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# THE IMPACT OF SILICON MASS ON THE RADIOLYSIS PROCESS OCCURRING UNDER THE INFLUENCE OF γ- QUANTA ON NANO-Si/$H_2O$ SUSPENSION SYSTEM

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***Abstract:*** The amount, formation rate, and radiation-chemical yield of molecular hydrogen, obtained from water radiolysis process within the system under the influence of γ-quanta ($^{60}$Co, P=22 rad/sec, T=300K), have been defined by maintaining the water volume constant (V=6 ml) and by changing the silica mass ($m_{Si}$=0.0 (pure water), 0.0025, 0.005, 0.01, 0.015 0.02, 0.03, 0.04, 0.06, 0.12 g) in nano-Si/$H_2O$ system with d=50 nm particle size. It was found that the amount, formation rate and radiation-chemical yield of molecular hydrogen, defined according to the overall system from radiation-heterogeneous transformation of water, increase directly proportional to the silica mass in the $m_{Si}\leq 0.02$ g values of nano-silicon mass added to the water, but in $m_{Si}>0,02$ g values, the inclination angle decreases. The formation rate and radiation-chemical yield of molecular hydrogen, defined according to nano-silica, do not change at $m_{Si}\leq 0.02$g, but there is observed a decrease at $m_{Si}>0.02$g. The maximum radiation-chemical yield of molecular hydrogen, determined according to the overall system, got the value of 10.9; but according to the nano-silicon, the value of 1788 molecules/100 eV.

***Key words:*** nano-particles, radiolysis, radiation-chemical yield, Compton scattering

## 1. Introduction

Recently, the radiolysis of liquids, especially water in nano- and micro-heterogeneous systems occurring under the influence of ionizing rays (neutrons, protons, γ-quanta, electrons, ions, etc.) remains one of the most pressing problems of the day in the different fields of science and technology. It becomes clear from our [1,2,3,4] and some foreign authors' [5-13] studies that the amount, formation rate and radiation-chemical yield of molecular hydrogen, obtained from the radiation-heterogeneous transformation of water, occurring in metal or metal oxides/$H_2O$ systems under the influence of γ-quanta, were higher in nano-sized materials. Especially, the radiation-chemical yields of the products obtained in the suspension systems of these materials are higher than that of other systems.

The yield of solvated electrons according to the time has been reviewed during the influence of γ-quanta with 20 nsec-pulse on the suspension of glass nanoparticles with different pores (1-57nm) [14]. It has been found that the yield of electron solvated in 1nm particle pores is 2-times higher than pure water. This proves that some of the electrons formed inside the solid under the influence of radiation are emitted from the solid surface to the liquid phase.

Ouerdane H. and other authors have calculated the electron transfer from silica to water and vice versa in the physical and physical-chemical stages of the process by the influence of ionizing rays on amorphous silicon dioxide system suspended in water by using Monte Carlo method [15]. It has been established that the yield of electrons, emitted into the water, changes depending on the size of nanoparticles.

Electron emission from oxides into the water during impulses and γ-radiolysis in various nano-size oxides ($SiO_2$, ZnO, $Al_2O_3$, $Nd_2O_3$, $Sm_2O_3$, and $Er_2O_3$) suspended, in water, has been defined [16].

It has been investigated that there is always electron transfer in the semiconductor/liquid boundary. The spectrum of electrons solvated in the water proves that the picosecond-nanosecond region does not change, while there is a strong difference in the nanosecond-microsecond region.

These research works have been carried out in two directions:

1) suspension of the added metal or metal oxides in water by maintaining the water volume constant
2) water adsorption on the surface of metal or metal oxides.

In each of these cases, radiation-chemical yields of products (hydrogen, oxygen, hydrogen-peroxide, etc.) obtained from the radiation-heterogeneous transformation of water change depending on their type, bandgap width, particle size, filling rate of absorbed water, temperature of overall system, mass of metal and metal oxides suspended in the water.

Herein, it has been defined the amount, formation rate, radiation-chemical yield of molecular hydrogen obtained from radiation-heterogeneous transformation of water by maintaining the water volume constant (V=6 ml) and by changing the silica mass *(m=0.0025; 0.005; 0.01; 0.015; 0.02; 0.03, 0.04; 0.06; 0.12 g)*, added to the same amount of water and suspended through vibration during the irradiation under the influence of γ-quanta ($^{60}Co$, *P=22 rad/sec, T=300K*),

## 2. Experimental part

Silicon (made by "Skysping Nanomaterials. Inc" USA) with the particle size of d=50 nm, the purity of 99.9% was taken as a research object. The silicon was initially thermal (T=523K) processed in the air within t=72 hours, then the required mass was defined and added to the thermal processed (T=773K) ampoule (V=19 ml), which is purified in a specific mode. The silicon was cooled after a 4-hour thermal treatment (T=673K) in the ampoule under the vacuum ($P=10^{-3}$ mm c.st.) condition and then sealed adding V=6 ml bi-distillate water, which is purified from air under special conditions [17].

The ampoule was irradiated under special conditions (with the condition that nano-Si particle remains dependent in the water through the vibrator during the irradiation) at $^{60}Co$ source with P=22 rad/sec dose rate. The rate of the absorption dose was determined using ferrosulfate and methane methods. Besides, the rate of the absorption dose at a specific research site was calculated using the methods of comparing electron densities [17, 18].

The latest molecular products from the radiation-heterogeneous transformation of water in the nano-Si/$H_2O$ system were defined to be $H_2$, $O_2$, and $H_2O_2$ by using chromatographic methods. As some parts of $O_2$ are on the surface and $H_2O_2$ is in the solution, their amounts can be difficult to determine. Therefore, more accurate information about the kinetic regularities of products obtained from the processes of radiation-heterogeneous transformation of water was obtained based on the amount of molecular hydrogen.

The reaction products were analyzed on an Agilent-7890 chromatograph. A parallel modernized chromatograph "Tsvet-102" (8-10% accuracy) was used to confirm the results. A column with a length of 1 m and a diameter of 3 mm was used on the chromatograph "Tsvet-102". It has been used activated coal with a size of d=0.25÷0.6 mm and argon gas with a purity of 99.9% as a gas carrier in the column.

### 3. Results and discussions

Figure 1 shows time (τ) dependence of the amount of molecular hydrogen ($N(H_2)$) obtained from radiation-heterogeneous transformation of water in the system formed with the addition of V=6 ml pure water (curve 1) and silicon with the particle size of d=50nm and with the mass of m=0.005(2); 0.01(3); 0.015(4); 0.02(5); 0.04(6); 0.06(7) and 0.12 (8) g, suspended in the same amount water through vibrator during irradiation, by the influence of γ-quanta ($^{60}Co$, P = 22 rad/sec, T = 300K).

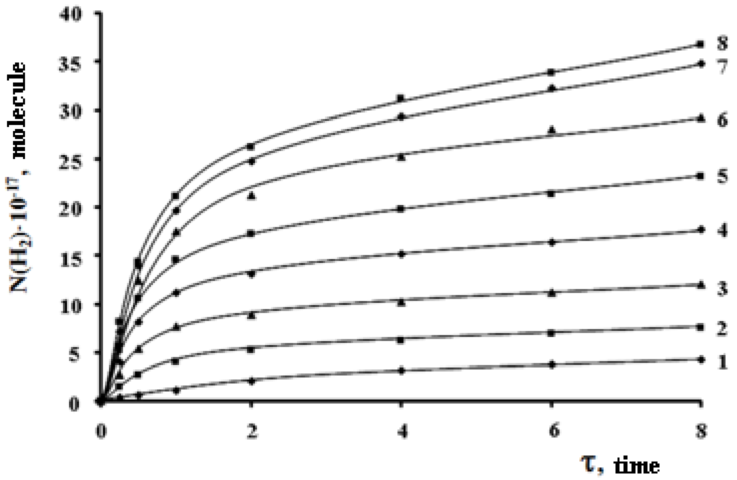


*Fig. 1. Time (τ) dependence of the amount of molecular hydrogen (N ($H_2$)) obtained from radiation-heterogeneous transformation (P=22 rad/sec, T=300K) of water in the systems formed with the addition of 6 ml pure water (curve 1) and nano-Si with the particle size of d=50nm and with the mass of m=0.005(2); 0.01(3); 0.015(4); 0.02(5); 0.04(6); 0.06(7) and 0.12 (8) g, suspended in the same amount water through vibrator during irradiation*

Formation rate and radiation-chemical yield of molecular hydrogen have been defined in two ways from the kinetic parts of the graphs obtained from the investigated systems:

1) according to the overall system;
2) according to nano-Si.

The formation rate of molecular hydrogen according to the general system:

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

On the basis of the above-mentioned expression, it has been calculated that moverall=mwater+mSi is the mass of the overall system, mwater- of water, mSi- of nanosilicon added into the water. The radiation-chemical yield of molecular hydrogen in the overall system has been calculated according to the speed for the overall system. The change in the amount of molecular hydrogen by the addition of nano-Si:

$$\Delta N = N(H_2) - N_0(H_2)$$

From the expression, it can be defined that here, *N ($H_2$)* - is the amount of molecular hydrogen obtained from the water radiolysis in the nano-Si/$H_2O$ system and *$N_0$ ($H_2$)* - that in pure water. The formation rate of molecular hydrogen according to Nano-Si:

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

It has been defined by the above-mentioned expression that here *t* is the irradiation period in the kinetic part. The radiation-chemical yield of the molecular hydrogen according to nano-Si was calculated based on the rate determined by the expression (3). The values for both cases are shown in the table.

**Table**

Mass ($m_{Si}$) dependence of formation rates ($w(H_2)$) and radiation-chemical yields ($G(H_2)$) of molecular hydrogen obtained from radiation-heterogeneous (P=22rad/sec, T=300K) transformation of water in the systems formed by the addition of 6 ml pure water and nano-Si with the particle size of d=50nm suspended in the same amount of water through vibrator during the irradiation.

| $m_{Si}$ , (g) | 0 | 0.0025 | 0.005 | 0.01 | 0.015 | 0.02 | 0.03 | 0.04 | 0.06 | 0.12 |
|---|---|---|---|---|---|---|---|---|---|---|
| according to overall system | | | | | | | | | | |
| $w_{ov}(H_2) \cdot 10^{-13}$, molecule/(g·sec) | 0.61 | 1.91 | 3.1 | 5 | 8.05 | 9,67 | 10.625 | 11.3 | 13.4 | 15 |
| $G(H_2)$ , molecule/100eV | 0.436 | 1.37 | 2.21 | 3.64 | 5.75 | 7,03 | 7.6 | 8.07 | 9.05 | 10.9 |
| according to Nano-Si | | | | | | | | | | |
| $w_{Si}$ $H_2) \cdot 10^{-16}$, molecule/(g·sec) | - | 2.448 | 2.497 | 2.394 | 2.502 | 2.447 | 1.873 | 1.434 | 1.148 | 0.649 |
| $G(H_2)$ , molecule/100eV | - | 1750 | 1785 | 1712 | 1788 | 1749 | 1339 | 1025 | 821 | 454 |

The obtained results show that the radiation-chemical yield of molecular hydrogen, obtained from the radiation-heterogeneous transformation of water in nano-Si/$H_2O$ system with 50nm particle size, defined according to the overall system, increases directly proportional at mSi≤ 0.02 g nano-silicon, added to water, but the inclination angle decreases at mSi>0.02 g. But the radiation chemical yield of molecular hydrogen, defined according to nano-silicon, remains almost constant at mSi≤ 0.02 g nano-silicon, but there is observed a decrease at mSi>0.02 g. In these systems, the maximum radiation-chemical yield of molecular hydrogen, calculated according to the overall system, is 10.9; but that calculated according to nano-silicon is 1788 molecule/100 eV.

In nano-Si/$H_2O$ system, the dependence of the obtaining process of molecular hydrogen from the radiation-heterogeneous transformation of water on the silicon mass can be explained on the basis of the current mechanisms of radiation-heterogeneous processes. Compton scattering mainly occurs in both phases compared to other processes due to the interaction of γ-quanta ($^{60}$Co, Eγ=1.25 MeV) with atoms and molecules that constitute the system while passing through the nano-Si/$H_2O$ system. The kinetic energy of Compton electrons varies in the range of 0÷1.02 MeV depending on the scattering angle. Compton electrons, whose kinetic energies are higher, and new generation δ-electrons formed by them transform to the thermal electrons gradually losing their kinetic energies in elastic and non-elastic collisions in both phases.

Experiments [19-22] and theoretical calculations [23-27] show that radiation-chemical yield of electron-ion pair formed in the water by the influence of γ- quanta in the physical (<$10^{-15}$ sec.) and physico-chemical ($10^{-15} \div 10^{-12}$ sec) stages of the process is 3.4 pair/100 eV, but the radiation chemical yield of electron-hole [28, 29] pair ($E_h$=2.5$E_g$ for the formation of an electron-hole pair, $E_g$=1.5 eV) inside the silicon particle is 26.6 pair/100 eV. Some electrons formed inside the particle can be localized in the volume and surface defects as a result of migration, and some can be emitted into the water from the nanoparticle surface. In the solid phase, the electron [14, 15, 16] concentrations of electrons are higher than that of the other parts in the liquid phase around the solid-state particle due to the electrons emitted from the solid-state to the liquid phase. Electrons emitted from the surface of the particle into the water can become thermal electrons gradually losing their kinetic energy, and then be able to be solvated. Molecular hydrogen is derived from the reactions between solvated electrons *($e^-_{aq}$)* band water molecules, protonated (*$H_3O^+$)* water molecules and H atoms (1-3).

$$2e_{aq}^- + 2H_2O \rightarrow H_2 + 2OH^- \quad (1)$$

$$e_{aq}^- + H + H_2O \rightarrow H_2 + OH^- \quad (2)$$

$$e_{aq}^- + H_3O^+ \rightarrow H_2 + H_2O \quad (3)$$

On the other hand, the holes inside the silicon particle formed by the influence of γ-quanta are migrated to the surface some of which are captured by the defects inside the particle, and some migrate to the surface, and create positive charge centers being captured by surface defects, and they, in turn, play a role of adsorption center (4) for water molecules with electro donor nature and at subsequent stages recombination center (5) for electrons [30]. The electron-excitation complex obtained due to recombination plays a key role in the process of obtaining molecular hydrogen from the radiation-heterogeneous transformation of water (6).

$$\equiv Si^+ + H_2O \rightarrow [\equiv Si....H_2O]^+ \quad (4)$$

$$[\equiv Si....H_2O]^+ + e^- \rightarrow [\equiv Si....H_2O]^* \quad (5)$$

$$[\equiv Si....H_2O]^* \rightarrow \equiv Si - OH + H \quad (6)$$

So if both the mass and the size of the silicon change, the energy transmitted to the water and the radiation-chemical yield of the corresponding molecular hydrogen change too. However, after a certain concentration of each-size particles in the medium, there is a balance formed between the formation processes of energy carriers and intermediate products.

## 4. Conclusion

Based on the research, the following conclusions can be reached:

- It has been established that the amount, formation rate, radiation-chemical yield of molecular hydrogen defined according to the overall system obtained from the radiation-heterogeneous transformation of water in the nano-Si/$H_2O$ system with d=50 nm particle size by the influence of γ-quanta ($^{60}$Co, P=22 rad/sec, T=300K), increase directly proportional to the silicon mass added to the water at mSi≤0.02 g but there is a decrease in the inclination angle at $m_{Si}$> 0.02 g.

The radiation-chemical yield of molecular hydrogen in this system was $G(H_2)$ = 10.,9 molecule/100 eV, which is significantly higher than that of pure water ($G_0(H_2)$ = 0.45 molecule/100 eV).

- The formation rate and radiation-chemical yield of the molecular hydrogen defined according to nano-silica do not change at $m_{Si} \leq 0.02$ g, while there is a decrease at $m_{Si} > 0.02$ g. The maximum radiation-chemical yield of molecular hydrogen determined by nano-silicon was 1788 molecule/100 eV.

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## ВЛИЯНИЕ МАССЫ КРЕМНИЯ НА γ-РАДИОЛИЗ СУСПЕНЗИРОВАННОЙ СИСТЕМЫ Нано-Si/$H_2O$

**С.М. Баширова**

***Резюме:*** Определены количество, скорость образования и радиационно-химический выход молекулярного водорода, полученного γ-радиолизом воды (*$^{60}Co$, P=22 рад/с, T=300K*) в системе нано-Si/$H_2O$ с размерами частиц Si, равными *d=50 нм,* при постоянном объеме воды (*(V=6 мл)* и изменяющейся массе кремния (*$m_{Si}$=0.0 (чистая вода), 0.0025, 0.005, 0.01, 0.015 0.02, 0.03, 0.04, 0.06, 0.12 г)*. Выявлено, что в пересчете на общую энергию, поглощенную системой, количество, скорость образования и радиационно-химический выход молекулярного водорода, полученного радиационно-гетерогенным разложением воды, увеличиваются прямо пропорционально массе кремния при значениях *$m_{Si}\leq 0,02$ г*, а при *$m_{Si}>0,02$ г* угол наклона, наоборот, уменьшается. Скорость образования и радиационно-химический выход водорода, вычисленные в пересчете на энергию, поглощенную нано-Si, практически не изменяются при *$m_{Si}\leq 0,02$ г*, а при *$m_{Si}>0,02$ г*

наблюдается их уменьшение. Полученные максимальные значения радиационно-химического выхода молекулярного водорода, в пересчете на общую систему и нано-Si, равны *10,9* и *1788* молекул/100эВ соответственно.

***Ключевые слова:*** нано-частицы, радиолиз, радиационно-химический выход, Комптоновское рассеяние

## Nano-Si/$H_2O$ SUSPENZİYALI SİSTEMİNƏ γ-KVANTLARIN TƏSİRİLƏ GEDƏN RADİOLİZ PROSESİNƏ SİLİSİUMUN KÜTLƏSİNİN TƏSİRİ

**S.M. Bəşirova**

***Xülasə:*** d=50 nm hissəcik ölçülü nano-Si/$H_2O$ sistemində suyun həcmini sabit saxlamaqla (V=6 ml) silisiumun kütləsini ($m_{Si}$=0.0 (təmiz su), 0.0025, 0.005, 0.01, 0.015 0.02, 0.03, 0.04, 0.06, 0.12 q) dəyişməklə, γ-kvantlarin ($^{60}$Co, P=22 rad/san, T=300K) təsirilə sistem daxilində suyun radilizi prosesindən alınan molekulyar hidrogenin miqdarı, əmələgəlmə sürəti və radiasiya-kimyəvi çıxımı təyin edilmişdir. Müəyyən edilmişdir ki, suyun radiasiya-heterogen çevrilməsindən alınan ümumi sistemə görə təyin edilmiş molekulyar hidrogenin miqdarı, əmələgəlmə sürəti və radiasiya-kimyəvi çıxımı suya əlavə olunan nano-silisiumun kütləsinin $m_{Si}$≤0,02 q qiymətlərində, silisiumun kütləsi ilə düz mütənasib olaraq artır, $m_{Si}$>0,02 q qiymətlərində isə meyl bucağı azalır. Nano-silisiuma görə təyin edilən molekulyar hidrogenin əmələgəlmə sürəti və radiasiya-kimyəvi çıxımı $m_{Si}$≤0,02 q qimətlərində demək olarki dəyişməyir, $m_{Si}$>0,02 q qiymətlərində isə azalma müşahidə olunur. Ümumi sistemə görə təyin edilən molekulyar hidrogenin maksimum radiasiya-kimyəvi çıxımı 10,9; nano-silisuma görə isə 1788 molekul/100 eV qiymətləri alınmışdır.

***Açar sözlər:*** nano-hissəciklər, radioliz, radiasiya-kimyəvi çıxım, Kompton səpilməsi