

Experimental and Calculation Investigation of Fire and Explosion Safety during Handling RBMK-1000 Spent Fuel

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Introduction

To date the NPP with RBMK-1000 reactors have accumulated a great quantity of spent fuel. The dry storage with its subsequent removal to the centralized storage is considered as the main technology for the RBMK-1000 spent fuel handling.

This solution was proved for the defective spent fuel assemblies, i.e. spent fuel assemblies suitable for the longterm dry storage. The methods of handling defective fuels assemblies are being developed now. One of the ways to solve the problem of handling the defective spent fuel is its reprocessing at the RT-1 plant at Mayak.

In 2010 Rosatom State Corporation initiated the project "Safe Handling of the RBMK Spent Nuclear Fuel. Reprocessing Feasibility Evaluation". This project provides for the preparation for the regular transportations of the defective RBMK spent fuel assemblies for reprocessing.

Transportation of the defective (mainly, leaky) spent fuel assemblies must be performed in the leaktight capsules in the TUK-11 shipping casks. During the temporary storage and transportation of the wet RBMK spent fuel assemblies in TUK-11 cask there is a hazard of accumulation of the radiolysis products, in particular, hydrogen.

The feasibility evaluation of the fire and explosion safety that was made during the successful pilot removal in 2011 showed that handling of the leaktight canisters containing two leaky fuel rods (without the preliminary drying) was admissible for no longer than five months. But this period is too short for the regular transportations. Therefore, the calculation results were verified during the experimental investigations into the accumulation of hydrogen in the capsules containing real leaky RBMK fuel rods.

The purpose of this paper is determine experimentally the kinetics of the hydrogen accumulation in the conditions simulating the transportation of the RBMK-1000 spent fuel assemblies with water found under the leaky fuel rod claddings and compare them with the calculation results of accumulation of radiolytic hydrogen in the leaktight capsules containing RBMK-1000 fuel rods.

1. Subjects of research

The experiments for determination of the hydrogen accumulation kinetics in the leaktight capsules containing the RBMK-1000 leaky fuel rods with water under claddings were carried out using the fuel rods the parameters of which provided the conservative evaluation of the hydrogen accumulation kinetics. The experiments were carried out in the shielded cell conditions.

The RBMK-1000 spent fuel assemblies intended for transportation and reprocessing are enriched to 2.0-2.4 %. With respect to the radiation safety, the cooling period before transportation and reprocessing must be no less than 10 years. With respect to the cost effectiveness of the spent fuel reprocessing, the fuel burnup must not exceed 24 MWd/kgU.

Therefore, four RBMK-1000 spent fuel rods (enrichment 2.4%, burnup 24 MWd/kgU, cooling time 10 years as of the beginning of the experiment) were selected for the experiment. Before placing into the capsule the upper plugs of fuel rods were cut off.

The kinetics of the hydrogen accumulation was determined at the shielding cell temperature (30–40 °C) and at 80 °C that corresponds to the maximum calculated temperature of the fuel claddings during transportation of the RBMK-1000 spent fuel assemblies with the given parameters in TUK-11.

2. Experimental facility and processing of measurement results

The equipment for filling fuel rods with water and simulation of the transportation conditions consisted of the following components:

- capsule for location of the fuel rods;
- auxiliary system for the capsule preparation;
- sampling system.

After operation and storage in the cooling pool the plenum of the leaky fuel rod can be completely filled with water. The capsule with fuel rods (the plenum upward) were previously evacuated and then filled with water along the entire height of the fuel column. Water from the capsule spare volume was drained out under the argon pressure that created an inert atmosphere in the capsule. The fuel rods, including the plenum, were completely filled with water. After sealing the capsule the fuel rods were in an argon atmosphere at a pressure of about 1 bar.

The schematic diagram of the sampling facility is shown in Fig. 1. The volume of the sampling system (including the sampler) is about 100 less than the capsule volume that provides a minimum sampling effect on the processes inside the capsule. Analysis of the sample composition in the gas phase from the capsule was performed out using an MI-1201 mass spectrometer.

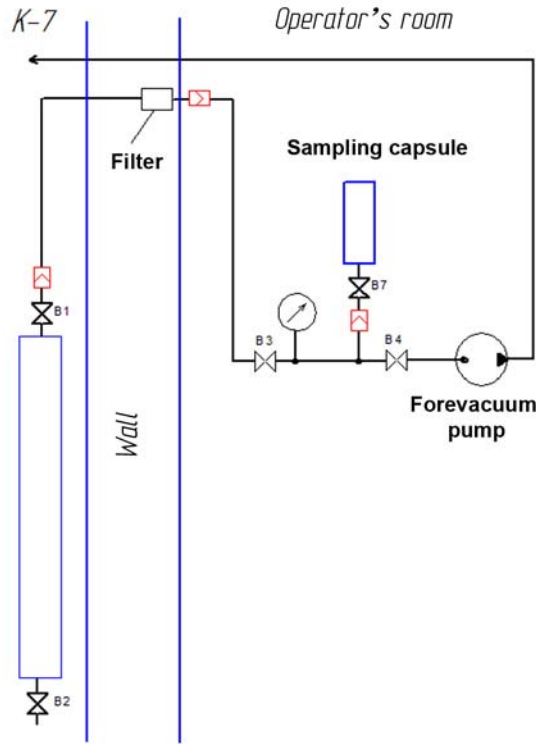


Fig.1. Layout of the gas sampling facility

The amount of hydrogen in the capsule as of the time of sampling is calculated as

$$v_{H_2cap} = \frac{C_{H_2}}{100\%} \cdot \frac{(V_{cap} + V_{ss})}{V_M} \cdot \frac{P}{P_n} \cdot \frac{T_n}{T}, \quad (1)$$

where v_{H_2cap} – the amount of hydrogen in the capsule, mol;

V_{cap} – free volume in the capsule, cm^3 ;

V_{ss} – sampling system volume, cm^3 ;

V_M – molar volume, cm^3/mol ;

C_{H_2} – volume fraction of hydrogen in the sample, %;

P_n – normal pressure, kPa;

T_n – normal temperature, K;

P – sampling pressure, kPa;

T – sampling temperature, K.

The amount of hydrogen generated by the water electrolysis was determined considering the decrease of the amount of hydrogen in the capsule due to sampling by the formula:

$$v_{H_2\Sigma}^i = v_{H_2\Sigma}^{i-1} + \left(v_{H_2cap}^i - v_{H_2cap}^{i-1} \cdot \frac{V_{cap}}{V_{cap} + V_{ss}} \right), \quad (2)$$

where i – sample number.

The relative error of the hydrogen amount in the capsule is practically fully determined by the mass spectrometry error of the gas composition and makes up no more than 20.1 % of the measured value in the concentration range of up to 0.1 vol.% and no more than 10.1 % of the measured value in the concentration range exceeding 0.1vol. %.

An in-cell dry storage testing facility was used (Fig.2) to maintain the temperature of fuel rods at about 80 °C for a long time.

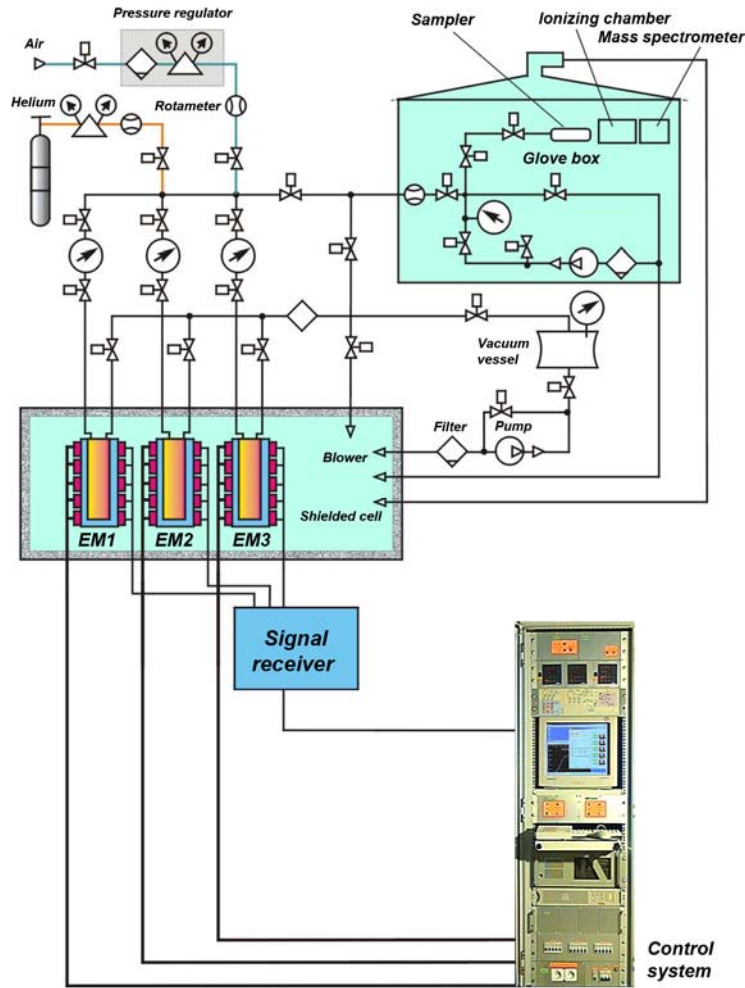


Fig. 2. Block diagram of the dry storage facility

On completion of the investigation of the hydrogen accumulation at the shielded cell temperature of 30–40 °C the capsule cover was opened. Water was removed from all four fuel rods using a syringe fitted with an extension. Then the fuel rods were reloaded from the RBMK spent fuel transportation simulation facility into the dry storage facility capsule. The fuel rods in the capsule were located vertically (plenum upwards). The capsule was tightly closed with a cover.

The temperature control error of the heating elements in the dry storage facility (± 4 °C) causes vibrations inside the capsule that hinders the calculation of the gas phase in the fuel rod capsule. In this connection a method was developed to calculate the quantity of hydrogen accumulated in the capsule based on possibility to calculate the quantity of argon in the capsule at each sampling. The amount of argon as of the moment of the i -th sampling is determined as:

$$V_{\text{Ar cap}}^i = V_{\text{Ar cap}}^{i-1} \cdot \alpha^{i-1}, \quad (3)$$

where $V_{\text{Ar cap}}$ – quantity of argon in capsule, mol;

α^i – sampling coefficient.

The gas fraction (sampling coefficient) remaining in the capsule after the i -th sampling:

$$\alpha^i = \left[1 + \frac{T_{\text{cap}}^i \cdot V_{\text{ss}}}{T_{\text{ss}}^i \cdot V_{\text{cap}}} \right]^{-1}, \quad (4)$$

where T_{cap}^i – gas temperature in capsule, K;

T_{ss}^i – gas temperature in the sampling system, K;

V_{cap} – spare volume in the capsule, cm^3 ($V_{\text{cap}} = 8548.3 \text{ cm}^3$);

V_{ss} – volume of the sampling system, cm^3 ($V_{\text{ss}} = 92.4 \text{ cm}^3$).

The amount of argon in the capsule as of the time of taking sample No.0 is determined as:

$$v_{\text{Ar cap}}^0 = \frac{C_{\text{Ar}}^0}{100\%} \cdot \frac{P^0}{R} \cdot \frac{(V_{\text{cap}} + V_{\text{ss}})^2}{T_{\text{cap}}^0 \cdot V_{\text{cap}} + T_{\text{ss}}^0 \cdot V_{\text{ss}}}, \quad (5)$$

where C_{Ar}^0 –argon volume fraction in sample No. 0, %;

P^0 – sampling pressure, Pa;

T^0 – sampling temperature, K.

The amount of hydrogen in the capsule as of the time of the i -th sampling is determined as:

$$v_{\text{H}_2\text{cap}}^i = \frac{C_{\text{H}_2}^i}{C_{\text{Ar}}^i} \cdot v_{\text{Ar cap}}^i, \quad (6)$$

where $v_{\text{H}_2\text{cap}}$ – hydrogen amount in the capsule, mol;

$v_{\text{Ar cap}}$ – argon amount in the capsule, mol;

C_{H_2} –hydrogen volume fraction in the sample, %;

C_{Ar} –argon volume fraction in the sample,

The amount of hydrogen accumulated as of the time of the i -th sampling is determined as

$$v_{\text{H}_2 \Sigma}^i = v_{\text{H}_2 \Sigma}^{i-1} + (v_{\text{H}_2\text{cap}}^i - v_{\text{H}_2\text{cap}}^{i-1} \cdot \alpha^{i-1}). \quad (7)$$

3. Discussion of results

Kinetics of radiolytic generation of hydrogen in the capsule containing leaky RBMK-1000 fuel rods at 30 and 80° is shown in Fig. 3. The highest hydrogen accumulation rate was obtained in the experiment at 80°C in the absence of water in the fuel rod plenum and was as high as $3.03 \cdot 10^{-6}$ mol/hr for four fuel rods.

The significant difference between the hydrogen generation rate at 30 and 80 °C cannot be explained by only the temperature dependence of the primary hydrogen release. Probably, the great effect is made by the presence or absence of water in the plenum.

In accordance with the Henry's law, hydrogen concentration in the capsule gas volume is directly proportional to its concentration in water found in the leaky fuel rod plenum. Water radiolysis in the plenum is caused only by the fuel gamma radiation (the path of alpha and beta particles in water is ~0.04 and ~0.85 mm, respectively, for a plenum of 170 mm height), i.e. the absorbed dose rate for water in the plenum is several times less than for water in the fuel part of the fuel rod, and concentration of hydrogen in the plenum water will be always lower. The removal of the molecular hydrogen by diffusion to the liquid/gas phase interface and desorption from the water surface into the gas phase is in competition with the back chemical reactions running in water and leading to the recombination of the radiolysis products with formation of water molecules. The higher is the water column, the less is the probability of its recombination in the back reactions, for instance: $\text{H}_2 + \text{OH} \rightarrow \text{H} + \text{H}_2\text{O}$.

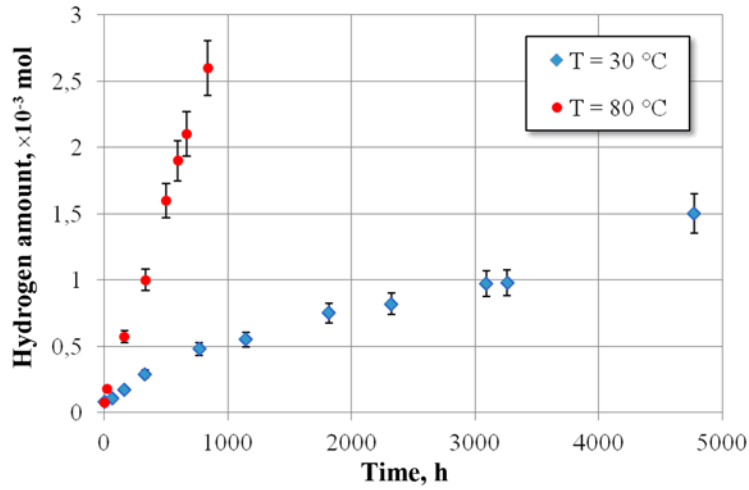


Fig. 3. Kinetics of radiolytic generation of hydrogen in the capsule containing four leaky RBMK-1000 fuel rods at 30 and 80°

The hydrogen accumulation rate inside the leaktight capsule with one fuel rod bundle was calculated based on the following radiation characteristics of the RBMK-1000 fuel assembly [1]:

- initial enrichment in ²³⁵U 2%;
- fuel burnup 24.9 MWd/kgU;
- cooling time 10 years.

The temperature of the fuel rod claddings and gas inside the capsule was taken equal to 80 °C based on the results of the thermal calculation. It was assumed that the inner volume of the leaky fuel rod was fully filled with water. In this case the hydrogen generation is caused by the water vapor radiolysis in the capsule gas volume due to the gamma radiation and from radiolysis of water in the leaky fuel rod due to alpha, beta and gamma radiation.

Evaluation of the hydrogen generation rate from radiolysis of water or water vapor was performed using equation [2], which does not take into account the back reactions:

$$\frac{dn_{H_2}}{dt} = \frac{G_{H_2} \cdot E}{100 \cdot N_A} \quad (8)$$

where dn_{H_2} / dt – hydrogen formation rate, mol/s;

G_{H_2} – primary radiolytic hydrogen release for water or water vapor, molecules/100ev;

N_A – Avogadro's number;

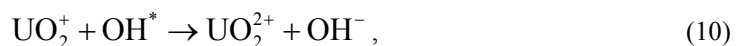
E – energy flux absorbed by energy or water vapor, ev/s.

The calculated hydrogen generation rate only due water radiolysis inside one leaky fuel rod was $3.17 \cdot 10^{-6}$ mol/h that is more than four times exceeds the hydrogen generation rate obtained experimentally at 80 °C ($3.03 \cdot 10^{-6}$ mol/h for a capsule containing four fuel rods). The difference can be obviously explained by the factors described above:

- the dependence of the hydrogen concentration in the gas capsule volume on its concentration in water of the leaky fuel rod plenum (Henry's law);
- the presence of the back reactions running in water and leading to the recombination of the radiolysis products with production of water molecules.

Another important factor effecting the reduction of the radiolytic hydrogen release is related to physical-chemical processes of the uranium dioxide oxidation-reduction at the fuel/water boundary of contact.

Oxidizing agents produced in the process of radiolysis (mainly, peroxide, oxygen and free radicals) react with U(IV) at the fuel boundary forming U(VI). When the stoichiometric matrix achieves the U_3O_7 composition, the uranium matrix is destroyed and dissolved in water [3]. This process does not reduce the probability of the molecular hydrogen recombination by the oxidants. Peroxide makes the main contribution (98-99%) to oxidation of uranium dioxide in the reaction chain



generating hydroxyl radicals.

However, the multiple experiments revealed a significant effect of suppression of the water dissolution process in the presence of the molecular hydrogen [3, 4].

Besides the recombination reactions the other mechanisms are available: hydrogen reduction of the dissolved U(IV) or U(VI) in the solid phase on the fuel surface. It was proposed that the reduction of the solid uranium phase can be catalyzed by nanometric particles of noble metals available in spent fuel [5] by the reaction:



The noble metal particles, called ϵ -particles, consist of the mixture of the metal fission products (Mo, Pd, Ru, Nc и Rh) [6]. The reaction between H_2 and radiolytic peroxide catalyzed by ϵ -particles is also possible [7].

Nevertheless, in any case molecular hydrogen is used for suppression of the fuel dissolution by hydrogen peroxide.

Conclusion

The experimental data were obtained for the radiolytic hydrogen accumulation rate in the leaktight capsule containing the leaky RBMK-1000 fuel rods filled with water. The results were compared with the evaluation of the fire and explosion safety during transportation of the defective RBMK-1000 fuel assemblies in the leaktight capsules without the preliminary drying.

The significant difference between the calculation results and the experimental data can be explained by the ignorance of the following factors:

- the dependence of the hydrogen concentration in the gas phase on its concentration in liquid;
- the presence of the back reactions leading to the recombination of the radiolysis products with production of water molecules.

While ensuring the transportation safety of the leaky RBMK-1000 fuel, in case, if the leaky fuel rod, including its plenum, is fully filled with water, the rate of the hydrogen release into the capsule gas volume is determined by the water radiolysis conditions in the plenum due to gamma radiation and the hydrogen mass transfer conditions between the liquid and gas phases. Alpha and beta radiation of the leaky fuel rod has an insignificant impact on the hydrogen accumulation rate in the capsule gas volume, and the plenum filled with water acts as a “gas lock”. This was demonstrated by about an n-th order difference of the hydrogen release rate in the experiments with and the plenum filled with water and without it given the fact that the dependence of the radiation hydrogen release on the temperature was much lower.

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