULTS AND DISCUSSION**

It is well known that when \(^{235}\text{U}\) absorbs thermal neutron will form unstable \(^{236}\text{U}\) compound which splits to give two fission fragments and 2-3 fast neutrons. The fission process is accompanied with the release of about 200 MeV. Fission products are released with a kinetic energy of about 167 MeV \(^{(5)}\). The balance of the energy is carried out by fast neutrons (2 MeV), gamma rays (prompt and delayed), beta particles and neutrinos \(^{(5)}\). The mass distribution of the fission products is represented by a fission yield curve, in which the percentage of the different products is plotted against mass number \(^{(6)}\). Yields vary from \(10^{-7}\%\) to about \(6\%\) and the range is so large that a logarithmic scale is used for the ordinate of the yield curve. The maximum yields lie near mass numbers of 95 and 139. The fission process is highly asymmetric. The energy of the fission products is inversely proportional to their mass numbers. Accordingly, the elements corresponding to maximum mass numbers are \(^{94}\text{Sr}\) and \(^{140}\text{Xe}\), and their corresponding kinetic energies are 99.9 MeV and 67.1 MeV respectively, while the light and heavy ones at the tail of the distribution curve are \(^{72}\text{Zn}\) and \(^{160}\text{Gd}\). Although the contribution of Zn and Gd in the damage of the fuel material is negligible since their released amounts are \(10^{-4}\%\) and \(10^{-3}\%\), respectively. The importance of the lightest metal Zn lie in knowing its maximum range in the fuel rod since its kinetic energy is 114 MeV \(^{(2)}\).

The released fission products are highly charged and can be presented as ionized ions accelerated to their corresponding kinetic energies. The released fast neutrons with energy ranging from an average energy of 2 MeV to the maximum 14 MeV were presented in a previous investigation by protons with kinetic energy higher than 2 and 14 MeV i.e., about 3 MeV\(^{(2)}\) and 17 MeV\(^{(7)}\) respectively. The contribution of fast neutrons in the damage of the fuel is considerably small if compared with the heavy fission products \(^{(8,9)}\). Consequently, the contribution of fast neutrons in the damage of the fuel is neglected.

The fission ions are used as projectiles incident perpendicularly on fuel target material and the corresponding radiation damage is simulated using the SRIM-2010 computer program \(^{(2,4)}\). The computer programs for the calculation of the stopping and the ranges of ions in matter SRIM 2010 \(^{(3)}\) is used to simulate the implantation process. It uses quantum mechanical treatment of ion-atom collisions (assuming a moving atom as an 'ion', and all target atoms as 'atoms'). These calculations are made very efficient by the use of statistical algorithms which allow the ion to make jumps between calculated collisions and then averaging the collision results over the intervening gap. During collision, the ion and atom have a screened Coulomb collision, including exchange and correlation interactions between the overlapping electron shells. The ion has long range interactions creating electron excitations and plasmons within the target. These are described by including a description of the target’s collective electronic structure and inter atomic bond structure when the calculation is set. The charge-state of the ion within the target is described using the concept of effective charge, which includes a velocity dependent charge state and long range screening due to the collective electron-sea of the target. Fifty ions are used in the simulation process.

The tracks of the accelerated Sr and Xe ions of fission products as well as the corresponding released recoils are shown in Figs (1 & 2). Figures (1 & 2) are those for Sr and Xe ions accelerated to 99.9 MeV and 67.1 MeV, respectively. The maximum longitudinal range of the ions resulted from fission process are 10.3 \(\mu\text{m}\) and 7.17 \(\mu\text{m}\) for Sr and Xe, respectively. It is obvious from the results that the range increases with the decrease in the mass number of the fission fragment. Again, neutrons being neutral have little effect in the damage of the fuel \(^{(8)}\). Moreover, it is worth mentioning that the
maximum value for the scale of the target depth axes for Sr and Xe ions are 30 µm and 15 µm, respectively.

During the trip of the accelerated fission products different processes occur. The main process is concerned with the collision events of the ions and recoils with the fuel material. The collision events begin from the start up to the maximum range of the ions in the material. Energy transfer from the ions to U and O atoms takes place during the collision process. The energy loss varies from a maximum value during head-on collision to a minimum at glancing collisions (5). The energy loss is given to create recoil-vacancy pairs, kinetic energy of recoils, electron-hole pairs during the ionization process, and lastly as heat or phonons production. The following gives the details of the involved processes.

**1. Collision Events**

The total displacement target depth distribution curves for Sr and Xe are shown in Figs (3& 4). Figure (3) shows that the target displacements for Sr ions increases slowly with the increase in target depth reaching 0.39 displacements/ion A° up to about 6.3 µm followed by an increasing rate till reaching a peak of 2.05 displacements /ion A° at a range around 9.0 µm and drops suddenly to zero at the maximum range of 9.35 µm. Area under the curve for Sr displacements gives $5.26 \times 10^4$ recoils/ion and vacancies/ion. Xenon ions show collision events higher than those of Sr ions. The starting slope for the increase in the distribution curve is much faster than that of Sr reaching a value of 0.55 recoils/ion A° at 2.93 µm from the surface followed resonating peaks with increasing values reaching a maximum of 3.06 recoils/ion A° up to 9.44 µm. An extremely fast drop in the maximum value to 1.0 displacements /ion A° is observed at 6.5 µm reaching the end of the range at 7.17 µm. Area under the curve for Xe ions gives $9.64 \times 10^4$ recoils/ion and vacancies/ion. The high values of the area under the curves for Sr and Xe ions are attributed to their high masses, charges and kinetic energies.
2. Energy to Recoil

Energy to recoils-target depth distribution curves for Sr and Xe ions are shown in Figs (5& 6). The curves represent the energy absorbed by U and O atoms. It is obvious that the energy absorbed by both types of atoms from their collision with Sr ions amounts to 3.87 MeV/ion while that absorbed by Xe ions is 7.99 MeV. Although the kinetic energy of Xe ions is less than that of Sr ions its absorbed energy by recoils is higher. This can be attributed to the higher mass and high collision cross section of Xe ions with target atoms. The absorbed energy by U and O atoms is equivalent to 3.87% and 11.91% of the total kinetic energy of Sr and Xe ions, respectively. Moreover, it should be noticed that the position and amplitude as well as the number of collision resonance peaks of both ions are much higher for Xe ions than those of Sr ions and characterize the type of fission fragment.

3. The Ionization Process

The ionization by Sr and Xe ions as well as U and O recoils are shown in Figs (7& 8). It is obvious from the results that the energy loss in the ionization by ions is considered the main process while that
resulted from recoils is extremely small (the lower dark part at the end of the curves). Area under the curves for Sr and Xe ions show that the magnitude of energy loss in the ionization process amount to 96.06% and 87.97% of the total kinetic energy, respectively while that resulted from U and O recoils are 1.85% and 6.33%, respectively.

Fig. (7): Ionization energy-target depth distribution curve for Sr ions and recoils.  
Fig. (8): Ionization energy–target depth distribution curves for Xe ions and recoils.

Although the energy loss in the ionization process is considered extremely high their effect in causing the damage in the fuel material is negligible. The resulted electron – hole pairs recombine and annihilate at the temperature of the PWR (300°C). The energy released from the recombination process is given as heat energy to the surrounding and thus reduces the internal stresses resulted from the interstitial recoils.

4. Phonon Production

Energy to phonons – target depth distribution curves for Sr and Xe ions are shown in Figs (9& 10). It is obvious from the results that the energy given as phonons from the collision of recoils is extremely higher than that from ions. Phonon energy results from non-displacement collisions of U and O recoils with the target material. When the energy of recoils is below that of the displacement energy of the target atoms (E_d = 25 and 28 eV for U and O atoms, respectively) non-displacement collisions take place and the energy is lost as heat (phonons). Phonon production from recoils resulted from Sr and Xe ions were calculated at 1.92% and 5.26% of the total kinetic energy of both ions, respectively. Again the position and amplitude of phonons production resonance peaks from both ions are different and characterize the type of fission fragment.

Fig. (9): Phonons distribution curve for Sr ions and recoils.  
Fig.(10): Phonons distribution curve for Xe ions and recoils.
5. Damage Resulting From Fission Fragments

The number of recoils released from the collision of Sr and Xe with the fuel material is presented in this section concerning collision events. The total number of displacements resulted from Sr ions amounted to \(4.937 \times 10^4\) recoils/ion and vacancies/ion while that produced from Xe ions is \(9.05 \times 10^4\) recoils/ion and vacancies/ion after subtracting the replacement collisions of \(5.927 \times 10^3\) and \(3.230 \times 10^3\) ions, respectively. The released recoils from neutrons are extremely small if compared with Sr and Xe ions. The actual number of produced recoils is much higher than the given values since the number of fission fragments resulted from the fission process is unbelievably high and can be given in the following section. The fission rate \(R_f\) resulting from 4% enriched UO\(_2\) pellets stacked in 3.658 m\(^3\) active fuel length can be calculated from the equation:

\[
R_f = 0.04 \left( S_f F_{th} V \right)
\]

Where, \(S_f\), \(F_{th}\), and \(V\) are the macroscopic fission cross-section, the thermal neutron flux and the volume of UO\(_2\) in the fuel rod, respectively. The macroscopic fission cross-section is calculated from the density of UO\(_2\) (\(\rho\)), Avogadro's number (No) and the molecular weight of UO\(_2\) (M) as well as its microscopic fission cross-section (\(s_f\)), respectively. Hence, the value of \(S_f\) is given by \((\rho No/M) s_f\). The values given in parameters of the previous equation are; the density \(\rho\) (10.4 g cm\(^{-3}\)), Avogadro's number \(No\) (6.02 \(\times\) \(10^{23}\)) atom/mol, molecular weight \(M\) (267 g), the microscopic fission cross section \(s_f\) (380 b), the flux \(F_{th}\) (4.5\(\times\)10\(^{13}\) n/cm\(^2\)s\(^{-1}\))\(^{(9)}\) and the volume \(V\) (cm\(^3\)). For 0.819 cm diameter pellet and 365.8 cm\(^3\) the volume of the fuel rod is 192.9 cm\(^3\). Using the above given values in the above equation the \(R_f\) is calculated at 7.735 \(\times\) \(10^{16}\) fissions/s. For 4% enriched fuel the resulting fission rate 3.1 \(\times\) \(10^{15}\) fissions/s.

Since each fission process produces two fission fragments (Sr and Xe) and 2.5 neutrons, the number of fission fragments produced per second is 3.1 \(\times\) \(10^{15}\) /s for each Sr and Xe ion. As given above, the number of recoils produced from Sr and Xe ions is 4.9737 \(\times\) \(10^4\) and 9.05 \(\times\) \(10^4\) recoils/ion, respectively. The corresponding value for fast neutrons is negligible if compared with those of Sr and Xe. However, the total number of recoils produced by the fission products per second is 1.542 \(\times\) \(10^{20}\) and 2.81 \(\times\) \(10^{20}\) for each Sr and Xe recoil, respectively. This rate of recoil production is considered very high from the point of view of the formation of interstitials in the UO\(_2\) matrix since for a reactor working time of about 1.5 years (4.73 \(\times\) \(10^7\) s) the number of interstitials becomes extremely high. For example the total interstitials resulting from Xe becomes 1.33 \(\times\) \(10^{28}\) recoils per fuel rod. The corresponding value for Sr is 7.294 \(\times\) \(10^{27}\). Because of annealing of part of U and O atoms residing in interstitial position in the fuel matrix during irradiation at the reactor operating condition, the situation in the fuel materials becomes very bad.

Materials in a high radiation environment (such as a reactor) can undergo unique behaviours such as swelling, and non-thermal creep. If there are nuclear reactions within the material (such as what happens in the fuel), the stoichiometry will also change slowly over time. These behaviors can lead to new material properties, cracking, and fission gas release. As the fuel is degraded or heated the more volatile fission products which are trapped within the uranium dioxide may become free\(^{(8)}\). As the fuel expands on heating, the core of the pellet expands more than the rim which may lead to cracking. Because of the thermal stress thus formed the fuel cracks, the cracks tend to go from the centre to the edge in a star shaped pattern. The expansion of the core of the fuel pellet will result in its contact with the zircaloy cladding material. On prolonged irradiation time, reaction between the fuel and the clad will result in buckling the fuel rod. The distortion in the fuel rods results in blocking the cooling channels end consequently affects considerably the heat transfer system of the reactor.
In order to better understand and control these changes in materials, these behaviours have to be studied. A common experiment to do this is post irradiation examination, in which fuel will be examined after it is put through reactor-like conditions. Due to the intensely radioactive nature of the used fuel this is done in a hot cell. A combination of nondestructive and destructive methods of PIE is common methods for testing radioactive materials\(^{(11)}\). Post Irradiation Examination (PIE) is used to check that the fuel is both safe and effective. After major accidents the core (or what is left of it) is normally subject to PIE in order to find out what happened. In addition to the effects of radiation and the fission products on materials, a need to consider the temperature of materials in a reactor, and in particular, the fuel is required\(^{(12,14)}\).

6. Fission-Product Poisoning

During the operation of a reactor, the total quantity of fission products (FP) present increases. Although most of the FP has relatively low cross sections, there are some which are very strong parasitic absorbers of thermal neutrons and act to poison the reactor. The most of these is \(^{135}\)Xe which has the high cross section of \(2.7 \times 10^6\) barns at \(0.025\) eV energy\(^{(15)}\). Although \(^{135}\)Xe is produced as a direct fission product with a yield of \(0.3\) percent, the main source of \(^{135}\)Xe is from the radioactive decay of \(^{135}\)Te \((T_{1/2} \text{ less than } 2 \text{ min})\) which in turn decay to \(^{135}\)Xe \((T_{1/2} 6.7 \text{ h})\)\(^{(15)}\). The \(^{135}\)Xe production is dependent on the reactor flux.

The poisoning of a reactor is defined as the ratio of the number of thermal neutrons absorbed by the poison to the number absorbed in fissionable material. The poisoning factor increases considerably with the increase in reactor flux in the range \(10^{12}-10^{14}\) n/cm\(^2\) reaching a maximum of about \(0.056\) at a flux of \(10^{15}\) n/cm\(^2\) sec. Consequently, sufficient excess reactivity must be available in a reactor to override the xenon poisoning which is not felt until several hours' operation at high power.

CONCLUSIONS

The study of the effect of fission fragments on the properties of UO\(_2\) fuel of pressurized water reactors showed several important results. The fission of \(4\)% enriched UO\(_2\) resulted in the release of two fragments and an average of 2.5 neutrons with high energy. Strontium and Xenon fission products with kinetic energies 99.9 MeV and 67.1 MeV, and neutrons with 2 MeV collide with UO\(_2\) with the release of energetic U and O recoils. The effects of the ionization and phonon processes which are responsible for the absorption of more than 95% and 87% of the total energy of Sr and Xe fission products are irrelevant if compared with the collision events since most of the energy is released as heat. In the collision process the ejected U and O atoms or recoils reside in interstitial places in the matrix of the fuel material. The calculated number of recoils introduced in UO\(_2\) per fuel rod at reactor working time of 1.5 years is \(7.9 \times 10^{27}\) and \(1.31 \times 10^{28}\) for Sr and Xe, respectively. The extremely high value of recoils in the fuel results in the creation of considerable high internal stresses, enhances crack formation, and causes cracking of the pellets. These processes as well as the release of gaseous fission products in the primary cycle of PWR should be considered seriously in handling fuel materials. Post irradiation examination of irradiated fuel rods is a must to understand and avoid fuel damage resulting from fission products. The poisoning effect of the released \(^{135}\)Xe requires sufficient excess reactivity at high operating power of reactors.

REFERENCES