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IMPACT OF MASS AND SIZE EFFECTS OF BERYLLIUM OXIDE ON WATER RADIOLYSIS PROCESS OCCURRING IN BeO+H₂O SYSTEM UNDER THE INFLUENCE OF GAMMA-QUANTA

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Abstract: It has been studied the influence of mass and size effects on amount, formation rate and radiation-chemical yield of molecular hydrogen, formed in water radiolysis process under the influence of γ -quanta (^{60}Co , $P=19,05$ rad/sec, $T=300\text{K}$), by changing the mass of beryllium oxide ($m = 0.0$ (pure water), 0.01, 0.02, 0.04, 0.08, 0.2 g) and size of its particles ($d =$ lower than 4 mkm, 32-53 and 75-106 mkm) and keeping the volume of water constant ($V=5$ ml) in BeO+H₂O system. Given size particles – maximum radiation-chemical yields of molecular hydrogen, obtaining from radiolytic decomposition of water in water system, are defined as $G(\text{H}_2)=3,1; 1,83$ and $0,92$ molecule/100 eV, respectively. The yield of hydrogen is directly proportional to beryllium oxide mass at lower values of the ratio of beryllium oxide mass to water, but there is observed saturation state after certain value of ratio depending on size of particle (ratio, appropriate to saturation state, is equal to 1/150 for the particles lower than $d<4\text{mkm}$, 1/120 for $d=32-53$ mkm and 1/80 for $d=75-106\text{mkm}$).

Key words: *micro particle, radiolysis, radiation-chemical yield, Compton scattering*

1. Introduction

Radiolysis process of liquids, especially water in nano- and micro-heterogeneous systems remains as one of the actual problems of the day. The amount, formation rate, radiation-chemical yield of molecular hydrogen, obtained in water radiolysis process which occurs in metal or metal oxides + H₂O systems under γ -quanta influence, are several times higher than pure water. Water radiolysis process [1-12], occurring in metal or metal oxides + H₂O systems under γ -quanta influence, has been carried out in two direction. First one is the suspension of these materials in water, second one is the change of amount, formation rate and radiation chemical yield of products (hydrogen, oxygen, hydrogen peroxide) obtaining from water radiolysis process, which takes place during absorption of water in some monolayers in the surface of these materials, depending on:

- type of solids,
- band gap width of solids
- size of solid particles
- thickness of the water layer, adsorbed on the surface of solids;
- temperature of general system
- mass of solids, subjected to suspension in water
- nature of water-soluble substances

In the presented work, it has been studied the influence of beryllium oxide mass, suspended in water and size of its particles on the amount, formation rate and radiation-chemical yield of molecular hydrogen, formed in water radiolysis process that takes place under the influence of γ -quanta (^{60}Co , $P=19,05$ rad/sec, $T=300\text{K}$) on different size (lower than $d=4$ mkm, 32-53 mkm, and 75-106 mkm) beryllium oxide system, suspended through vibrator during radiation in 5ml water.

2. Experimental part

It has been used different size (lower than $d=4 \text{ mkm}$, 32-53, and 75-106 mkm) high-purity (99,9%) beryllium oxide in order to study the influence of beryllium oxide mass suspended in water and size of its particles on the amount, formation rate and radiation-chemical yield of molecular hydrogen formed in water radiolysis process which takes place under the influence of γ -quanta in $\text{BeO} + \text{H}_2\text{O}$. Required mass ($m = 0,01; 0,02; 0,04; 0,08$ and $0,2 \text{ g}$) has been purified in special mode and added to thermal processed ($T=773\text{K}$) ampoule ($V=46 \text{ ml}$) after perfunctory processing of beryllium oxide ($t=72\text{hours}$) in the open air at $T=773\text{K}$. Beryllium oxide has been covered in an ampoule by cooling and adding 5ml bidstillated water, which is purified from air, to it after thermal processing ($T=673\text{K}$) in a vacuum ($P=10^{-3} \text{ mm c.st.}$) within 4 hours [13]. The ampoule has been irradiated in special condition (with the condition of suspension of beryllium oxide in water by vibrator within irradiation). Absorption dose rate has been defined by Ferrosulphate and methane methods [14].

The amount of molecular hydrogen (accuracy 5-10%) obtained in radiolytic decomposition of water in $\text{BeO} + \text{H}_2\text{O}$ systems, has been defined by chromatographic method (Цбер-102). The column with 1m length and 3mm internal diameter has been used in chromatography. It has been used in the column, the activated choral with the size of $d=0,25 \div 0,6 \text{ mm}$ and high- purity (99,9%) argon as gas carrier.

3. Results and discussion

It has been given the graphic of time dependence of the amount of molecular hydrogen obtained in radiolytic decomposition of water, taking place under the influence of γ - quanta (^{60}Co , $P=19,05 \text{ rad/sec}$, $T=300\text{K}$) on beryllium oxide system with different the size (lower than 4mkm (fig.1) and 32-53 mkm (fig.2)) and mass ($m=0,01(1); 0,02(2); 0,04(3), 0,08 (4), 0,20 \text{ g}(5)$) in 5ml bidstillated water.

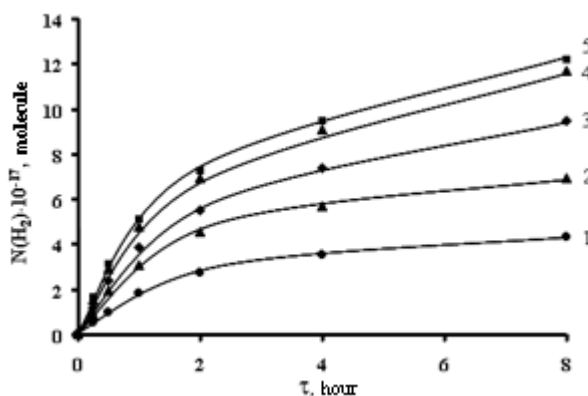


Fig. 1. Time dependence of the amount of molecular hydrogen, formed within radiation-catalytic ($P=19,05 \text{ rad/sec}$, $T=300\text{K}$) decomposition of water in beryllium oxide system with the size of $d < 4 \mu\text{m}$ and different mass- (1(0,01 g); 2(0,02 g); 3(0,04 g); 4(0,08 g); 5(0,20 g)), which is suspended in 5ml water.

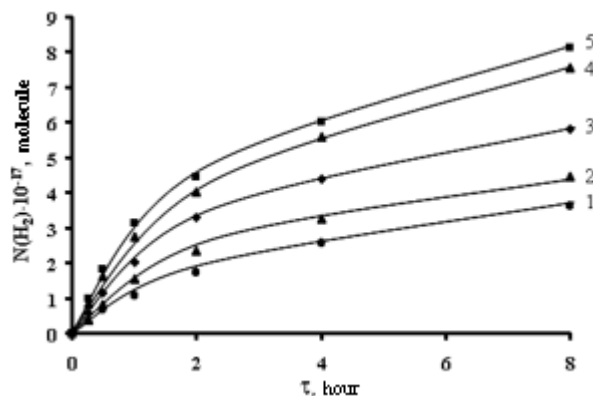


Fig. 2. Time dependence of the amount of molecular hydrogen, formed within radiation-catalytic ($P=19,05$ rad/sec, $T=300K$) decomposition of water in beryllium oxide system with the size of $d=32\div53 \mu m$ and different mass (1(0,01 g); 2(0,02 g); 3(0,04 g); 4(0,08 g); 5(0,20 g)), which is suspended in 5ml water

In figure 3, it has been given the graphic of time dependence of the amount of molecular hydrogen, formed from radiolytic decomposition of water, occurring in 5ml pure water (1st curve) and beryllium oxide system (2-6th curves) with the size of $d=75-106 mkm$ and mass of $m=0,01(2)$; $0,02(3)$; $0,04(4)$, $0,08(5)$, $0,20 q(6)$ under γ -quanta influence.

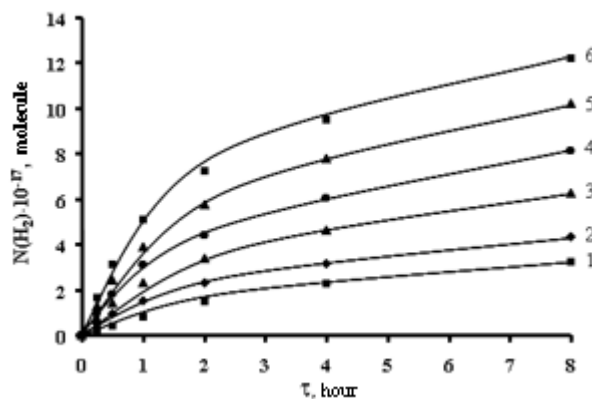


Fig. 3. Time dependence of the amount of molecular hydrogen, formed from radiolytic decomposition of water, occurring in 5ml pure water (1st curve) and beryllium oxide system (2-6th curves) with the size of $d=75-106 mkm$ and mass of $m=0,01(2)$; $0,02(3)$; $0,04(4)$, $0,08(5)$, $0,20 q(6)$ suspended through vibrator within irradiation in same amount of water.

It has been defined the formation rate $w(H_2)$ and radiation-chemical yield $G(H_2)$ with 100 eV absorption energy of molecular hydrogen from linear part of kinetic curves (figure 1-3) obtained from studied systems.

Table

The dependence of formation rate - $w(H_2)$ and radiation-chemical yield - $G(H_2)$ of molecular hydrogen, formed in radiolytic decomposition of water ($P=19,05$ rad/sec, $T=300K$), occurring in 5ml pure water and with the addition of different size beryllium oxide (lower than $d=4 mkm$, $32-53$, and $75-106 mkm$) which is suspended through vibrator during irradiation in the same amount of water, on the mass of beryllium oxide - $m(BeO)$.

m(BeO), g	4 μm<		32-53 μm		75-106 μm	
	w(H ₂)·10 ⁻¹³ , molecule/g·sec	G(H ₂), molecule/ 100 eV	w(H ₂)·10 ⁻¹³ , molecule/g·sec	G(H ₂), molecule/ 100eV	w(H ₂)·10 ⁻¹³ , molecule/g·sec	G(H ₂), molecule/ 100eV
0	0,61	0,436	-	-	-	-
0.01	1,13	0,95	0,69	0,58	0,62	0,52
0.02	1,93	1,63	0,82	0,69	0,77	0,65
0.04	2,66	2,24	1,24	1,05	0,91	0,77
0.08	3,35	2,82	1,84	1,55	1,04	0,88
0.20	3,66	3,1	2,18	1,83	1,08	0,92

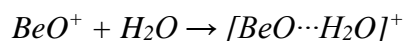
Compton scattering takes place in both phases under the influence of $E_\gamma=1,25$ MeV energy γ -quanta on $BeO + H_2O$ system.



The energy of Compton electrons varies between 0÷1,02 MeV depending on scattering angle. Compton electrons, of which kinetic energy is high, turn to thermal electrons by gradually losing their energy in elastic and non-elastic collision in both phases. A part of electron-ion pair, formed in BeO particle and water, recombines with its pair as a result of influence of Coulomb interaction, and some of them move away from each other. The main part of electrons in energy spectrum is the electrons with 100 eV energy. Experiments [15÷18] and theoretical calculations [19÷25] show that, radiation-chemical yield of electron-ion pair, formed in water under the influence of γ -quanta and electrons, becomes 3,4 pair/100 eV in physical and physical-chemical stages of process, but the radiation-chemical yield of electron-excitation states - 2,6 excitation/100 eV. But, the radiation-chemical yield of electron-ion pair, formed in BeO particle, becomes 3,5 pair/100 eV, and radiation-chemical yield of electron-excitation state-2,8 excitation/100 eV. The electrons can easily move from solid to liquid phase or vice versa. Calculations show that, absorption dose is 2,33 times higher in beryllium oxide than water. As the number of electrons, moved from solid to liquid phase, are more and most of them are 100 eV electrons, the concentration of electrons becomes higher around solid particle in $d=20$ nm space than other parts of liquid phase. Electrons, lost their energy, turned to thermal or solvated electrons in liquid. It can be described the reaction of molecular hydrogen, obtained in radiolytic decomposition occurring between solvated (e_{aq}^-) electrons and water molecules or protonated water ion as follows:



The complex of

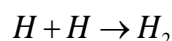


is formed as a result of interaction of ion-dipole on the border of $\text{BeO}+\text{H}_2\text{O}$.

Then that complex forms excited complex of $[\text{BeO}\cdots\text{H}_2\text{O}]^*$ by the recombination with tunnel and thermal electrons. Electron excitation energy formed in the complex and solids, causes dissociation of water molecule by passing to adsorbed water molecule and as a result, H and OH radicals with high reaction ability are formed:



As a result, molecular hydrogen



is formed.

It becomes clear from the results of the research work that:

- The amount, formation rate, radiation-chemical yield of molecular hydrogen formed in water radiolysis process, which occurs under the influence of γ -quanta on $\text{BeO} + \text{H}_2\text{O}$ system, decrease by the increase of beryllium oxide size. The yield with the particles in the size of $d < 4$ mkm, 32-53 and 75-106 mkm got the values of 3.1, 1.83 and 0.92, respectively
- Radiation-chemical yield of molecular hydrogen is directly proportional to beryllium oxide mass at lower values of the ratio of beryllium oxide mass to water, but there is observed saturation state after certain value of ratio depending on size of particle (ratio, appropriate to saturation state, is equal to 1/150 for the particles lower than $d < 4$ mkm, 1/120 for $d = 32-53$ mkm and 1/80 for $d = 75-106$ mkm).

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ВЛИЯНИЕ МАССОВЫХ И РАЗМЕРНЫХ ЭФФЕКТОВ ОКСИДА БЕРИЛЛИЯ НА ПРОЦЕСС РАДИОЛИЗА ВОДЫ, ПРОТЕКАЮЩЕГО В СИСТЕМЕ BeO+H₂O ПОД ВОЗДЕЙСТВИЕМ ГАММА – КВАНТОВ.

Я. Д. Джафаров, С. Р. Гаджиева, Н. К. Рамазанова, С. М. Алиев

Резюме: Было изучено влияние массовых и размерных эффектов на количество, скорость образования и радиационно – химический выход молекулярного водорода, образующегося в процессе радиолитического превращения воды под воздействием γ - квантов (^{60}Co , $P=19,05$ рад/сек, $T=300\text{K}$) на систему $\text{BeO}+\text{H}_2\text{O}$, при условии сохранения постоянным объем воды ($V=5$ мл) и при изменении массы оксида бериллия ($m=0.0$ (чистая вода), 0.01, 0.02, 0.04, 0.08, 0.2 г) и размеров его составляющих частиц (d = меньше 4 мкм, 32-53 и 75-106 мкм). Учитывая размеры данных частиц, были определены соответственно максимальный радиационно-химический выход молекулярного водорода $G(\text{H}_2)=3,1; 1,83$ и $0,92$ молекул/100 эВ, образующегося в процессе радиолитического превращения воды в водной системе. При малых массах частиц оксида бериллия по сравнению с массой воды, выход молекулярного водорода прямо пропорционально массе оксида бериллия, а после определенных значений соответствия, в зависимости от размеров частиц наблюдается процесс насыщения (соотношение процессу насыщения при малых размерах частиц $d<4$ мкм соответственно равно 1/150, для частиц с размерами $d=32-53$ мкм равно 1/120, а для частиц с размерами $d=75 - 106$ мкм равно 1/80).

Ключевые слова: микрочастица, радиолит, радиационно-химический выход, Комптоновское рассеяние

**QAMMA-KVANTLARIN TƏSİRİLƏ BeO+H₂O SİSTEMİNDƏ GEDƏN SUYUN
RADIOLİZİ PROSESİNƏ BERİLLIUM OKSİDİN KÜTLƏ VƏ ÖLÇÜ
EFFEKTƏRİNİN TƏSİRİ**

Y. D. Cəfərov, S .R. Hacıyeva, N. K. Ramazanova, S. M. Əliyev

Xülasə: BeO+H₂O sistemində suyun həcmi sabit saxlamaqla (V=5 ml) berillium oksidin kütləsini (m=0.0 (təmiz su), 0.01, 0.02, 0.04, 0.08, 0.2 q) və onu təşkil edən hissəciklərinin ölçüsünü (d=4 mkm-dən kiçik, 32-53 və 75-106 mkm) dəyişməklə, γ-kvantların (⁶⁰Co, P=19,05 rad/san, T=300K) təsiri ilə suyun radiolizi prosesindən əmələ gələn molekulyar hidrogenin miqdarına, əmələgəlmə sürətinə və radiasiya-kimyəvi çıxımına kütlə və ölçü effektlərinin təsiri öyrənilmişdir. Verilmiş ölçülü hissəciklər – su sistemində suyun radiolitik çevrilməsindən alınan molekulyar hidrogenin maksimal radiasiya-kimyəvi çıxımları uyğun olaraq G(H₂)=3,1; 1,83 və 0,92 molekul/100 eV təyin edilmişdir. Berillium oksidin kütləsinin suyun kütləsinə nisbətinin kiçik qiymətlərində hidrogenin çıxımı berillium oksidin kütləsi ilə düz, nisbət müəyyən qiymətindən sonra, hissəciyin ölçüsündən asılı olaraq doyma halı (doyma halına uyğun nisbət d<4 mkm-dən kiçik hissəciklər üçün 1/150-yə, d=32-53 mkm hissəciklər üçün 1/120-yə və d=75-106 mkm ölçülü hissəciklər üçün isə 1/80-ə bərabər olur) müşahidə olunur.

Açar sözlər: mikrohissəcik, radioliz, radiasiya-kimyəvi çıxım, kompton səpilməsi