

Radiation-Catalytic Processes in the Nano-SiO₂/H₂O System under the Influence of Gamma Radiation

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Abstract

The amount, rate of formation and radiation-chemical yields of molecular hydrogen obtained from the process of radiolysis under the influence of gamma quanta (⁶⁰Co, P=9.276 rad/s, T=300 K) in the nano-SiO₂/H₂O system with a mass of particle of m=0.2 g and size of silicon particle of d=20÷60 nm by changing of water mass (m=0.01÷0.8 g) were studied. It was found that the rate of formation and radiation-chemical yield of molecular hydrogen determined by increasing the mass of water

- decrease by 10 times for water,
- increase by 8 times for nano-silicon dioxide,
- for common system, increase at the values of water mass of 0.01 gm H₂O 0.2 g, have the maximum at m_{H₂O} =0.2 g and decrease at 0.2 g< m_{H₂O} 0.8 g.

Keywords: Nanoparticles • Radiolysis • Radiation-chemical yield • Compton scattering

Introduction

Recently, due to the rapid development of industry, the demand for energy has increased sharply and the use of nuclear energy is increasing day by day, as traditional methods are both economically and environmentally unfavorable. Transforming nuclear energy into a more affordable form of energy remains one of today's needs. Unique physical, physicochemical and chemical properties of nano-sized materials have been discovered. Therefore, these materials are widely used in all fields of science and technology. One of these applications is the method of obtaining molecular hydrogen from the conversion of water used for the transition from nuclear energy to hydrogen energy with the help of nanoscale catalysts. These methods are preferred in three forms of heterogeneous radiolysis of water. In the first case, water molecules are adsorbed on the nano-particle surface in the form of several monolayers and the process is mainly occurs at surface levels, in the second case in the system of nano-sized catalysts suspended in liquid water at room temperature the process is going on both at the catalyst-water boundary and with electrons emitted from the catalyst surface and released into the water and in the third case, radiation-thermocatalytic processes in these systems under the influence of temperature occur as a sum of two independent thermocatalytic and radiation-catalytic processes.

In the presented work, the amount, rate of formation and radiation-chemical yields of molecular hydrogen obtained from the process of radiolysis under the influence of gamma quanta (⁶⁰Co, P=9.276 rad/s, T=300K) in the nano-SiO₂/H₂O system with a mass of particle of m=0.2 g and size of particle of d=20÷60 nm by changing of

water mass (m=0.01÷0.8 g) were studied. The amount of molecular hydrogen obtained from radiolysis process, its formation rate and radiation-chemical yield are determined in the nano-SiO₂/H₂O system with a mass of m=0.2 g and d=20-60 nm particle size under the effect of gamma irradiation. In systems created by the adsorption of water on the surface of nano-SiO₂ and under the influence of gamma rays, the radiation-chemical yield of molecular hydrogen obtained from the decomposition of water was less than 0.36 molecules/(100 eV). This indicates the surface density of the energy transfer centers on the surface of nano-SiO₂ is very small. As the mass of water increases, the radiation of the nano-SiO₂ emitted from the surface of the nanoparticles in the liquid space between the particles increases, and the radiation of the resulting molecular hydrogen also increases. However, the radiation-chemical yield of molecular hydrogen obtained from the decomposition of water was less than 0.36 molecules/(100 eV) in systems created by the adsorption of water on the surface of nano-SiO₂ irradiated by gamma rays. This means that the surface density of the energy transfer centers on the surface of nano-SiO₂ is very small. When the intergranular space is filled with water, the electrons emitted from the surface of the solid to the liquid phase and the radiation-chemical yield of salvaged electrons in liquid phase increases [1].

Materials and Methods

Experimental part

After heat treatment in the open air at a temperature of T=773K (t=72 hours), the required mass of the SiO₂ (m=0.2 g) is determined

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and then transferred to a heat-treated and cleaned ampoule ($V=5$ ml) under special conditions ($T=773$ K). After 4 hours of thermal treatment ($T=673$ K) of the nano-SiO₂ at vacuum conditions ($P=10$ mm. Hg to 3 mm. Hg) in the ampoule it was cooled, then the required amount of air-purified bidistilled water was added and the ampoule was sealed. The ampoule was irradiated by ⁶⁰Co source with a dose rate of $P=9.276$ rad/s at room temperature. The absorbed dose rate was determined using ferrosulfate and methane methods and in a specific research object it was calculated using electron density comparison methods. It was revealed that in the nano-SiO₂/H₂O system, the final molecular products obtained from the radiation-heterogeneous transformation of water are H₂, O₂ and H₂O₂. Since some part of O₂ is captured on the surface and H₂O₂ remains in solution, there are great difficulties in determining their quantities. Therefore, more accurate information on the kinetic regularity of products obtained from the processes of radiation-heterogeneous transformation of water was based on the amount of molecular hydrogen. The amount of molecular hydrogen obtained from the radiation-heterogeneous conversion of water in the nano-SiO₂/H₂O system was analyzed on the "Agilent-7890" chromatograph. In parallel, a modernized "Chvet-102" chromatograph (accuracy 8%-10%) was used to confirm the results. A column with a length of 1 m and an inner diameter of 3 mm was used in the chromatograph "Chvet-102". Activated carbon with a particle size of $d=0.25\div0.6$ mm inside the column and argon gas with a purity of 99.99% on both chromatographs as the gas carrier were used (Figure 1).

Results

Graphs of the dependence of the amount of molecular hydrogen obtained from the radiation-catalytic transformation of water in the nano-SiO₂/H₂O system (particle size of n-SiO₂ of $d=20\div60$ nm and mass of $m(\text{SiO}_2)=0.2$ g) on the duration (dose) of radiation of γ - quanta (⁶⁰Co, $P=9.276$ rad/s, $T=300$ K) by changing the mass of water ($m=0.01$ (1), 0.02 (2), 0.04 (3), 0.08 (4), 0.2 (5), 0.4 (6), 0.8 (7) g) are given.

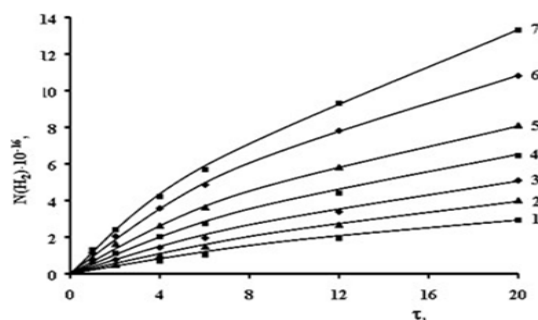


Figure 1. The dependence of the amount of molecular hydrogen obtained from the radiation-catalytic transformation of water in the nano-SiO₂/H₂O system on a duration (dose) of radiation under an influence of gamma-quanta (⁶⁰Co, $P=9.276$ rad/s, $T=300$ K).

Discussion

The rates of formation of molecular hydrogen from the linear parts of the kinetic curves (curve 1-7) obtained from the studied systems (nano-SiO₂/H₂O) were determined for water, nano-silicon dioxide and the general system. Formation rate are determined for Clean water as

$$w_0(H_2) = \frac{N_0(H_2)}{m_{H_2O}t}$$

Water added to nano-SiO₂ as

$$w_{H_2O}(H_2) = \frac{N(H_2)}{m_{H_2O}t}$$

The total nano-SiO₂/H₂O system as

$$w_{tot}(H_2) = \frac{N(H_2)}{m_{tot}t} = \frac{N(H_2)}{(m_{H_2O} + m_{SiO_2})t} = \frac{m_{H_2O}}{m_{H_2O} + m_{SiO_2}} \frac{N(H_2)}{m_{H_2O}t} = \frac{m_{H_2O}}{m_{H_2O} + m_{SiO_2}} w_{H_2O}(H_2)$$

Finally for the nano-SiO₂

$$w_{SiO_2}(H_2) = \frac{\Delta N(H_2)}{m_{SiO_2}t} = \frac{N(H_2) - N_0(H_2)}{m_{SiO_2}t}$$

If we multiply the nominator and denominator of (4) by m_{HO} , make simple transformations, and take into account the expressions (1) and (2), we get the expression (5)

$$w_{SiO_2}(H_2) = \frac{m_{H_2O}}{m_{SiO_2}} \frac{N(H_2) - N_0(H_2)}{m_{SiO_2}t} = \frac{m_{H_2O}}{m_{SiO_2}} \frac{N(H_2) - N_0(H_2)}{m_{H_2O}t} = \frac{m_{H_2O}}{m_{SiO_2}} \left[\frac{N(H_2)}{m_{H_2O}t} - \frac{N_0(H_2)}{m_{H_2O}t} \right] = \frac{m_{H_2O}}{m_{SiO_2}} [w_{H_2O}(H_2) - w_0(H_2)]$$

Here, $N_0(H_2)$ is the amount of molecular hydrogen obtained from the radiolysis of pure water and $N(H_2)$ is the amount of molecular hydrogen obtained from the radiolysis of water in the nano-SiO₂/H₂O system are masses of water, nano-SiO₂ and the total system respectively [2-5]. The table shows the values obtained for the rate of formation of molecular hydrogen from the radiation-catalytic transformation of water in systems created by the addition of water with a mass of $m=0.01$ g, 0.02 g, 0.04 g, 0.08 g, 0.2 g, 0.4 g and 0.8 g on nano-SiO₂ under the influence of γ - quanta. This shows the values obtained for the rate of formation of molecular hydrogen from the radiation-catalytic transformation of water in systems created by the addition of water with a mass of $m=0.01$ g, 0.02 g, 0.04 g, 0.08 g, 0.2 g, 0.4 g and 0.8 g on nano-SiO₂ under the influence of γ - quanta (Table 1).

Table 1. Rates of formation of molecular hydrogen obtained from the radiation-catalytic transformation of water in systems created by the addition of water of the mass $m=0.01$ g, 0.02 g, 0.04 g, 0.08 g, 0.2 g, 0.4 g and 0.8 g to nano-SiO₂ ($m=0.2$ g, $d=20 \div 60$ nm) under the influence of γ - quanta (^{60}Co , $P=9.276$ rad/s, $T=300$ K).

w(H ₂) 10-13, molecules/g × s	mH ₂ O, g						
	0.01	0.02	0.04	0.08	0.2	0.4	0.8
wSiO ₂ (H ₂)	0.23	0.35	0.53	0.83	1.22	1.55	1.8
wH ₂ O (H ₂)	4.55	3.5	2.65	2.1	1.33	0.775	0.45
wtot (H ₂)	0.22	0.32	0.44	0.59	0.665	0.52	0.36

This shows the dependence of the radiation-chemical yield of molecular hydrogen on the mass of water calculated on the basis of these rates ($w_{\text{tot}}(\text{H}_2)$, system (curve 1), water (curve 2) and nano-SiO₂ (curve 3) (Figure 2).

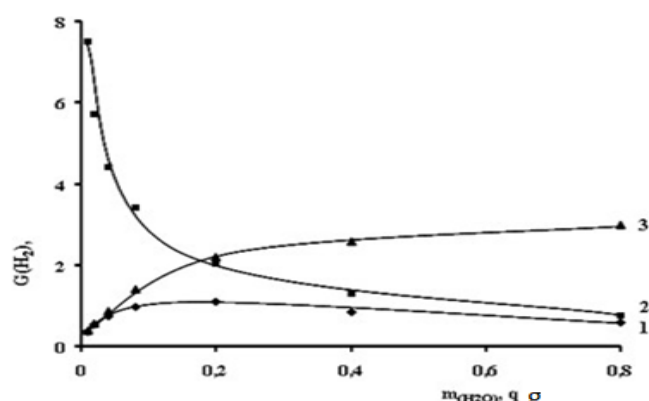
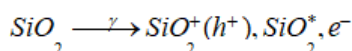
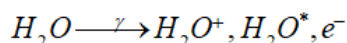


Figure 2. The dependence of the radiation-chemical yield of molecular hydrogen on the mass of water in the nano-SiO₂/H₂O system under an influence of gamma-quanta (^{60}Co , $P=9.276$ rad/s, $T=300$ K).

The results obtained can be explained on the basis of known mechanisms of radiation chemistry. Thus, when γ - quanta with energy $E_\gamma=1.25$ MeV (^{60}Co) pass through SiO₂ and H₂O, Compton scattering occurs mainly in comparison with other processes (various scattering, photoeffect, electron-positron pair formation, photonuclear reaction, etc). Depending on the angle of scattering in Compton scattering, the energies of Compton electrons vary in the range of 0 ÷ 1.02 MeV. Compton electrons, which have large kinetic energies, gradually lose their kinetic energies in both elastic and inelastic collisions in both phases and become thermal electrons. In an inelastic collision in the physical phase of the process the next intermediate particles are created; electron-hole pair (SiO₂⁺ (h⁺)) and electron-excitation (SiO₂^{*}) and electron (e⁻) from direct single ionization of silicon dioxide molecules within nano-Si [6].



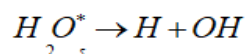
as well as electron-ion pair O₂H⁻ electron excitation H₂O^{*} and electron (e⁻) from direct single ionization of water molecules in water [7].



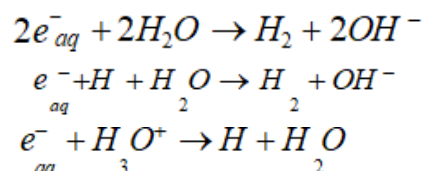
These particles (h⁺, H O⁺, e⁻, SiO₂^{*}, H O^{*}) play an important role in the processes of obtaining the molecular hydrogen in the radiation

heterogeneous conversion of water in the SiO₂/H₂O system. The holes formed inside the nanoparticles under the influence of ionizing radiation can migrate, some of them inside the particle and some of them to the surface of the particle and can be localized on the surface [8,9].

Holes localized in the nanoparticle-water boundary form H₂O ions in contact with adsorbed water molecules. By recombining this ion with thermal electrons or tunnel electrons, an electron-excited water molecule is formed. An excited water molecule with a short lifespan dissociates to form intermediate H and OH products;



On the other hand, some of the electrons formed inside the nanoparticles are emitted inside the particle, some on the particle surface, and some over the particle surface into the water [10]. Electrons emitted from a solid into a liquid phase can gradually lose their kinetic energy in an elastic and inelastic collision in water first transform in thermal electrons and then convert to solvated electrons in water. The process of obtaining molecular hydrogen (9 ÷ 11) during radiolytic reactions between solvated electrons and water molecules and protonated water molecules can be described as follows;



Conclusion

The holes formed inside the nanoparticles under the influence of ionizing radiation can migrate, some of them inside the particle and some of them to the surface of the particle and can be localized on the surface.

Holes localized in the nanoparticle-water boundary form H₂O ions in contact with adsorbed water molecules. By recombining this ion with thermal electrons or tunnel electrons, an electron-excited water molecule is formed.

The results of the study show that both the rate of formation of molecular hydrogen and its radiation-chemical yield, determined by increasing the mass of water;

- Decrease by 10 times if determined for water,
- Increase by 8 times if determined for nano-silicon dioxide.

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