Studying the Effect of Fuel Elements Structural Materials Corrosion on their Operating Life

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Abstract. The paper deals with experimental data regarding the effect of internal and external factors on the corrosion decay of Zr1Nb alloy fuel elements. Based on the analysis results, losses of zirconium that transfers to oxide or coolant as per the fuel element wall weight and thickness as well as economic losses from their corrosion decay have been theoretically calculated. To avoid a state-level emergency occurrence, an increase in the fuel element wall thickness up to 660 μm is proposed, which can increase the operating life under the conditions of trouble-free coolant mass transfer hydrodynamic mode.

Introduction

As of today, there are four nuclear power plants are operating in Ukraine, where operate 15 VVER type (water-water energetic reactor) units with total capacity of 13,880 MW. Over the past decade, the total annual production of electric power in Ukraine increased, the share of energy production at nuclear power plants increased as well. In other words, atomic power has turned into the main stabilizing factor of the country's energy system. All these facts as well as the analysis of the country's energy needs and capability to meet them is evidence of expedience and necessity of the atomic energetics development in Ukraine [1,2]. Choice of this exact way will correspond with the global trend as well.

The main direction of efforts of the global atomic energy scientific community is ensuring the safety of nuclear power plants operation. One of the most relevant directions of ensuring the nuclear power plants operation safety is studying the fuel elements corrosion processes in order to solve the issue of their operating life increasing. The physical and chemical peculiarities of the fuel elements operation are of the holistic character and depend on the zirconium alloys chemical composition in the body of fuel elements, coolant temperature and chemical composition (water steam) and coolant hydrodynamic motion through the fuel elements components. Therefore, researching the kinetics and mechanisms of fuel elements corrosion is a relevant area of research.

Main Part

One of the primary issues that designers or production engineers consider when developing fuel elements is the issue of materials and fuel assemblies corrosion resistance in the working fluid [3].

Zirconium, due to its low thermal-neutron capture cross-sections and other properties, is an excellent structure material for the atomics. However, it has some disadvantages. One of them is its high chemical activity when interacting with other elements. It easily reacts with oxygen, nitrogen, hydrogen, water with halogens: fluorine, chlorine, iodine as well as caesium, tellurium and other elements that can accumulate inside fuel elements [4].

In its pure form, it does not meet the requirements as per the corrosion resistance and mechanical properties as a structural material of fuel elements. Therefore, there was an issue on the creation of an alloy without this disadvantage occurred. One of these alloys is Zr1Nb – zircaloy [5].

At the time being, researchers from many countries work over the improvement of the existing zirconium alloys and creation of new ones that capable to operate reliably under the reactor conditions at temperatures of 300-350°C in the high-parameters water. The currently studied for use in Ukraine alloy on the basis of calciothermic zirconium contains increased amounts of oxygen (up to 0.16 wt %) [5].

Zirconium is highly oxidized by air at a temperature of 300- 400°C and is quite stable in water. It is appropriate for fabrication of the fuel elements protective shells cooled by water or rare metals (sodium, potassium) [6]. Unalloyed zirconium losses its stability in the water at a temperature of 300-320°C. Therefore, its stability is highly dependent on temperature. Adding 1.5% of tin, 0.12% of iron, 0.05% of nickel and 0.1% of chromium (zircaloy 2) to zirconium the oxide film is not destructed. The zircaloy 2 alloy is stable in water and steam at high temperatures [7].

Autoclave test results of plates samples rolled of Zr1Nb triple furnace-charge cast alloy at 350 ^{0}C, show a high corrosion resistance during the accelerated test.

The effect of the medium pH acidity on the Zr1Nb and Zr-Fe-Sn-Nb alloys corrosion compared to the pure zirconium was studied in the paper. The research results showed that to reduce the corrosion processes one has to maintain the pH level within 7±0.2[8].

Besides researching the corrosion processes under normal operating conditions, tests that simulated emergencies upon large breaks and 1st circuit pipe rupture of VVER-1000 reactor with the active zone dewatering, pressure drop down to 0.1 MPa in reactor and rise of fuel elements temperature up to 1200 °C have been conducted. Resulting from a hypothetical violation of operating conditions, an increase in the fuel elements shells temperatures from the operating ones of 300 - 350 up to 600 - 800 ºC is possible [9]. The oxide films structure studies revealed a significant unevenness of their thickness after testing at all temperatures . After holding at the temperatures of 500 and 600 °C they are integral, while after holding at the temperatures of 660 i 770 °C, there are well visible cracks in the oxides layers that spread to the deep but not reach the metal due to the protective oxide films building-up in the cracks.

Currently, in many countries that use nuclear power, the behaviour of fuel elements shells materials is widely studied regarding the design accidents temperatures (500 - 600 to 1100 - 1200 °C) and at higher temperatures. The objective of these works is obtaining additional knowledge and increasing the safety of nuclear reactors [10]. At the National Science Center Kharkiv Institute of Physics and Technology, studying the zirconium alloys properties during corrosion testing in water and steam in a wide range of temperatures is also conducted. This paper additionally deals with the characteristics of pipes made of Zr1Nb experimental alloy when oxidation in the design emergencies range of temperatures that can occur due to depressurization of the VVER type reactor facilities first circuit.

However, a disadvantage of the existing studies of the fuel elements corrosion processes is the neglect of the water radiolysis processes taking into account. Under the conditions of high temperatures and radioactivity, water radiolysis proceeds quite quickly. In addition, the process is complicated by the availability of special corrosion inhibitors and radiolysis additives (hydrogen, ammonia, hydrazine), neutrons absorbers (boric acid, gadolinium nitrate) as well as structural materials corrosion products in water. Therefore, water coolant (moderator) is often a diluted, and sometimes concentrated aqueous solution of one or more substances [11].

Thus, the task to be solved is zirconium alloys corrosion processes study, both under the conditions of fuel elements operation (t=300-360 °C) and in an emergency (up to 1200 °C) with account for the hydrolysis processes of the coolant aqueous solution.

The fuel element rod of a nuclear reactor is assembled of uranium oxide pellets. Nuclear fuel is placed in a zirconium alloy protective shell - a material that weakly absorbs thermal neutrons. There is a gap between the fuel rod and shell – a thin gas interlayer filled with chemically neutral and highly thermally conductive helium (refer with Fig.1).

The power of internal heat sources in fuel elements reaches 109 W/m. One has to provide for efficient cooling so that the temperatures level is appropriate for the available materials. The heat transfer coefficient is approximately 30,000 W/(m^{2}C). For uranium oxide that belongs to the

ceramic nuclear fuel type, the temperature can be very high, since the UO_2 melting point is 2,800 °C. Yet the zirconium shells allowable temperature is much lower – up to 400°C [8]. In case of this limit exceedance, destructive corrosion is quickly developed in contact with water
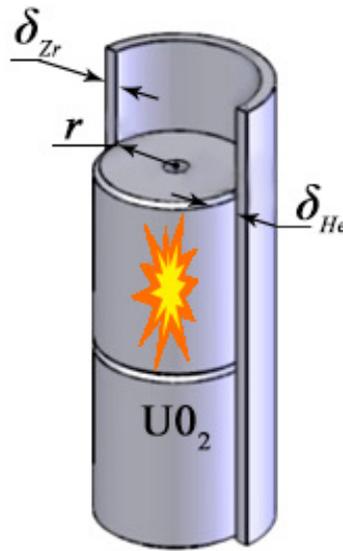

Fig. 1. Fuel element structure

Materials and Research Methods.

Even though the water irradiation process has been studied for decades, as of today, one can say surprisingly little of its decomposition regularities under the operating conditions of nuclear reactors.

First of all, enormous amounts of water reaching many dozens of cubic meters. The liquid phase volume ratio to the evaporation surface exceeds 102 cm instead of the index of 1 cm, which is peculiar to the laboratory conditions. In such a situation, the conditions of gaseous products of radiolysis removal from the liquid phase are not always favourable; they are largely determined by the convective mass exchange intensity at the phases liquid-gas interface. Sometimes establishment of the steady state can take many hours and even days.

Secondly, a wide temperature range, starting from the "room" temperature, when it acts as a moderator in reactor, and ending with about 300 °C, when water is a coolant. In a power reactor, radiolysis often proceeds at a spur increase in temperature (from the channel inlet and at its outlet) and variables power of dose and LET (maximum in the channel centre). The dose intensity varies from 1 to 104 Gy/s.

Thirdly, the proximity of the reactor radiation effective LET to the critical value, at which the radiolysis steady state stops existing. The evident variations in the radiolysis products concentrations follow from this.

Fourthly, availability of special corrosion inhibitors and radiolysis additives (hydrogen, ammonia, hydrazine), neutrons absorbers (boric acid, gadolinium nitrate) as well as structural materials corrosion products in water. Therefore, water coolant (moderator) is often a diluted, and sometimes concentrated aqueous solution of one or more substances [15].

Concerning the hydrogen and oxygen radicals generation in coolant, several chemical effects are considered (reaction 1-7):

$$H_2O \rightarrow H_2O^+ + e^- \tag{1}$$
$$H_2O^+ + H_2O \rightarrow OH + H_3O^+ \tag{2}$$
$$H_2O^+ + e^- \rightarrow H_2O^* \rightarrow H + OH; \tag{3}$$
$$e^- + H_2O \rightarrow H + OH^-; \tag{4}$$
$$H + H \rightarrow H_2; \tag{5}$$
$$H + OH \rightarrow H_2O; \tag{6}$$

$$OH + OH \rightarrow H_2O_2. \tag{7}$$

Studying the processes in diluted aqueous solutions under the effect of X-ray and gamma radiation showed that formation of H_2 and H_2O – the number of molecules formed per 100 eV adsorbed energy – in various substances solutions, except for organic ones in a wide range of their concentration is almost unchanged and equal to 0.4 i 0.7 molecules/100 eV respectively. To clarify these facts, A. Allen admitted that H_2 and H_2O_2 are formed resulting from H atoms and OH radicals recombination. To clarify these facts, A. Allen admitted that H_2 and H_2O_2 are formed resulting from H atoms and OH radicals recombination. To clarify these facts, A. Allen admitted that H_2 and H_2O_2 are formed resulting from H atoms and OH radicals recombination [12].

A number of mechanisms for these products of radiolysis formation has been proposed. Thus, for example, the interaction of excited water molecules leads to the generation of hydrogen and hydrogen peroxide:

$$(H_2O)^* + (H_2O)^* = H_2 + H_2O_2$$

An important role is played by radicals formed during irradiation: $H_2O = H + OH$. Interaction of each of these radicals with each other yields the main products of radiolysis: $2H = H_2$, $2OH = H_2O_2$. No doubt, an important role in water radiolysis is played by the free electron, formed during irradiation: $H_2O_2 = H_2O + e$. This process gives rise to a number of ion-molecular reactions or reactions involving an electron, for example: $H_2O^+ + H_2O = H_3O^+ + OH$, $H_3O^+ + e = H + H_2O$, $H_2O + e = H + OH^-$. Oxygen generation is the consequence of various processes: $OH + HO_2 = H_2O + O_2$, $H_2O_2 + HO_2 = H_2O + OH + O_2$, $2H_2O = H_2O_2 + O_2$

A great help in the water radiolysis products studying is provided by this process material balance equations, which links the radiation and chemical outputs of radiolysis various products:

$$G_H + G_{H2} = G_{OH} + 2G_{H2O2} \text{ *}$$

or a more detailed equation that takes into account the H_2O radical formation:

$$G_H + 2G_{H2} - G_{HO2} = G_{OH} + 2G_{H2O2} + 2G_{HO2}$$

The equations allow determining the amount of some radiolysis products, calculating the amount (output) of other products of this process.

Research Results.

Experimental determination of the radiolysis products nature and amount is conducted using various physical and chemical methods, among which the most important are the pulse radiolysis method and electronic paramagnetic resonance method, which allows free radicals studying; various chemical methods are also widely used, for example, a method based on determining the components ratio in oxidation-reduction systems.

Chemical transformations of substances dissolved in water exposed to irradiation are, first of all, due to the interaction of these substances with the products of water radiolysis. Since when water radiolysis both strong reducing agents and strong oxidizing agents are formed, substances under dissolution can be both reduced and oxidized depending on their oxidation extent. Easily reduced ions (for example, Co^{3+}, Ce^{4+}, Hq^{2+}, Sn^{4+}, CrO^{2-}_4, MnO^-_4) are reduced to 4 lowest oxidation degrees either by H radical or by hydrogen peroxide. In some cases, H_2O_2 can demonstrate its oxidizing action. Thus, taking into account the aqueous solutions pH value, one can to conduct $Co^{2+} = Co^{3+}$ or $Sn^{2+} = Sn^{4+}$ oxidation reactions that are difficult to be carried out by chemical methods.

Naturally, that easily oxidizable substances, when their aqueous solutions radiolysis, evolve to the oxidized form. Thus, halogens are oxidized to molecular halogens:
$$2Cl^- + H_2O_2 + 2H^+ = 2H_2O + Cl_2.$$

The most important characteristics of some inorganic substances aqueous solutions radiolysis process are given in Table 1.

When concentrated aqueous solutions irradiating, the diluted substances chemical transformations are the consequence of not only these products interaction with the water radiolysis products, but also of the irradiation effect itself [13].

Table 1. Radiation and chemical outputs of some inorganic substances aqueous solutions radiolysis process

Radiation object	Concentration, [mol/l]	Radiation type	Products output
Ti^{3+} (0,4 M H_2SO_4)	10^{-3}	X* (50 кеВ)	G (Tl^+) = 1.34
Ce^{4+} (0,4 M H_2SO_4)	10^{-4}	L Co^{60}	G (Cl^{3+}) = 2.39
Sn^{2+} (0,4 M H_2SO_4)	10^{-3}	L Co^{60}	G (Sn^{4+}) = 0.49
NH_4 (pH = 13)	0.1	L Co^{60}	G (No_2^-) = 1.5
NO_3^- (0,4 M H_2SO_4)	10^{-3}	L Co^{60}	G (No_2^-) = 2.0
H_3PO_4 (0,4 M H_2SO_4)	10^{-3}	L Co^{60}	G (PO_3^{3-}) = 3.15
$Cr_2O_7^{2-}$ (0,4 M H_2SO_4)	10^{-4}	L Co^{60}	G (Cr^{3+}) = 0.78
U^{4+} (0,4 M H_2SO_4)	10^{-3}	L Co^{60}	G (UO_2^{2+}) = 1.3
ClO_2^-	10^{-3}	X (50 кеВ)	G (ClO^-) = 1.34 G (Cl^-) = 0.6 G (ClO_2^-) = 1.1 G (ClO_3^-) = 0.2
Fe^{2+} (0,4 M H_2SO_4)	10^{-2}	L Co^{60}	G (Fe^{3+}) = 8.1

The substances used as a coolant in power reactors, a number of specific requirements conditioned by the presence of large specific heat fluxes in the active zone (heat flux density from fuel element to coolant reaches $(1 - 2) \cdot 10^6$ W/m^2 and more, which is significantly greater than in water-wall tubes of modern steam boilers, where a similar value does not exceed $0.5 \cdot 10^6$ W/m^2), the need for increased operational reliability of the reactor equipment, continuity of the coolant physical and chemical properties under the ionizing irradiation, personnel radiation safety, etc., is specified to.

Based on the material balance equation, taking into account the water radiolysis in a nuclear reactor, its products and performance characteristics, theoretical calculation of fuel element wall thickness reducing under normal operating conditions for 460 days was proposed.

To determine the change in the fuel element wall thickness under normal operating conditions, a mathematical calculation based on the experimental and reference data was theoretically carried out.

The following data were used and designation introduced when calculating:
The experimental value of gain:

ZrO_2 V^{Zr}_{corr}= 240 mg/dm^2 for 5500 hours (230 days).
$V^{Zr}_{corr} \rightarrow ZrO_2$ for 1 day =240 mg/dm^2/230 days = 1 mg/dm^2 per day

Calculation of Zr active surface in fuel elements:

$$\sum S_{f.el.} = S_{1f.el.} \cdot N_{f.el.}$$

One fuel element active surface:

$$S_1^{work}{}_{f.el} = 2,6cm \cdot 353cm = 917cm^2$$

Fuel elements active surface:

$$S^{work}{}_{f.el} = 917 \text{cm}^2 \cdot 50856 = 46634952 \text{ cm}^2 = 466349/52 \text{ dm}^2$$

ZrO_2 amount calculation in all fuel elements per day:

$$V_{ZrO2} = S \cdot V^{Zr}{}_{corr} = (466349 \text{ dm}^2 \cdot 1 \text{ mg})/\text{dm}^2 \text{ per day} = 466/349 \text{ mg/day}$$

Transfer to ZrO_2 from all fuel elements per day: 466.352 mg Zr
For further calculation, based on the requests on power plant capacity, 460 days were adopted as the average time cycle of the fuel element complete serving out.
ZrO_2 amount calculation in all fuel elements over 460 days:

$$V_{ZrO2} = S \cdot V^{Zr}{}_{cor} = (466.349 \text{ dm}^2 \cdot 460 \text{ days}) = 214.521 \text{ g } ZrO_2$$

To calculate the pure zirconium losses the following proportion is used:

$M(ZrO_2) = 123$ g/mol
in 123 g (ZrO_2) – 91 g Zr
214521 g (ZrO_2) – X
$X = (214521 \text{ g} \cdot 91 \text{ g})/123 = 158700 \text{ g} = 158.7 \text{ kg Zr}$

Over the period of fuel elements complete serving out in reactor, approximately 158.7 kg of pure zirconium evolves to zirconium oxide.
Calculation of the total reduction in the fuel element wall thickness due to corrosion. As calculated for 460 days.
The total mass of oxidized zirconium over 460 days: $m_{Zr} = 158.7$ kg
Total area and density according to reference guide: $S_{total}{}^{Zr} = 4818836 \text{ cm}^2$, $d_{Zr} = 6.5 \text{ g/cm}^3$;

$$V_{Zr} = m_{Zr}/d_{Zr} = 158700/6,5 = 24415,38 \text{ cm}^3$$

$$\Delta h = V_{Zr} / S_{total}{}^{Zr} = 24415.38 \text{ cm}^3/48188836 \text{ cm}^2 = 0,000506 \text{ cm} = 5,1 \text{ μm}$$

The mass of pure zirconium in its oxide was calculated above: $m_{Zr} = 158,7$ kg
The amount of $Zr(OH)_n$ that passes to the coolant solution in 460 days at a conditional concentration of Zr^{4+} ions = 0.5 g/l.

$$M^{+4}{}_{Zr} = C \cdot V_{H2O} = 0,5 \text{ g/l} \cdot 100 \cdot 10^3 \text{ l} = 50 \text{ kg } Zr^{4+} (Zr^0)$$

Calculation of the aggregate amount of Zr in ZrO_2 in coolant:

$$\sum m = 158,7 \text{ kg} + 50 \text{ kg} = 208,7 \text{ kg}$$

Total fuel elements surface at full load of reactor = 466,349 dm². Based on the zirconium density and its aggregate amount that transfers to the coolant, the total surface area is calculated:

$$V^{surface}{}_{Rz} = \sum m/p = 208,7 \text{ kg} \cdot 103/6,5 \text{ g/cm}^3 = 32108 \text{ cm}^3$$

Calculation of the thickness of zirconium that transferred to the coolant based on the spent zirconium total surface area and all fuel elements total area at a full load of reactor:

$$h^{thickness}{}_{Zr} = V/S = 32108 \text{ cm}^3 / 46634900 \text{ cm}^2 = 0,00069 \text{ cm} = 6,9 \text{ μm} = 7 \text{ μm}$$

According to the reference guide, the fuel element wall: $h_0 = 650$ μm

Calculation of the thickness losses as a percentage: $h^{thickness}_{Zr} = (7 \ \mu m \cdot 100)/650 \ \mu m = 1,08\%$.

The total reduction in the wall thickness at an individual calculation:

$$\sum \Delta h = h^{thickness}_{Zr} + \Delta h$$

$h^{thickness}_{Zr}$ – wall thickness reducing based on m_{ZrO2}; Δh – wall thickness reduce as per the amount of Zr in the coolant

$$\sum \Delta h = 5,1 + 1,6 = 6,7 \ (\mu m)$$

Reducing the wall thickness from the fuel element total thickness: $(6,7 \ \mu m \cdot 100)/650 \ \mu m = 1,03\%$.

Conclusion

Reduce in the fuel element wall, which equals 7 µm that corresponds to losses up to 1% of fuel elements wall thickness, was for the first time calculated based on the experimental data array concerning the nuclear reactor zirconium fuel elements corrosion.

The fuel elements internal corrosion mechanism has been proposed, the theoretical justification of this phenomenon has been performed. The fact that the internal corrosion mechanism can be launched under the condition of steam penetration through the metal crystalline structure imperfection has been established.

Material and financial expenses (zirconium) when fuel elements operation in a nuclear reactor for 460 days that amounted to 208.7 kg of zirconium, which is financially expressed as 2,608,750 UAH have been calculated. These calculations have been carried out with account for zirconium hydroxides transition to the primary coolant.

An increase in the fuel element wall thickens of up to 660 µm is recommended, which will allow operating the reactor upon increasing the operating life for 500-600 days under the conditions of trouble-free coolant mass transfer hydrodynamic mode.

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