THE INFLUENCE OF RADIATION ON STAINLESS STEEL X16H6**МГЮ**

T.N. Agayev, A.A. Garibov, G.Z. Velibekova, U.M. Gasumova
Insitute of Radiation Problems of Azerbaijan NAS.
АЗ 1143 Baku, 9F.Agayev str., E-маил;Улвиййе_аз@рамблер.ру

ABSTRACT

Kinetics of thermal and radiation-thermal processes of oxidation of metal and accumulation of molecular hydrogen in contact of stainless steel with water at various temperatures was investigated. It was established that radiation processes in contact of stainless steel with water lead acceleration radiolytic water decomposition to process.

1. INTRODUCTION

Austenitic stainless steels (s.s.) with contents of Fe-69.5%, Cr-14.7%, Ni-5.48% are widely applied in nuclear reactors as constructional materials and for manufacturing of FE (Fuel Elements) shells, as well as in the field of oil and gas production and transportation [1-12]. Constructional materials of active zones of nuclear reactors at operation in with heat-carriers contact, expose to effect of irradiation and temperature. Today all leading world manufacturers are engaged in development of new updating of old stainless alloys. The basic motivation of these works is increase of their corrosion resistance [1-9].

It was established before that increase of resistance of stainless materials to the influence of ionizing radiation can be achieved by creation of thin oxide film on the surface of metals [11-15].

In the given kinetics of thermal and radiation-thermal processes of oxidation of metal and accumulation of molecular hydrogen in contact of stainless steel with water at various temperatures.

2. TECHNIQUE OF EXPERIMENTS

The study was conducted in static conditions in special quartz ampoules with volume of 1.0-1.1 sm^3. Stainless steel of grade X16H6МГЮ in the form of thin film was taken as object of the study. A contact surface of the samples was determined on the basis of their geometrical sizes and it equaled to 17,23 sm^2/g. In order to eliminate the contribution of organic pollutions of the surface in the process of accumulation of molecular hydrogen samples were preliminarily cleaned by organic solvents - ethanol, acetone, and then washed out by distilled water. Then samples were dried up at temperature 300÷320K in inert gas atmosphere-Ar. The dried up samples were weighed up to $\pm 5 \cdot 10^{-5}$ g and transferred in quartz ampoules. Ampoules with samples were evacuated up to $P \approx 10^{-3}$ Pa first at T=300K, and then at T=673K.

Samples were subjected to preliminary radiation exposure of gamma-rays (J=0,70 Gy/s at various times, then dried up, weighed and transferred to special ampoules for detecting radiation-catalytic activity in the processes of radiolytic decomposition of water. The required amount of water were injected into ampoules with samples by condensation of water vapors from the graduated volume of vacuum-adsorption plants. Accuracy of water injection into ampoules with samples from vacuum-adsorption plant within the studied limit of values of water vapors' density is ±5 %. The temperature while carrying out the experiments was supported with accuracy ±1C. Radiation and radiation-thermal processes were carried out in an isotope source of γ-radiation Co^{60}. Dosimetry of the source was conducted by chemical dosimeters - ferrosulfate, cyclohexanone and methane. Recalculation of the absorbed doze (D) in the studied system was made by comparison of electronic densities [10].

Gas products of the processes were transferred to the special graduated volumes and analyzed by the method of a gas chromatography («GASCHROM 3101»). At radiolytic process at T=300K besides H_2O_2 was also observed in structure of gas products but at thermal radiolysis, basically, H_2 was detected. Corrosion of stainless materials was studied by a gravimetric method. For this purpose the virgin samples of an alloy exposed to experiment which had been dried up in vacuum ($P \approx 10^{-3}$ Pa) weighed to $\pm 5 \cdot 10^{-5}$. Corrosion of materials at the result of radiation-thermal and thermal processes was characterized on an additional weight of samples $\Delta m = m_i - m_0$.

3. RESULTS AND THEIR DISCUSSION

With the purpose of revealing the influence of stainless materials on water radiolysis kinetics of H_2 accumulation at radiolytic decomposition of water in the system « water+reactor stainless steel » at T=300K was investigated. Radiation-chemical yield G (H_2) was 0.44 and 6.5 molec./100 eV. The observable increase of values G (H_2) at H_2O radiolysis in the presence of stainless steel in comparison with the yield of radiolysis of pure water can be explained by the contribution of electrons emitted from metal under the influence of γ-quanta and formation of the additional active centers on the surface essentially influencing the reaction of water decomposition. Thus rate of the process equals to $\Delta w = 3{,}89 \cdot 10^{13}$ molecul/s.

In working conditions of nuclear reactors metal constructional materials are exposed to simultaneous influence of temperature and radiation in contact with the heat-carrier. Kinetics of accumulation of molecular hydrogen at radiation-thermal and thermal processes in contact of stainless steel with water at T=300÷773K and both ρ =5 mg/sm^3 and $D = 1,05Qy/s$ was investigated.

For the purpose of revealing of the contribution of radiation processes to radiation-thermal processes thermal and radiation processes of decomposition water are spent in identical conditions.

In fig. 1 and 2 characteristic kinetic curve of accumulation of molecular hydrogen at radiation-thermal and thermal processes of water decomposition at the presence of metallic stainless steel were given. On the basis of initial, linear sites of experimental kinetic curves values of rates of these processes were determined. Rate of w_R (H_2) radiation-thermal process of accumulation of hydrogen was determined according to the difference of speeds of radiation-thermal and thermal processes w_{RT} (H_2) and w_T (H_2) accordingly.

$$w_K(H_2) = w_{RT}(H_2) - w_T(H_2)$$

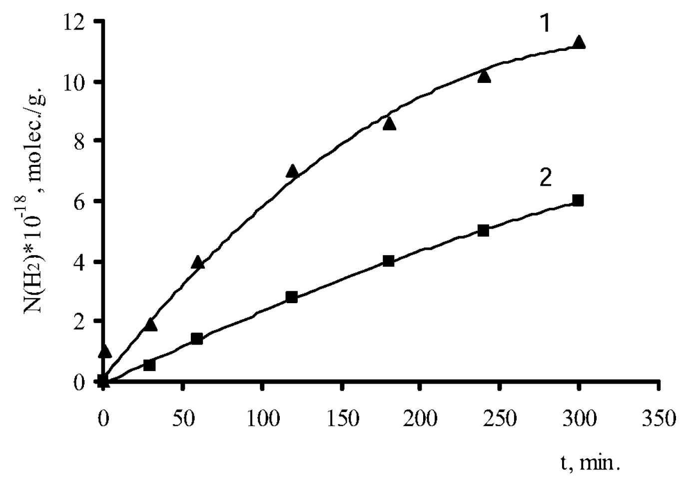

Fig. 1. Kinetic curve accumulation of molecular hydrogen at radiation-thermal (1) and thermal (2) processes of water decomposition at $T = 673K$, $\rho_{H_2O} = 5mg/sm^3$ in the presence of samples of stainless steel, $D = 1,05Qy/s$

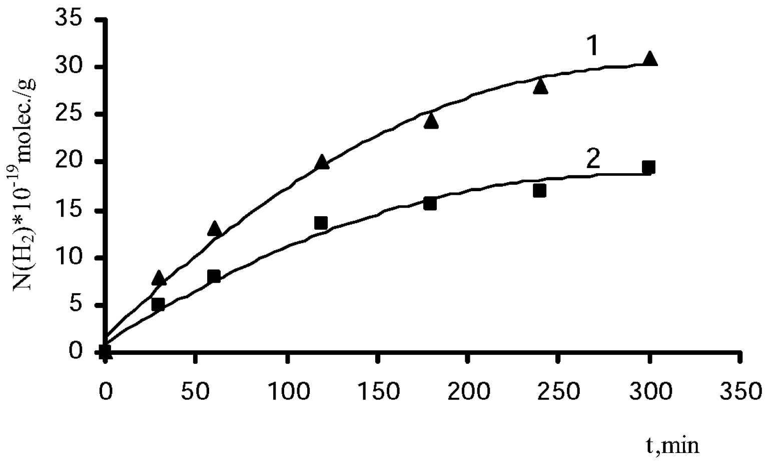

Fig. 2. Kinetic curve accumulation of molecular hydrogen at radiation-thermal (1) and thermal (2) processes of water decomposition at $T = 673K$, $\rho_{H_2O} = 5mg / sm^3$ in the presence of samples of stainless steel, $D = 1,05Qy / s$

In figure 3 dependence of a radiation-chemical yield of molecular hydrogen on temperature is presented.

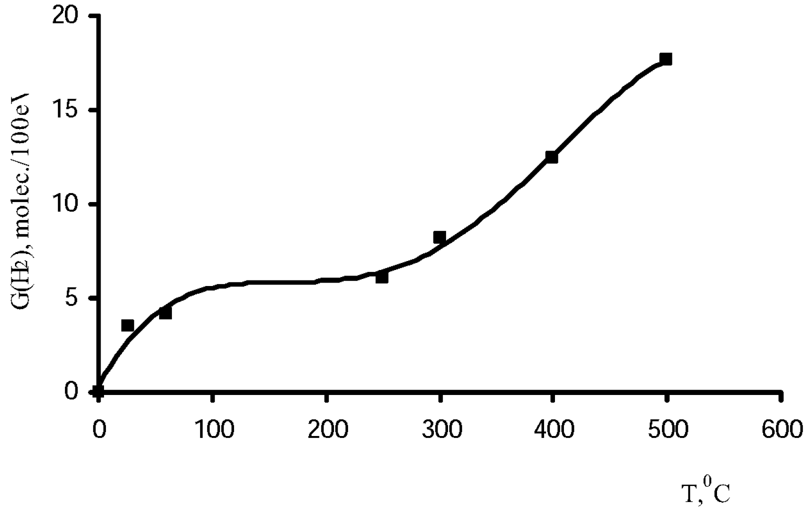

Fig. 3. Dependence of radiation-chemical yield G (H_2) of molecular hydrogen of stainless steel from temperature. $T = 673K$, $P_{H_2O} = 5mg / sm^3$, $D = 1,05Qy / s$

Apparently from fig. 3 on a stainless steel surface at low temperatures the protective oxiditing layer forms and a radiation-chemical yield of hydrogen reaches the values G (H_2) =5-8 molecule/100eV up superficial activity remains stable to value of temperature 523K. Higher values of temperature lead to increase of superficial activity and increase of a radiation-chemical yield of hydrogen to G (H_2) =17,7 molecules/100 eV at T=773K.

During the further temperature increase up to 873 K radiation constituent of radiation-thermal process in comparison with thermal one becomes unnoticeable and $W_T(H_2) \approx W_P(H_2)$.

The kinetics of oxide phase formation of investigated sample surface at the result of thermal and radiation-thermal processes in their contact with water has been studied.

It shows that radiation leads to protective oxidation process rate increase and promotes the beginning of stainless steel destruction oxidation in contact with water.

Thus, on the basis of the obtained results the made following conclusions were made:

At T≥573 K insoluble oxide phase is formed on metal surface that promotes increase of stainless steel samples electrical resistivity.

On the base of obtained results the following conclusions have been made:

Radiation defects in s.s. and influence of secondary electron radiation on contacting medium causes molecular hydrogen yield increase during water radiolysis in contact with s.s.

At the result of radiation-heterogeneous processes in stainless steel contact with water at T≤523 K the formation of stable oxide film highly connected with the surface can be observed.

During radiation-heterogeneous processes in stainless steel contact with water at T≥773 K the connection of oxide film with the surface weakens but at T≥873 K it carries a destructive character.

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