Review Of The Radiation Effect On The Cladding Of Zirconium Alloy In Nuclear Reactors

Abdulmalik Mohammed AlShareef
Department of Nuclear Engineering | King Abdulaziz University
E-mail address: *Abdulmalikalshareef1993@gmail.com*

Abstract – *In a nuclear reactor, one of the most important parts is the cladding, which is used to cover the fuel rod. The cladding will face many factors that impact the mechanical structure of the materials while in operation due to its position between the reactor coolant and the nuclear fuel. This cladding should be made of highly corrosion-resistant material with low thermal neutron absorbance.*
The zirconium alloy is the preferred material for use as cladding in nuclear reactors. As previously stated, the zirconium alloy is highly corrosion-resistant, and a thermal neutron absorption cross-section is estimated to be 0.18 x 10-24 cm2. In the reactors, we are concerned with three types of zirconium alloys: Zr-Sn-Fe-Ni-Cr alloy (Zircaloy 2), Zr-Sn-Fe-Cr alloy (Zircaloy 4), and Zr-Sn-NbFe-Cr alloy (Zircaloy 2.5).
This material is exposed to pressure from coolant, temperature, and the irradiation process; as well as changes in the crystallization in the microstructure of the alloy. Thus, the zirconium alloy was found to achieve the purpose.
The article reviews the radiation effect on zirconium alloys in nuclear power plants and emphasizes the impact of radiation on the material's mechanical structure. It also explains some phenomena that occur within the cladding during reactor operation and impact the material's quality and life span.

Keywords: *Zircaloy, Radiation Effect, Nuclear Reactors, cladding, hydrogen embrittlement, irradiation growth.*

I. Introduction:

Nuclear reactors produce many radiations that affect materials, one of these materials is zirconium alloys which are used as the cladding for pellets fuel, the main function of the cladding is prevented corrosion of the fuel by the coolant and prevent the fission products release out of the cladding, thus we must use material having high corrosion resistance, high radiation resistance, and excellent heat exchange, from fuel rod to coolant. [1]
Therefore, used zirconium alloys as cladding and structure of core in light and heavy water nuclear reactor because have a good corrosion resistance between 280–350°C (PWR or BWR) and have a low macroscopic cross-section for thermal neutrons but with absent hafnium, cause hafnium has high absorption cross-section for neutrons.
Consequently, the zirconium alloy used in reactor components (431 light or heavy water reactors in operation out of 448 nuclear reactors in operation in 2017).
Also, the zirconium alloy is included in the structure of guide tubes in a pressurized water reactor (PWR), and cladding tubes and channel boxes in the boiling water reactor (BWR). [2] [3]
Furthermore, these advantageous properties. Zirconium cladding exhibits dimensional instabilities in a nuclear reactor exposed to high-temperature irradiation due to irradiation growth, irradiation creep, and hydrogen embrittlement.

The reactor's initial operation causes rapid growth, which then gradually slows. The initial growth stage is characterized by an increase in the intensity of the dislocation line, which eventually reaches saturation during the slow growth stage.

Also, large safety margins have been built into the design for creeping, and during normal reactor operation, components are unlikely to fail due to creeping rupture. [4]

In this paper will review some properties of zirconium alloy and discuss the radiation effect on zirconium alloy, also discuss hydrogen embrittlement, irradiation growth, the effect of fission products on the alloy.

I.A Neutron–zirconium interaction:

Zirconium alloy has a low cross-section of thermal neutrons and is corrosion-resistant as well. If the atomic transmission threshold is equal to 1 Mev. This means that if the neutron's energy is below the threshold, the effect on the atom only increases the atomic vibration capacity, resulting in the heating of the atom without transmission. If the energy transferred exceeds the threshold value, the atom can be transmitted from its position and is referred to as PKA. Following its transition, high energy can be transferred to the other atoms of the alloy along its path, resulting in further displacement within the material. [5]

It uses fuel cladding in structural components of light nuclear reactor cores and heavy water. On the other side, these alloys show instability in their dimensions when operating due to the high radiation conditions and the heat present at the core of the nuclear reactor, resulting in phenomena such as irradiation growth, increased irradiation creep, and hydrogen embrittlement. [6] [7] [8]

Zirconium alloys are usually exposed to a large reaction with fast neutrons with an energy estimated at 1−2 MeV, resulting in damage to their mechanical properties due to the elastic interaction between fast neutrons and alloy atoms. This collision displaces the atoms and changes their locations, creating a significant change in their order, making them vulnerable to defect formation. [9]

During this stage, most defects are sent to the sink, such as loops, grain boundaries, and cavities. Other defects combine with each other to form so-called clusters. [10]

I.B Displacement energy in zirconium

To Know the displacement energy threshold for neutrons, M. Griffiths has made direct measurements using the HVEM at Birmingham University. The mean energy was 25 eV for irradiation at about 300 K; this energy is dependent on the crystallographic orientation.

Table 1 Displacement energy in zirconium

	Crystallographic Orientation			
	[0001]	[1123]	[1010]	[1120]
Threshold T_d (ev)	**25.5 +- 0.5**	**24.0 +- 0.5**	**24.5 +- 1.0**	**27.5 +- 1.0**

In some cases, damage first appeared near the surface of the material indicating that displacement energies were affected by surface effects. This reduction in threshold energies is caused by the indirect effects of oxygen due to oxides on the surface. [11]

By using the method of Torrens and Robinson formula to calculate the number of atoms displaced. Assume that the energy to generate atomic displacements by elastic collisions is 21.7 keV PKA and using displacement energy of Ed = 40 eV and the displacement efficiency is 0.8. the number of atoms displaced will be 217 atoms according to the equation below.

$$np = \frac{0.8\,(T)}{2(Ed)} \qquad (1)$$

Where n_p is number of atoms displaced, and T is energy to generate atomic displacements by elastic collisions and E_d is displacement energy threshold. [12]

II. Background:

There are three types of zirconium alloys used in the nuclear power plant as cladding, Zr-Sn-Fe-Ni-Cr alloy (Zircaloy 2), Zr-Sn-Fe-Cr alloy (Zircaloy 4), and Zr- Sn-Nb-Fe-Cr (Zircaloy 2.5).

Zirconium alloys used in many locations in reactor power plant from cladding to nuclear waste disposal components, zircaloy-2 originally developed to improve the corrosion resistance of zr-1 to achieve this target should adding amounts of Cr, Ni, and Fe.

Therefore, the zr-2 contains Zr, Sn (1.5%), Fe (0.15%), Cr (0.1%), and NI (0.05%), used in BWR as fuel cladding and calandria tubing in Canadian Deuterium Uranium (CANDU). [13]

Furthermore, the difference between zircaloy-2 and zircaloy-4 is the concentration of iron because removed the nickel and increased the iron content will contribute to reducing hydrogen absorption in the reactor. The zircaloy-4 composition is containing Zr, Sn (1.5%), Fe (0.2%), tin (1.3%), and Cr (0.1%), and it's used as cladding in Pressurized Water Reactors (PWR) and CANDU reactors. [14]

Zr-2.5Nb alloy contains niobium to increase strength and used in pressure tubes in CANDU reactors, the table below summarizes the chemical compositions of some types of zirconium alloys.

Table 2 Typical elemental compositions of Zircaloys [16]

Ele	Sn	Fe	Cr	Ni	O	Hf	Nb
Zr-2	1.2-1.7	0.07-0.20	0.05-0.15	0.03-0.08	0.1	<100 PPM	-
Zr-4	1.2-1.7	0.18-0.24	0.07-0.13	-	0.1	<100 PPM	-
Zirlo	0-0.99	0.11	-	-	0.1	<40 PPM	0.98
Zr-2.5	1	0.1	-	-	0.1	-	1

Now, the studies are beginning to improve the alloys used in the fourth-generation reactors, consequently, the zirconium alloy isn't a candidate for use in the fourth-gen reactor due to higher fuel cladding temperatures which reach to 1000ºC, Therefore, it is possible to replace zirconium with stainless steel. As shown in table 3. [15]

Table 3 Types of fourth-generation reactors with the alloys expected to be used as cladding [15]

Reactor	Temperature	Cladding
GFR	850ºC, Fast	Ceramic
LFR	550 ºC and 800 ºC, Fast	High – Si F-M , Ceramic , or refractory alloy
MSR	Thermal 700-800 ºC	Not applicable
SFR	Fast, 520ºC	F-M (HT9 or ODs)
SCWR	Fast, 550 ºC	F-M (12Cr,9Cr,etc), (Fe-35Ni-25Cr-0.3Ti) incoloy 800,ODS incoloy 690&625
VHTR	Thermal 1000 ºC	ZrC coating and surrounding graphite

In the experiment to study irradiation growth at a reactor temperature of 555 K and a fluence up to 3.5 1025 n/m2, and the case of annealed zircaloy- 4 with 75% the irradiation growth linear increase with fluence. [16]

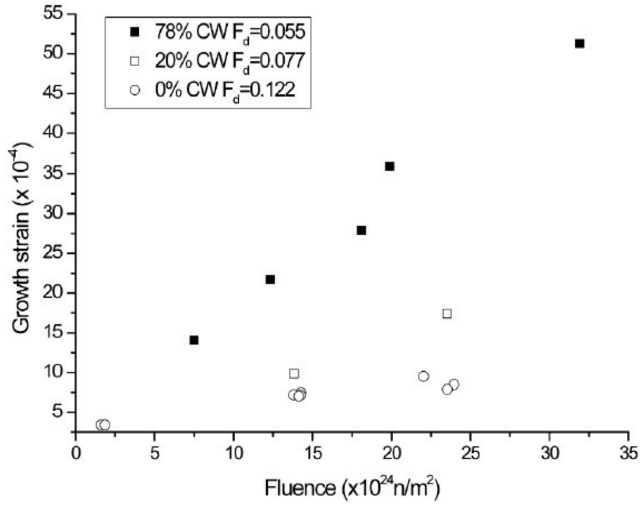

Figure 1 Irradiation Growth at 555 K in Cold Worked Zircaloy-4 [16]

In another experiment, regardless of the irradiation, there is another factor that affects the growth, according to fig.3 the cold worked is an impact on growth. The growth rate increases in cold-worked compare with annealed in zirconium-2 during neutrons irradiation, for example, neutrons irradiation with 60X1024nm–2 and 353K, the irradiation growth in annealed case is 5X104 and for 25% cold-worked is 20X104. Also, some impurity affects the growth phenomena, for example, polycrystalline RXA zirconium alloys at elevated temperature has been growth rate higher than pure zirconium alloy, in another hand, niobium impurities reduce a growth rate in the alloy, for this reason, it's used with zirconium-2.5. [17]

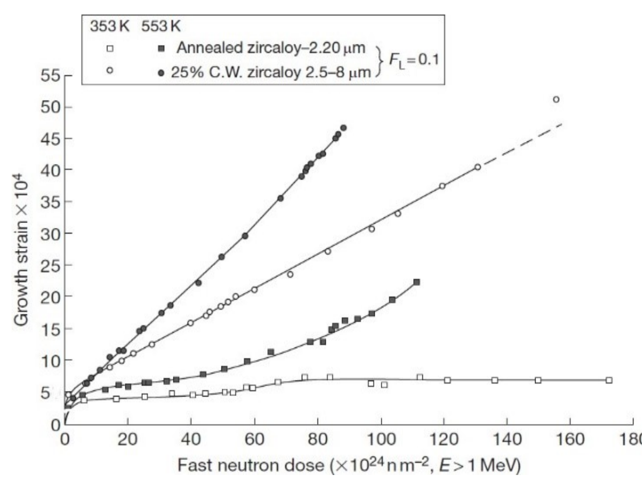

Figure 2 Irradiation growth in annealed and 25% cold-worked Zircaloy-2 at 353 and 553 K [17]

The results of experiments conducted on zirconium alloy showed that the iron concentration in the alloy plays an important role in the impact of the irradiation growth, whereas, Fe content reduced irradiation growth for Fe concentrations higher than 1000 wtppm for (Zr-1Sn>Zr-1Nb>Zr-1Nb-1Sn alloys),and (Zr-1Sn>Zr- 1Nb-1Sn>Zr-1Nb alloys) for iron concentrations lower than 1000 wtppm, Also, The concentration of H will be increase irradiation growth . [18]

III. Radiation effect on zirconium alloys

III.A. hydrogen embrittlement:

Hydrogen embrittlement is one of the issues that engineers face when using zirconium alloys. This phenomenon causes a significant reduction in the alloy's fracture toughness as it begins to appear in service when the reactor's fuel is burned. In fact, hydrogen is produced as a result of the oxidation of zirconium in the reactor by cooling water. It then spreads within the substance, forming layers of hydrides when its solubility is exceeded. [19] In the presence of concentration grades, temperature, and stress, hydrogen spreads in the solid solution. For example, hydrogen spreads towards cooler areas in the presence of a thermal gradient or towards higher hydrostatic stress areas in the presence of a stress gradient. [20] When a coolant (light water or heavy water) interacts with zirconium alloy is produced an oxidized layer and release two molecules of hydrogen, according to the below equation.

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2. \qquad (2)$$

Consequently, the existence of hydrides in zirconium alloy contributes to initial crack formation and subsequent crack propagation, also the concentration of hydrides may reduce ultimate tensile strength in zirconium alloy, this crack called delayed hydride cracking (DHC). [21] This mechanism enables the formation of the crack at some stages, such as crack stress-directed diffusion, hydride formation, and fracture. [22]

The most serious problem is hydrogen embrittlement, which happens when a crack along the rod axis occurs. During nuclear reactor operation, the rod was exposed to external pressure from coolant water and pressure from helium gas, which is came from the fission product. Furthermore, the stress from pellet-cladding mechanical interaction. [23]

A.G. Varias concluded in an experiment that stress was present in a hydrogen embrittlement near the crack. This effect is due to hydrogen's chemical potential and final solid solubility under hydrostatic stress. Away from the crack tip, heat transport is more important. [24]

In the experiment, used The High-Flux Advanced Neutron Application Reactor (HANARO) and chosen two a sample of cladding tubes 250 ppm-H and 500 ppm- H zirconium-4, also the cladding tubes were cut shape make 5-mm width ring, as shown in figure below.

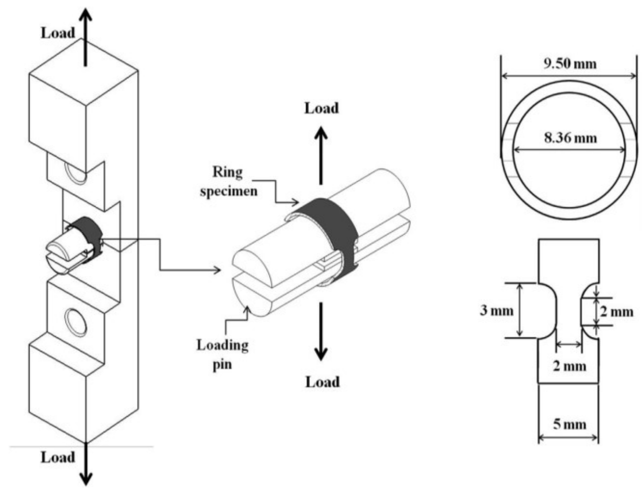

Figure 3 shows a schematic of the ring specimen

Irradiation the sample with 1,392 h at 380 oC at the HANRO Central Hole (CT), with neutrons fluence of 7.5X1024n/m2, after the irradiation process, the sample were heated with the heating rate of 2.0 oC/min.

The non-irradiated and irradiated samples were heated from 25 oC to 200 oC and 400 oC, we can be seen the non-irradiation sample produce circumferential hydrides, while the irradiated sample generated fractions of radial hydrides (see figure 4), therefore, could say the neutron irradiation in the HANARO at 380oC cause recrystallization in the microstructure of the zir-hydride alloy.

Consequently, under elevated temperatures, the hydrogen redistributes easily, furthermore, recrystallization caused a change in the mechanical property of a material and make it brittle, and under tensile hoop stress will form microcrack on the hydrides and may microcrack spread along in the hydrides. [25]

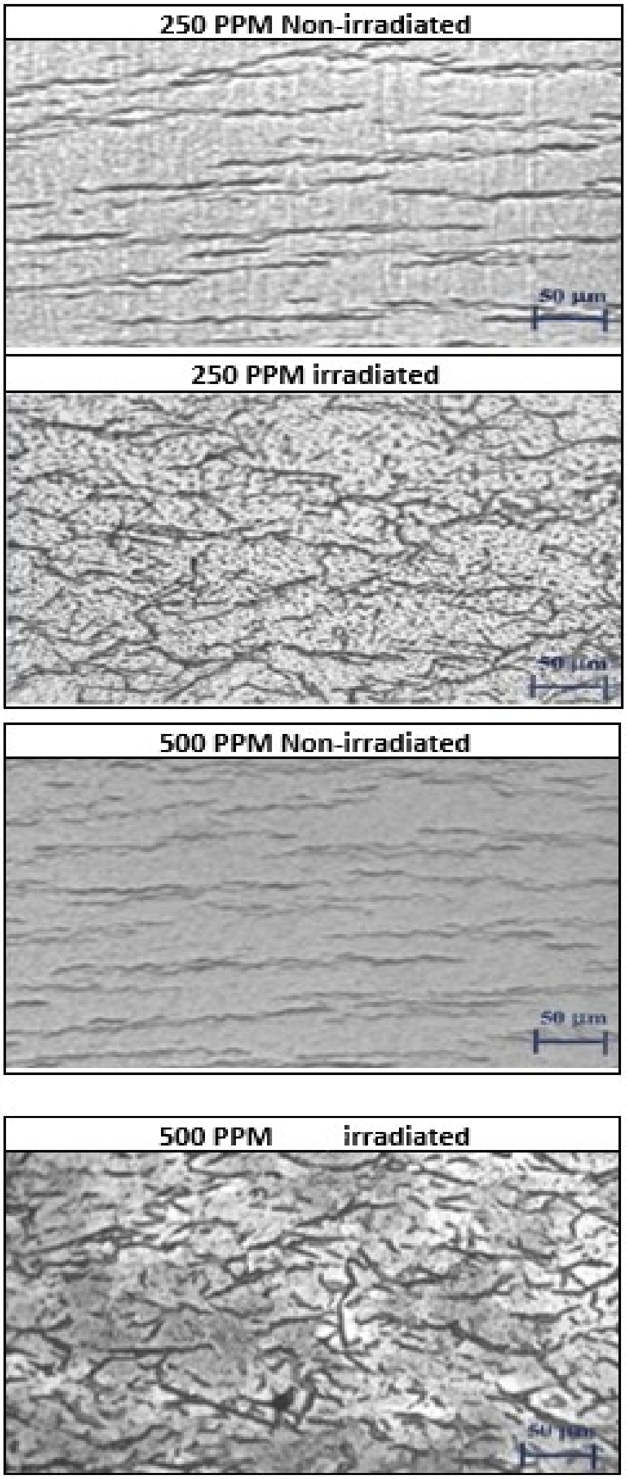

Figure 4 Optical micrographs at the heat-up temperature of 200C

III.B. Irradiation growth:

The most important factor in nuclear power plants is component size stability. In boiling water reactors (BWR) and pressurized water reactors (PWR) fuel assembly will distortion it will affect the movement of control rods and will be complicated to control burning the fuel. [26] In the same context, Fast Breeder Reactors, for example, alloys containing the elements Fe and Ni are affected by radiation and begin with growth, which means that their dimensions will change due to stress and their size will change due to radiation. Zirconium alloys do not swell in water reactors, but their dimensions change as a result of the radiation growth process. Each atom is removed from its normal position at least 20 times during the element's life in the reactor. The mechanical and physical properties of the materials used influence this displacement. [27]

Radiation-induced growth is characterized by dimensional variability, i.e., expansion in one direction and contraction in another. The growth phenomenon, unlike swelling and creep, is not accompanied by increased size or stress. However, even in the absence of stress and volume expansion, growth is a complex phenomenon that is dependent on a number of fundamental criteria. As a result, we had to take into account numerous physical parameters and metal conditions (texture, grain size, and so on).

Radiation growth is affected by several factors (texture, grain size, dislocation density, alloy element, heat treatment, cold work, fluence, and temperature). The source of radiation growth is Dislocation loops, which are created by neutron radiation in single-crystal zirconium. Dislocation loops are classified into two types: a or c-dislocation. [28]

The radiation growth of Zr is related to the interaction between defects and dislocation loops. A large number of defects, vacancies, and interstitials will be created during the first stage. Therefore, the migration energy of interstitials is different and lower than that of vacancies. This difference in vacancies and interstitials induces a tendency towards a-dislocation and loops on the prismatic plane. Thus, early expansion along a-directions. [29]

In Low Dpa stage, the interstitials are absorbed by dislocation-type sinks till achieved the quasi-equilibrium in atomic scale. Then, the ratio of defects to interstitial loops will be decrease or close to zero. Therefore, irradiation growth plateau will be formed. [30]

III.B.A Single crystal zirconium:

In a single crystal case, the irradiation growth starts with dislocation and dislocation loops. In the breakaway regions, the self-interstitial atom (SIA) is responsible for creating the elongation along the a-axis. Then, the vacancy loop on the basal plane is responsible for creating along the c-axis.

In the first stage, the self-interstitial atom will be a sink because the diffusion coefficient is greater than the vacancy, i.e., $DiCi >> DvCv$ (where $Di >> Dv$). Then, the vacancy concentration will increase. In addition, the self-interstitial atom will recombine with a vacancy, and the concentration of the vacancy will be greater than SIA $DiCi = DvCv$ (where $Cv >> Ci$), both SIA and vacancies form clusters. Then, expansion in the a-axis and shrinkage in the c-axis.

Finally, a c-dislocation loop is formed on the basal plane, and the dislocation loop absorbs SIA. Then, the SIA concentration will be increased again by the vacancy loop. And the irradiation growth increases with the dose. [31]

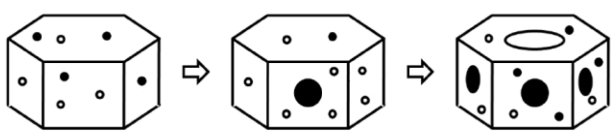

Figure 5 Single crystal zirconium alloy

III.C. The effect of fission products:

Today, the most commercial reactor operation from Light Water Reactor (LWR) type, these types of reactors used UO2 as fuel. The nuclear fuel is manufactured with the gap between the fuel rod and cladding to provide space for thermal expansion and swelling during the operation state (See Figure 6). The cladding suffers from different pressures, coolant pressure, and gas pressure, these pressures make a gap between pellet and clad close down and causes protrusions (need between one year to 3 years and its dependence on the type of fuel). [32]

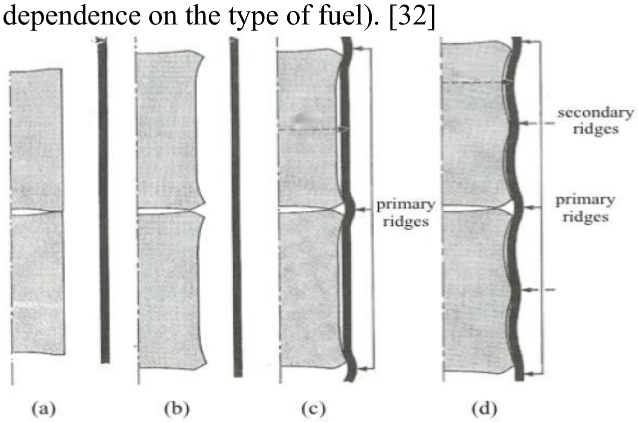

Figure 6 The change space between the cladding and the fuel rod during operation reactor

The fission products release as noble gases (xenon and krypton) approximation 30% of fission product release as gas, which is dissolve poorly in the UO2.

That gases migrate and form bubbles inside grain or in the grain boundary of UO2, after this, these bubbles will escape out of fuel and Settle down in the gap between cladding and fuel.

These bubbles reduce the thermal conductivity between cladding and fuel, furthermore, increase the temperature center of fuel (as shown in figure 7), also increase the pressure on the cladding.

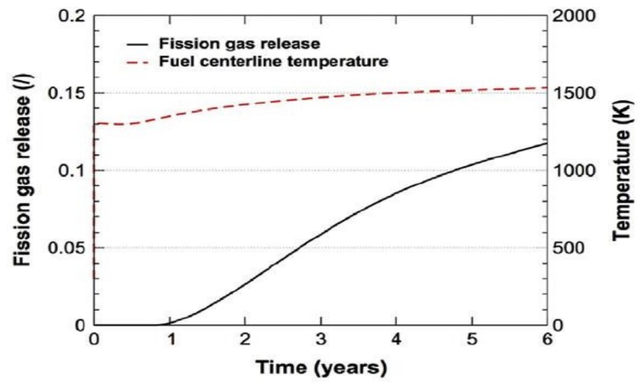

Figure 7 Effect of fission gas release on fuel centerline temperature

In the normal state, the fuel pellet prevents fission products gas release to the outside of the pellet as possible, but may the gas release with three stages:

The gas atoms transport through the grain (see figure 8A). Accumulate gas atoms and create bubbles in the face of grain (see figure 8B). Finally, these bubbles migrate to the grain edge, and release out of the surface (see figure 8C). [33]

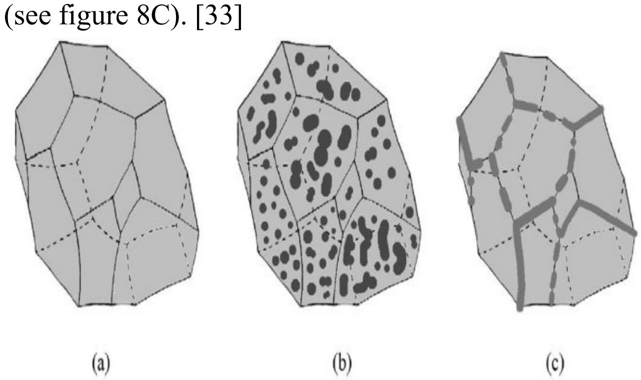

Figure 8 The three stages of release gas fission produce

Another phenomenon that arises due to the release of fission gases that we should concern about it is swelling, the Swelling is a 3-dimensional change in the material (fuel in this case) during the irradiation process, the

swelling appears in the fuel due to gas fission product such as (Xenon, Krypton) because this gas creates bubbles in the microstructure of fuel, thus impact significantly fuel swelling.
These bubbles migrate with the presence of temperature and neutron radiation to creates more void with contributions from some vacancy. After the formation of swelling, radiation, and temperature will create more inside pressure on the cladding and over time it may happen failure. Also, the fission products cause cracking in fuel rod and cladding due to temperature gradient during normal or abnormal operation of nuclear power plant, these cracks its increases the surface of fuel, thus it caused a change in thermal conductivity. As shown in figure 9, these cracks are appeared horizontal (z axis or X- axis) of fuel, the orientations of crack depend on thermal stresses between center temperature (1000 ºC) and outside temperature (400-500C) of the pellet. [32]

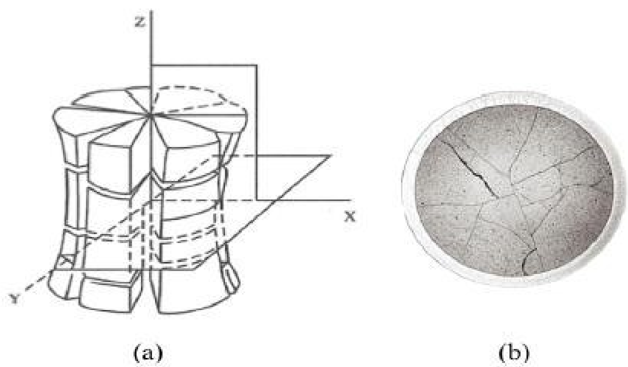

Figure 9 Fuel pellet cracked by the thermal gradient

IV. Conclusions

This paper has highlighted of Zirconium alloys and how it used as the cladding of fuel rod in nuclear reactors, the zircaloy used because it has high corrosion resistance and good heat exchange.
There are many types of Zirconium alloy, but we are concerned with three types of Zr-Sn-Fe-Ni-Cr alloy (Zircaloy 2), Zr-Sn-Fe-Cr alloy (Zircaloy 4), and Zr-Sn-Nb- Fe-Cr (Zircaloy 2.5) in the reactors. This material is exposed to pressure from coolant, temperature, and irradiation process; thus, we found the zirconium alloy achieve the purpose. One of the most important issues in using zirconium alloy as cladding is hydrogen embrittlement, the zirconium alloy interacts with water and produce H + ZO2 and produce delayed hydride cracking (DHC).
Also, the irradiation process makes recrystallization in the microstructure of the zirconium-hydride alloy.
Furthermore, I discuss the irradiation growth in the alloy, according to results in the review, when neutron radiation increased the growth also increase linearity.
The effect of fission products on zirconium alloy is significant, the fission product 30% as gas (xenon and krypton), these gasses create bubbles and accumulate to create a void, these voids caused pressure to the cladding, in addition to thermal pressure, water pressure and irradiation. The other effect of fission products is temperature gradient these phenomena cause cracking in fuel rod and cladding.
Now, studies are beginning to improve the alloys used in the fourth-generation reactors, the zirconium alloy isn't a candidate due to higher fuel cladding temperatures, which reach 1000ºC, Therefore, it is possible to replace zirconium with stainless steel.

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