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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

STUDY OF RADIATION CHEMICAL PROCESSES IN THE $(RaO)_x(SiO_2)_y$ +H_2O SYSTEM

Specialty:	2305.01 - Nuclear chemistry
Field of science:	Chemistry
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GENERAL CHARACTERISTICS OF THE WORK

In recent years, atomic-hydrogen energy has again become the most important scientific and technical problem of the era. So, since the use of energy sources of hydrocarbon based has created a catastrophic ecological situation, and other alternative energy sources are unstable and do not satisfy demand due to power factors, atomic hydrogen energy and nuclear energy are one of the most relevant scientific and technical research areas of that time in recent years. To this end, extensive research is being carried out in the field of atomic- hydrogen energy. As known, the research in this area has been carried out for many years at the Institute of Radiation Problems. One of the topical areas of atomic-hydrogen energy is the direct use of the energy of the main energy-carrying particles of the processes of nuclear transformations in the processes of water splitting. In the literature, due to the energy of fission fragments of uranium and plutonium isotopes under the action of neutrons, an increase in the yield of hydrogen during the radiolytic splitting of water and, mainly, the rate of absorption of hydrogen was observed. Considering that existing nuclear materials are alpha-active, and alpha particles, gamma quanta obtained in the process of fission fragments in radiolysis processes and estimating the effect of temperature on the kinetic energy of fragments, are of great importance in providing the mechanism of processes and developing technologies. Therefore, the study of alpha particles obtained during the decay of model radioactive isotopes, the radiolytic decomposition of water under the influence of temperature and gamma rays, and the identification of patterns are among the urgent tasks in the direction of atomic hydrogen energy and nuclear technology. The topic of the dissertation, which is devoted to the study of the processes of radiolysis and thermoradiolysis of water in the presence of the radioactive isotope Ra^{226}, which is one of the daughter isotopes of the decay of uranium elements, is very relevant and of great importance. In fragmentary radiolysis, the fragmented core is embedded in a radiation-catalytically active oxide carrier. In the presented dissertation work, the Ra^{226} isotope is included in the composition of the radiation-catalytically active silicate during the radiolysis of water. Thus, there is no doubt about the relevance of the dissertation devoted to clarifying the indicated problems in the process of obtaining hydrogen from water by the fragmentation radiolytic method based on a silicate system containing various amounts of radium.

The main purpose of the dissertation work - is the discovery of the regularities of the influence of internal alpha particles, external gamma-rays and temperature on the process of hydrogen formation during the radiation-chemical decomposition of water in the presence of radium silicates containing radioactive radium in various amounts (according to activity).

To achieve the goal, the following issues were resolved:

- synthesis of the $(RaO)_x(SiO_2)_y$ system containing radium of different activity,
- study of structural properties depending on the activity of radium in the obtained system $(RaO)_x(SiO_2)_y$,
- evaluation of the role of internal alpha, external gamma-radiation and temperature in the production of molecular hydrogen in the radiation-catalytic decomposition of water in the presence of $(RaO)_x(SiO_2)_y$,
- investigation of hydrogen generation during water splitting under the action of secondary electron beams emitted from $(RaO)_x(SiO_2)_y$ into the contact medium under the influence of gamma- rays,
- transfer of the internal and external energy of radium gamma-radiation to the surface level and the formation of molecular hydrogen at different temperatures as a result of the decomposition of water on the surface $(RaO)_x(SiO_2)_y$ in radiation-chemical processes,
- study of the effect of internal alpha radiation and external gamma-radiation on the electrical properties of the system $(RaO)_x(SiO_2)_y$,
- study of the formation of paramagnetic radiation defects in $(RaO)_x(SiO_2)_y$ under the action of γ-irradiation,
- discussion of the obtained results and description of the

process mechanism.

The objects of study were pure SiO_2 and radium-containing silicate systems of various activity, obtained by boiling in a given temperature range by a special method of hydrolysis and precipitation from solution.

The scientific novelty of the work:

For the first time:

• the kinetic regularities of the processes of obtaining molecular hydrogen from the process of radiation-heterogeneous decomposition of water with the participation of the $(RaO)_x(SiO_2)y$ system taken as a model version of fragmentation radiolytic processes were studied and the kinetic parameters were determined,

• the effect of radium activity on the yield of hydrogen during radiative and radiative-thermocatalytic decomposition of water with the participation of the $(RaO)_x(SiO_2)y$ system is revealed, and its energy fraction is estimated,

• in the presence of the $(RaO)_x(SiO_2)y$ system, a regularity of the influence of temperature on the rate of hydrogen formation during the radiation-catalytic decomposition of water and the radiation-chemical yield in the range of $T=300\div673K$ was revealed and the activation energies of the processes were determined,

• under the action of gamma rays, the role of secondary electron beams emerging from the $(RaO)_x(SiO_2)_y$ system into the contact medium in the radiation-heterogeneous decomposition of water was revealed,

• to characterize the formation of non-equilibrium charge carriers, which play the role of energy carriers in the radioactive system $(RaO)_x(SiO_2)_y$ under the action of gamma quanta, using the methods of electrical conductivity and electron paramagnetic resonance (EPR), identification of localized charge carriers was carried out, and an electrical effect on conductivity was detected.

The practical significance of the work: The results obtained are of great importance in the field of atomic-hydrogen energy, which is a topical scientific and technical problem of our time. Thus, the results obtained will be used in the selection of a catalyst to produce hydrogen by the fragmentation radiation-catalytic method.

In addition, radiation and radiation-thermal processes in a radioactive-silicate system can be proposed for use in ensuring hydrogen safety in storage systems for spent nuclear materials and radioactive substances, while characterizing the role of radiation-chemical conversion processes.

Main clauses defended:

• acquisition of a radium-silicate system containing a radium group of various activity, study of radioactivity, structure and hydrate surface;

• study of the processes of radiolysis in contact with water $(RaO)_x(SiO_2)_y$ depending on the activity of radium;

• study of the kinetics of formation of molecular hydrogen at different temperatures as a result of radiation-chemicals processes upon contact of radium silicate system with water;

• characterization of the role of energy transfer to the surface level and the contact medium in the process of radiolytic splitting of water with the participation of the radium-silicate system;

• the influence of alpha rays of radioactive radium and external gamma rays on the electrical conductivity of the radium-silicate system;

• formation of paramagnetic radiation defects under the influence of alpha-rays of radioactive radium and external gamma-rays;

• study of water splitting processes in radiation-heterogeneous processes as a result of the formation of unbalanced charge carriers in radium silicates under the action of γ-rays and their transfer to surface levels;

Approbation of work. The results of the dissertation were discussed at the following scientific conferences: International conference dedicated to the 100th anniversary of the birth of M. Malikzade "Peaceful use of atomic energy" November 8-10, Baku, Azerbaijan, 2010; Republican Conference dedicated to the 20th anniversary of the Independence of the Republic of Azerbaijan "Perspectives for the use of alternative and renewable energy sources" June 1-2, 2011, Baku, Azerbaijan. IV International Conference "Perspectives for the Peaceful Use of Atomic Energy"

November 23-25, 2011; 8th International Conference "Nuclear and Radiation Physics" September 20-23, 2011, Almaty, Kazakhstan; International Conference “Nuclear science and its application”, Samarkand, Uzbekistan, September 25-28, 2012; The V international Conference “Perspectives of peaceful use of nuclear energy”, November 21-23, 2012. Baku, Azerbaijan; Materials of the 4th republican conference on modern problems of physics. Baku: Azerbaijan 2012; 9th International Conference Nuclear and Radiation Physics, 24-27 September 24-27, Almaty, Kazakhstan-2013; 10th International Conference "Nuclear and Radiation Physics", September 08-11, 2015, Kazakhstan. International Symposium "New Trends in the Development of Fundamental and Applied Physics: Problems, Achievements and Prospects" Tashkent, November 10-11, 2016. 11th International Conference "Nuclear and Radiation Physics", September 12-15, 2017, p.194, Almaty, Republic of Kazakhstan. International Scientific Forum «Nuclear Science and Technologies» Almaty, 2017. VIII international conference Semipalatinsk test site, September 11-13, 2018.

Publications: 26 scientific papers have been published on the topic of the dissertation. These cases were discussed at international conferences and seminars and published as abstracts in local and foreign journals, which fully meets the requirements of the Higher Attestation Commission under the President of the Republic of Azerbaijan.

Structure and scope of work. The dissertation consists of an introduction, five chapters, a conclusion and a list of references. The work is explained on 134 pages, there are 38 graphs and figures, 15 tables. The bibliography includes 141 works, including personal articles of the author.

SUMMARY OF THE WORK

The introduction justifies the relevance of the problem being solved and its place in modern science, briefly and accurately sets out the purpose of the study and the questions posed, indicates the scientific novelty and practical significance of the work. Abstract chapters of the dissertation are annotated, and a list of published works is given.

The **first chapter** is dedicated to a review of the literature, detailed literature data of recent years are given on the production of oxygen-containing silicon compounds, their physicochemical properties, their application in the field of nuclear technology, the general laws of water radiolysis, the effect of ionizing rays and temperature on radium-silicate systems. In this case, a brief review of the published scientific literature on the processes occurring as a result of the action of external radiation on these systems, as well as on the processes occurring on the surface of silicates, is given. At the end of the chapter, the results of the search and analytical work carried out in the existing scientific literature are summed up, and the choice of the topic of the dissertation is justified, considering the existing characteristics of radium-silicate systems and areas of application.

In the **second chapter**, the synthesis of radium-silicate systems with different activity content and the method for determining the specific activity of these systems in a gamma spectrometer with a Ge-detector are given. To study the structural properties of the samples, Methods of X-ray diffraction analysis, infrared spectroscopy, thermal analysis, etc. were used.

For this, synthesis was carried out by known methods and radium-silicate systems containing various amounts of radium were obtained. For the synthesis of radium silicate, solutions of $RaCl_2$ and TEOS (tetraethylorthosilicate) were used, and acetic acid was used to purify the intermediate reaction products. First, $RaCl_2$ was added to the TEOS solution, and then CH_3COOH was added at a certain temperature ($T>60^0C$). The resulting intermediate product C_2H_5OH reacts with CH_3COOH to form an ethyl acetate ester. To obtain silicates of various compositions, solutions of TEOS and $RaCl_2$ were taken in the ratio 1:1, 1:10, 1:20, respectively. The reaction equation or the synthesis of the system was mainly carried out on the following chemical reactions:

$$Si\,(OC_2H_5)_4 + 4H_2O \rightarrow Si(OH)_4 + 4C_2H_5OH \qquad (1)$$

$$Si(OH)_4 + RaCl_2 \rightarrow (RaO)_x(SiO_2)_y + 2HCl \quad (2)$$
$$C_2H_5OH + CH_3COOH \rightarrow CH_3COOC_2H_5 + H_2O. \quad (3)$$

The composition of the obtained compounds can generally be expressed as $(RaO)_x(SiO_2)_y$. A three-component radium-silicate system was obtained, and the table gives their activity depending on the amount of radium, as well as the amount of radium:

I - $(RaO)_x(SiO_2)_y$ with 0,6% Ra
II - $(RaO)_x(SiO_2)_y$ with 4,8% Ra
III - $(RaO)_x(SiO_2)_y$ with 10,1% Ra

The resulting radium silicate systems of different activity were dried in vacuum at a temperature of T=373K. The specific and effective activity of radionuclides in the synthesized radium-silicate systems was determined by the gamma spectrometric method based on gamma-quanta with an energy of 186 keV in the HPGe gamma spectrometer with a high-purity germanium detector manufactured by Canberra (USA) and, respectively, was equal to 260, 2500, 6100 Bq/g.

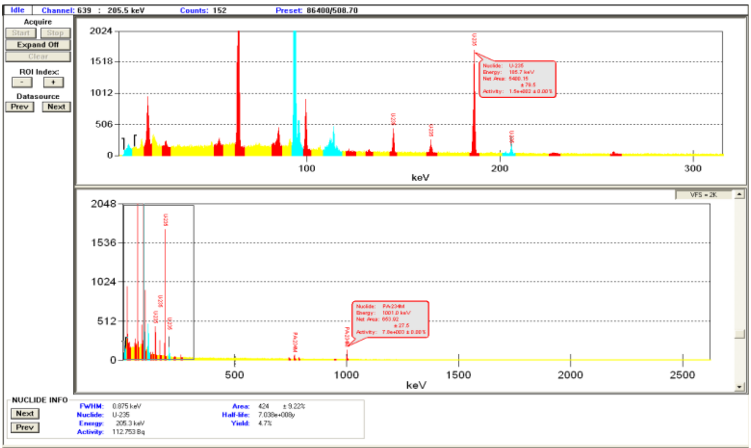

Figure 1. Gamma spectrum of a radium-silicate system containing 10,1% radium.

Table 1.
Activity and amount of radium in compounds $(RaO)_x(SiO_2)_y$

№	Sample name	Activity, Bq/g	The amount of radium in the sample with %.
1	$(RaO)_x(SiO_2)_y$	260	0,6
2	$(RaO)_x(SiO_2)_y$	2500	4,8
3	$(RaO)_x(SiO_2)_y$	6100	10,1

In addition, methods for studying the electrical properties of the system $(RaO)_x(SiO_2)_y$, methods for analyzing products formed in processes occurring on the surface of radiation-heterogeneous systems, and methods for estimating errors of given parameters. The structural shape of the obtained samples with different activities was determined by the X-ray diffraction method. X-ray phase analysis of the studied samples was performed on a D8ADVANCE diffractometer (Germany). Operating mode: 40 kV, 40 mA, CuK2 radiation, λ=1.5406Å, $05^0<2\theta<80^0$, temperature T=300K. Thermal analysis of the samples was carried out on a Perkin-Elmer STA 6000 instrument (USA). During the measurement, the samples were heated at a rate of 50°C/min, and the flow rate of argon was taken equal to 20 ml/min to maintain the homogeneity of the system. A technique is given for calculating the energies of endo- and exo-effects from the obtained spectra, as well as a technique for changing the mass in various temperature ranges depending on temperature. IR spectroscopy was used to study structural relationships. A technique for recording IR absorption spectra of radium-silicate samples on a “Specord 71 IR” spectrometer (Germany) in the frequency range 4600-400 cm^{-1} is presented. The study of paramagnetic centers formed in radium silicates under the action of internal and external gamma radiation was carried out on an EMX plus Bruker electron paramagnetic resonance (EPR) spectrometer and a technique for measuring electrical properties was presented. The products of radiation and radiation-thermal decomposition of water with the participation of radium-silicate systems were analyzed on chromatographs of the “Agilent-7890” and “Svet-102” brands. Experimental errors are given. The dose of absorption of gamma rays was determined by the sulfate-iron dosimetric method. Based on the electron densities of the systems under study and the reference dosimeter, the values of the dose absorbed by the radium-silicate composition were determined.

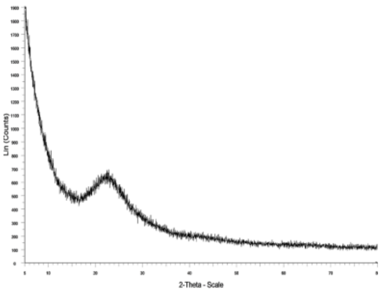

Figure 2. X-ray diffraction pattern of SiO_2 compound

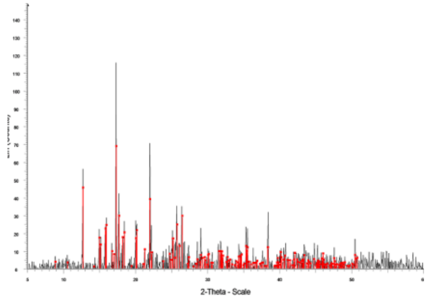

Figure 3. X-ray diffraction pattern of a radium silicate compound containing 10,1% radium.

For comparative studies, along with $(RaO)_x(SiO_2)_y$, pure compound SiO_2 was also synthesized and X-ray phase analysis of all synthesized samples was carried out. It is known from diffraction patterns that SiO_2 has an amorphous structure, a crystalline structure $(RaO)_x(SiO_2)_y$, and various water molecules are located at the interlayer levels.

İn the **third chapter** it is discussed about the nature of the radium isotope and the production of IR and DTA analyzes of the silicate and radium silicate system, mainly in the fission chain of the U^{238} isotope in nuclear materials:

$$U^{238} \xrightarrow{\alpha} Th^{234} \xrightarrow{\beta} \mathrm{Pr}^{234} \xrightarrow{\beta} U^{234} \xrightarrow{\alpha} Ra^{226} \qquad (4)$$

The resulting Ra^{226} isotope also has an unstable nucleus and undergoes alpha decay with a half-life of $T_{1/2}$ = 1600 years.

$$Ra^{226} \xrightarrow[5,4\%\gamma\ (T_{1/2}=1600\ years)]{94,6\%\alpha} Rn^{222} \xrightarrow[T_{1/2}=3,8235 days]{\alpha} Po^{218} \xrightarrow[T_{1/2}=3,11 min.]{\alpha} Pb^{214} \xrightarrow[T_{1/2}=26,8 min.]{\alpha} (5)$$

During the decay of the Ra^{226} isotope, α-particles with energy E_α = 4.7843 MeV are released with a yield of 94.45% and gamma-quanta with E_γ = 4.601 MeV with a yield of 5.55%. Rn^{222}, a daughter product of radium, undergoes alpha decay with a yield of 99.99% with energy E_α=5.46 MeV, half-life $T_{1/2}$ = 3.8235 days and gives the polonium isotope Po^{218}.

As can be seen, any system containing the Ra^{226} isotope will be energetically and, accordingly, electrically nonequilibrium. Therefore, in the processes of these changes in the electrophysical properties of such systems in contact with the environment, one should expect the presence of a nonequilibrium state.

Given that radium can affect the electrical conductivity of the hydrated layer and the aqueous layer formed upon contact with the environment, the change in these factors was initially studied in research facilities. The effect of radium on a hydrated coating of radium silicate was studied by IR-Fourier spectrometry and thermogravimetry. The effect of radium as a cation and energy source of radium on the properties of the silicate system was determined based on a comparative study with the properties of pure SiO_2.

The IR Fourier spectra of the initial SiO_2 and $(RaO)_x(SiO_2)_y$ systems are shown in figure 4. The observed absorption regions are identified by standards and results are given in table 2.

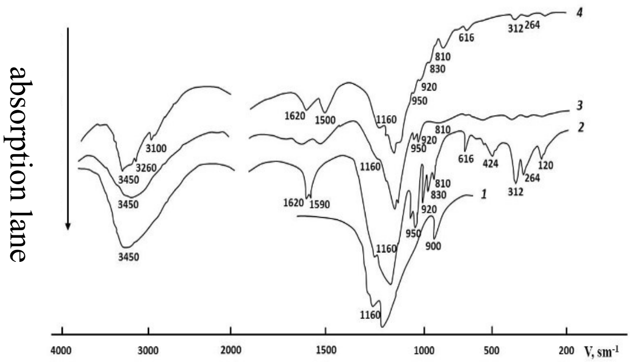

Figure 4. Infrared absorption spectra of silicon and radium-silicate samples containing different amounts of radium;(1)-SiO2;2 - radium-silicate sample containing 0,6% radium; (3) - radium-silicate sample containing 4,8% radium;(4)-radium silicate sample containing 10,1% radium.

Table 2.
Positions and intensities of absorption bands of silicates with silicate and radium groups

Literature results			Results from experience
Absorption spectrum, cm^{-1}	absorption spectrum, cm^{-1}	Valence [106,76]	absorption spectrum, cm^{-1}
3450	3450±10	ν(OH)	3450±10
1620	1650±5	δ(HOH)	1620±5
1580	1588	δ(HOH)	1590
1180	1180	$\nu(SiO_2)$	1180
955	963	$\nu(SiO_2)$	950
905	920±3	$\nu(RaO)\nu_3$	908±2
870	878	$\nu(SiO_2)$	880
830	835±4	$\nu(RaO)\nu_1$	830±2
809	812±1	$\nu(SiO_2)$	810±1
610	617	$\delta\ (SiO_2)$	616
500	520±10	$\delta\ (SiO_2)$	520±5
420	431±4	ν(RaO)	428
305	315±1	$\delta\ (SiO_2)$	312±2
268	278	$\delta(RaO)\nu_2$	266
-	-	ν(RaO)	128±1

As can be seen, with the introduction of radium, the intensity of the absorption bands associated with SiO_2 decreases and the number and intensity of absorption bands associated with the hydroxyl coating increase.

To characterize the effect of the radium-silicate system on the hydrate coating, the thermogravimetric method was used to study primary SiO_2 and the $(RaO)_x(SiO_2)_y$ system containing various amounts of radium. For example, the differential thermogram of the $(RaO)_x(SiO_2)_y$ system containing 10,1% radium is shown in Figure 5.

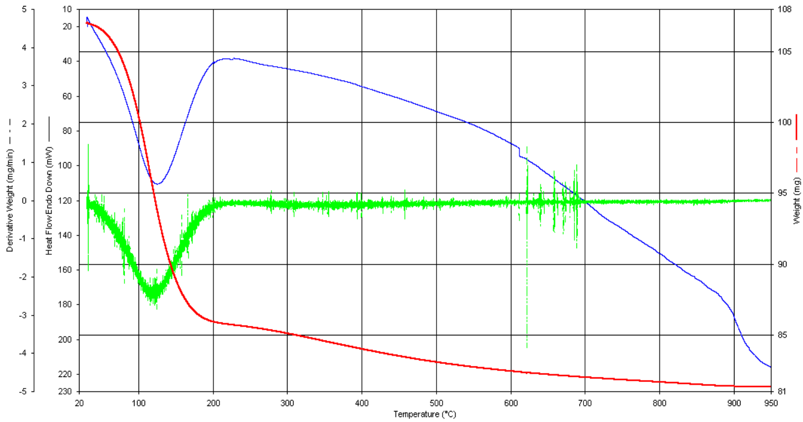

Figure 5. Differential thermogram of a radium-silicate system containing 10,1% radium

As it can be seen, in the DTA line, areas corresponding to various loss processes were observed in the TG line, which indicates extensive endopic and mass loss related to dehydration. In table 3 the mass loss in three temperature ranges found in the hydrated coating in $(RaO)_x(SiO_2)_y$ samples containing different amounts of radium are shown.

Table 3

Mass loss in non-irradiated $(RaO)_x(SiO_2)_y$ samples.

Sample's name	I group T≤280 °C	II group T≤300÷540°C	III group T≥650 °C
	Weight loss of samples depending on temperature, %		
Minimum activity $(RaO)_x(SiO_2)_y$	6	2	1
Medium activity $(RaO)_x(SiO_2)_y$	15	11	2
Maximum activity $(RaO)_x(SiO_2)_y$	20	17	2

As can be seen, with an increase in the amount of radium, the amount of hydrate coating increases. Subsequently, a sharp increase in the amount of adsorbed molecular water of various nature was observed.

Taking into account that the main goal of the work is to obtain hydrogen as a result of radiolytic splitting of water as a model radioactive compound of the $(RaO)_x(SiO_2)_y$ system, to study the regularities in the change in the absorption spectrum of gamma radiation with energy D ≤ 50 kGy of external gamma radiation on the hydration shell of the system (RaO)x(SiO_2)y is satisfied. On figure 6 shows the dependence of the mass loss in the hydrated layer on the doses of absorption of gamma-rays in the temperature ranges indicated in table 4.

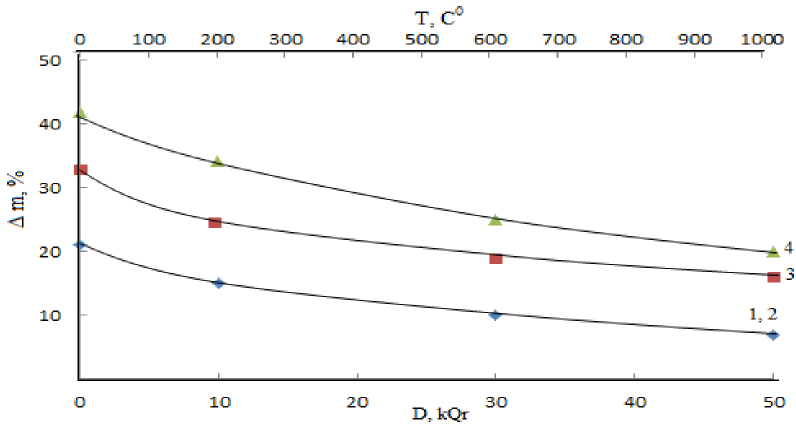

Figure 6. Dependence of mass loss on the absorption dose in the thermal analysis of radium-silicate compounds of different activity,
1) SiO_2 combination;
2) radium-silicate system with an activity of 260 Bq/g;
3) radium-silicate system with an activity of 2500 Bq/g;
4) radium-silicate system with an activity of 6100 Bq/g.

In table 4 the effect of gamma rays at a dose of D = 50 kGy on the desorbed hydrate cover in different regions are shown.

Table 4

Change in mass loss in radium silicate samples after irradiation

Sample's name	I group $T \leq 280$ °C	II group $T \leq 300 \div 540$ °C	III group $T \geq 650$ °C
	Weight loss of samples depending on temperature, %		
Minimum activity $(RaO)_x(SiO_2)_y$	4	2	1
Medium activity $(RaO)_x(SiO_2)_y$	11	4	3
Maximum activity $(RaO)_x(SiO_2)_y$	12	5	4

As can be seen, under the action of external gamma-rays, water molecules, mainly adsorbed at the molecular level, undergo strong radiolytic decay, and as a result of the interaction of decay products with structural fragments, an increase in the number of OH groups are observed. At high values of the radium content, the rate of decomposition of the hydrated coating is higher.

$$RaO\text{-}SiO_2\,(H_2O)_x \rightarrow Ra\text{-}SiO_2\text{-}OH(H_2O)_{x-1} + H \quad (6)$$

As can be seen from figure 6, the change in the hydrate cover under the action of gamma-radiation in samples with a low content of radium (A = 260 Bq/g) corresponds to SiO_2 within the error of determination. As the activity of radium in the $(RaO)_x(SiO_2)_y$ system increases, the amount of hydrate coating increases, and this increase is greater at A $\geq$ 5000 Bq/g. Thermograms were used to determine the enthalpy of dehydration processes of the $(RaO)_x(SiO_2)_y$ system with different activities. It has been determined that with an increase in the amount of radium, the enthalpy of dehydration processes increases within 45÷176 mC/mg. The rate of dehydration in the system $(RaO)_x(SiO_2)_y$ per 1 Gy of energy of external gamma radiation at constant 600 °C varies in the range of 0.43 ÷ 0.78% loss/Gy with an increase in radium activity in the range of 260 ÷ 6100 Bq/g. That is, radiation-thermal dehydration occurs faster in a highly active radium-silicate system.

As can be seen from the radium decomposition reaction, charged alpha particles with energies in the range E_α=4.7843 ÷ 5.5460 MeV and gamma-quanta with E_α=4.601 MeV are released in the $Ra^{226} \rightarrow Pb^{214}$ radioactive transformation chain, under their influence there is a high probability of obtaining charged particles in a silicate matrix. Therefore, the effect of both internal and external radiation on the electrical conductivity of the $(RaO)_x(SiO_2)_y$ system was studied. As can be seen, the $(RaO)_x(SiO_2)_y$ system is highly hygroscopic, and the hydrate coverage increases with increasing radium activity.

$$(RaO)_x(SiO_2)_y \cdot nH_2O \xrightarrow[\gamma]{\alpha} n + p + H_2O^+, OH^-, H^+ \quad (7)$$

Under the action of internal radiation, n-electron and p-hole charge carriers and a hydrate coating in the SiO_2 matrix are formed in the $(RaO)_x(SiO_2)_y \cdot nH_2O$ system, as shown in equation (7). The rate of these processes will increase as the activity of radium in the silicate system increases, and eventually the electrical conductivity will also increase. A decrease in the hydrate coverage during heating will lead to a decrease in the number of charge carriers in the system. The electrical conductivity of the $(RaO)_x(SiO_2)_y \cdot nH_2O$ system in the temperature range where dehydration occurs decreases with increasing temperature according to the pattern shown in figure 7.

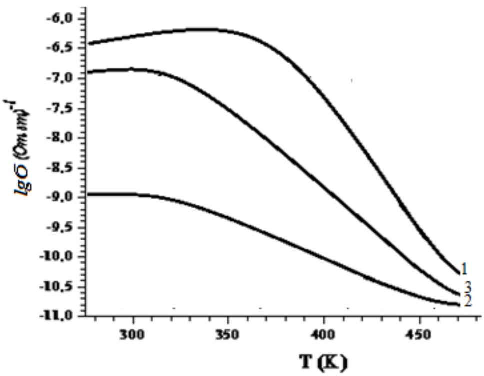

Figure 7. Temperature dependence of the electrical conductivity of the system $(RaO)_x(SiO_2)_y$+H_2O with A=6100 Bq/g; 1-initial, 2- after 1 hour, 3- after 24 hours.

Therefore, in the investigated temperature range (T= 300 ÷ 673 K), the dependence of $\delta = f(T)$ is different from the temperature dependence characteristic of dielectrics. Hydrated coated and measured at T ≈ 550 °C, dehydrated $(RaO)_x(SiO_2)_y$ dependences of electrical conductivity of the samples on the activity of radium in the sample content are given in figure 8.

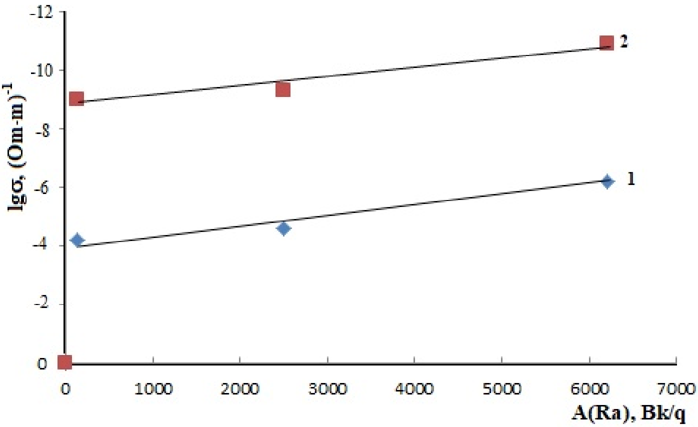

Figure 8. Dependence of electrical conductivity of $(RaO)_x(SiO_2)_y$ samples with hydrate coating (1) and dehydrated state (2) on the activity of radium in the samples, 1-T =300 K, 2-T = 550 K

As can be seen, as the activity of radium increases in $(RaO)_x(SiO_2)_y$ samples, the electrical conductivity increases linearly in both hydrated and dehydrated cases.

This shows that the decomposition products of radium in the $(RaO)_x(SiO_2)_y$ system create additional charge carriers in the system.

In order to characterize the generation of additional charge carriers under the influence of external gamma-radiation in the $(RaO)_x(SiO_2)_y$ system, the effect of gamma-radiation dose on the electrical conductivity of samples with different activity was studied. Figure 9 shows the dependence of the electrical conductivity of the $(RaO)_x(SiO_2)_y$ system on the dose of gamma-radiation.

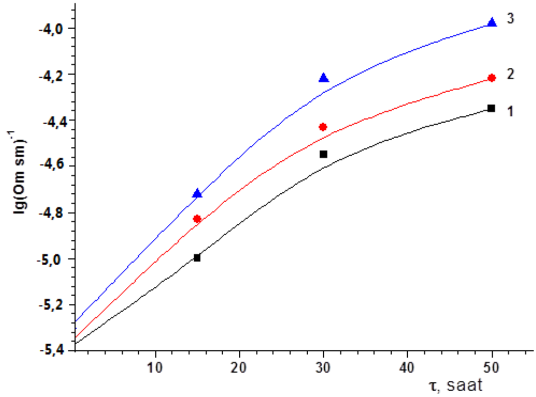

Figure 9. Dependences of the electrical conductivity of the $(RaO)_x(SiO_2)_y$ system at a temperature T= 300 K, D = 0.38 Gy/sec on the irradiation time. (1260 Bq/g, 2-2500 Bq/g, 3-6100 Bq/g).

As can be seen, the dependence lgδ = *f*(D) becomes linear in the initial region D≤10 kGy and becomes stationary in the region D ≈ 10–50 kGy. Based on the results obtained, it can be concluded that the rate of formation of charge carriers under the action of external gamma-rays in the $(RaO)_x(SiO_2)_y$ system and their stationary concentrations increase with increasing radium activity in the sample. Based on these results, it can be expected that the yield of molecular hydrogen will increase as the activity of radium increases as a result of the radiation-chemical decomposition of water with the participation of the $(RaO)_x(SiO_2)_y$ system within the existing mechanisms of radiation-chemical processes.

İn the fourth chapter initially, the energy that can be supplied to the system as a result of internal radiation in radium-containing silicate was evaluated. Radium is the daughter element of the U^{238} series and forms the radioactive gas radon primarily through alpha-decay. In radium-radon decay, the hundred-year equilibrium state is reached mainly over a period $\tau \geq 10T_{1/2}$. Experimental observations show that at times there is a tendency towards equilibrium.

$$Ra^{226} \rightarrow Rn^{222} + \alpha\ (94,6\ \%) + \gamma\ (5.4\%) \quad (8)$$

The dependence of the activity of Rn^{222} as a result of radioactive decay on this chain on the time of contact with the initial isotope can be determined by the following empirical expression.

$$A(Rn) = A(Ra)(1 - e^{\lambda\tau}) \qquad (9)$$

Here A(Ra) is the activity of the obtained radon, A(Ra) is the activity of Ra^{226} in the sample, τ is the observation time, and λ is the decay constant of Rn^{222}. As can be seen from this equation, at approximately $t_t = \frac{3}{4} T_{1/2}$, the activity of Rn^{222} is approximately equal to the activity of initial Ra^{226}. In experiments on the study of decomposition processes with the participation of a silicate system containing radium in the composition of water, it can be assumed that the retention time of the $(RaO)_x(SiO_2)_y + H_2O$ system corresponds to the radiation time, as well as the presence of an equilibrium state during the decay of radioactivity Ra → Rn inside ampoules. Under the experimental conditions, Rn^{222} decomposition processes proceed at the same time:

$$Rn^{222} \rightarrow Po^{218} + \alpha \qquad (10)$$

As a result, the emmonization of radon gas takes place inside the reactor. If we look at this order of expansion, then the century equilibrium condition can be applied as Po^{218}. So, if we look at line (5), the condition $T_{1/2}$ (mother) >> $T_{1/2}$ (daughter), which is the basis of the centenary balance, is fulfilled as soon as Po^{218} is formed. Therefore, we can write the following expression for the first three splits in accordance with the age equilibrium condition:

$$\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3 \qquad (11)$$

According to the century equilibrium condition, the amounts of Rn^{222} and Po^{218} present in each substance in the system $\tau >> T_{1/2}$ (Rn) >> $T_{1/2}$ (Po) after a certain time can be determined. For this, their radioactive decay coefficients can be determined based on the $T_{1/2}$ values of isotopes according to the following expression.

$$\lambda = \frac{0,693}{T_{1/2}} \qquad (12)$$

The radioactive decay constants and the number of radioactive isotopes present under experimental conditions are given in the table below.

Table 5
Amounts of isotopes in the reactor in case of century equilibrium.

Isotope name	A, Bq/g·sec	λ, sec^{-1}	N, atom/g
Mother isotope, Ra^{226}	6100	$1.356 \cdot 10^{-11}$	$4.5 \cdot 10^{14}$
Rn^{222}	6100	$2.9 \cdot 10^{-6}$	$2.1 \cdot 10^{9}$
Po^{218}	6100	$3.79 \cdot 10^{-3}$	$1.61 \cdot 10^{6}$

Since hydrogen, which is a reaction product during the radiation-catalytic decomposition of water, is calculated relative to a unit mass of the catalyst, the energy given to the catalyst can be calculated from the amount of decomposition of isotopes that are in equilibrium per unit time. The energy of alpha particles emitted into the system per unit time by primary Ra226 can be determined by the following expression:

$$E_r = A \cdot E_\alpha \quad (13)$$

Here, E_{rel} is the energy communicated by radium to silicates through alpha particles, E_α is the energy of each alpha particle, A is the activity.

Let's estimate the energy given for the maximum activity of the samples taken in the experiments A = 6100 Bq/g·sec.

$$E(Ra) = 6100 \cdot 4.77MeV = 2.92 \cdot 10^4 \text{ MeV/Gy·sec} \quad (14)$$

Accordingly, it is possible to estimate the energy transferred to the silicate by other isotopes.

$$E(Rn) = 6100 \cdot 5.59MeV = 3.41 \cdot 10^4 \text{ MeV/Gy·sec} \quad (15)$$

$$E(Po) = 6100 \cdot 5.46MeV = 3.33 \cdot 10^4 \text{ MeV/Gy·sec} \quad (16)$$

The energies of gamma and beta rays released during the alpha decay of these elements are relatively small, so they are not taken into account. Energy imparted within the silicate by conventional alpha active isotopes

$$\Sigma E_i(\alpha) = 2.92 \cdot 10^4 + 3.41 \cdot 10^4 + 3.33 \cdot 10^4 = 9.66 \cdot 10^4 \text{ MeV} \quad (17)$$

$$\Sigma E_i(\alpha) = 9.66 \cdot 10^{10} \text{ eV/Gy·sec} \quad (18)$$

Since in these cases they are energy carriers, ionization, excitation, and structural defects are more likely to occur when they interact with silicate systems. Intensity of external irradiation during the study

$\dot{D}$=0.15 Gy/sec ≈ $0.15\cdot10^2$ ˙$6.24\cdot10^{13}$ eV/Gy·sec=$0.936\cdot10^{15}$ eV/Gy·sec=$9.36\cdot10^{14}$ eV/Gy·sec (19)

However, considering that the energy exchange between the external gamma radiation and the element atoms included in the catalyst mainly occurs by Compton scattering, the role of internal radiation in the radiative heterogeneous decomposition of water should be expected. Therefore, in order to evaluate this effect and to clarify the role of alpha rays in the future processes of radiolysis of fragments, the kinetics of hydrogen production from gamma radiolysis at temperature T= 300 K in the presence of radium-containing silicate of water was studied. For this purpose, m ≈ $4\cdot10^{-2}$ g of radium-containing silicate in special ampoules was adsorbed in the water adsorption device corresponding to the vapor density ρ = 5 mg/cm^3 in the volume of the ampoule, by irradiating in a closed Co^{60} -isotope source at an intensity of $\dot{D}\approx$ 0.15 Gy/sec the kinetics of the process was studied at different times.

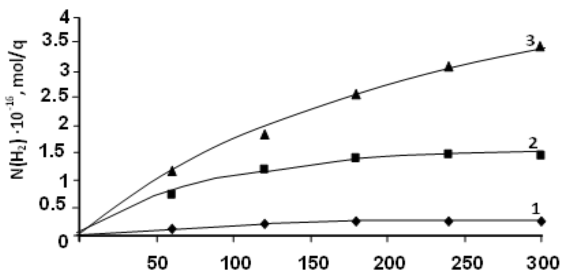

Figure 10. Kinetics of hydrogen formation during radiation-catalytic decomposition of water in the presence of $(RaO)_x(SiO_2)_y$ of different activity, T=300 K, $\dot{D}$=0.15 Gy/s, ρ=5 mg/cm^3, 1.A= 260 Bq/ G; 2. A=2500 Bq/g; 3. A= 6100 Bq/g,

Using a suitable technique, the kinetics of hydrogen formation during the radiation-catalytic
decomposition of water in the presence of a radium-containing silicate of various activity was studied. The radiation-chemical yield of molecular hydrogen production was calculated in two ways.

$$G_{gen}(H_2) = \frac{W(H_2)}{D_{cat} + D_{water}} \cdot 10^{-2} \qquad (20)$$

$$G_{ads}(H_2) = \frac{W(H_2)}{D_{water}} \cdot 10^{-2} \qquad (21)$$

The first quantity was calculated to evaluate the efficiency of general radiation-heterogeneous
processes, the useful work coefficient of energy conversion, and the second quantity to characterize the processes of energy transfer from the solid phase to the adsorbed phase.

The results obtained from the experimental results are given in table 6.

Table 6.

Results of kinetics of hydrogen formation during radiation-catalytic decomposition of water in the presence of $(RaO)_x(SiO_2)_y$ of different activity

Ra^{226} activity in $(RaO)_x(SiO_2)_y$ system, Bq/Gy·sec	$W(H_2)$, molecule. $Gy^{-1}.sec^{-1}$	$G_{ads}(H_2)$, molecule / 100 eV	$G_{gen}(H_2)$, molecule / 100 eV
260 ($\dot{D}$=0.5 Gy/sec)	$0.28\cdot10^{12}$	1.86	0.009
2500 ($\dot{D}$=0.5 Gy/sec)	$0.21\cdot10^{13}$	13.5	0.068
6100($\dot{D}$=0.5 Gy/sec)	$0.35\cdot10^{13}$	22.58	0.113
(SiO_2), ($\dot{D}$=0.28 Gy/sec)	$9.0\cdot10^{11}$	10	0.05
In homogeneous gamma-radiolysis of pure water, $G_{gen}(H_2)$, molecule/100 eV	-	0.45	-

As can be seen, the release of hydrogen during the radiolysis of water in the presence of primary silicate and radium-containing silicate is 4–50 times higher than during the radiolysis of pure water. This shows that the silicate and the radium-containing silicate have radiation-catalytic activity in the process of water radiolysis. As the activity of radium increases in the radium-containing silicate, the radiation-catalytic activity increases. This increase can be explained mainly for two reasons:

• creation of favorable conditions for energy transfer as a result of radium Ra^{+2} cation playing the role of a strong adsorption center for water molecules

• Acceleration of the decay process due to the additional energy provided by radium and its daughter fission isotopes

In the process of radiolysis of water with the presence of radium-containing silicate, as a result of both internal and external radiation, imbalanced charge carriers and excited states, which are the main energy carriers in typical dielectric silicate systems, are created (according to the 7th sechem).

As a result of the interaction of charge carriers and excited states formed by water molecules adsorbed on the surface level and migrated to the surface level, water molecules undergo disintegration according to the following schemes.

$$H_2O_s + p \rightarrow H_2O_s^+ \quad (22)$$
$$H_6O_s^+ + e \rightarrow H_2O_s^* \quad (23)$$
$$H_2O_s^* \rightarrow H + OH \quad (24)$$
$$H_2O_s + ex \rightarrow H_2O^* \rightarrow H + OH \quad (25)$$

At low temperatures, intermediate products are converted from hydrogen atoms to molecular hydrogen according to the following reactions.

$$H + H \rightarrow H_2 \quad (26)$$

At this time, most likely,

$$H + OH \rightarrow H_2O \quad (27)$$

the opposite process also takes place.

If we take into account that the threshold energy of formation of nonequilibrium charge carriers in silicate systems with a band gap corresponding to $E_g \approx 10$ eV is $E_h = 2 E_g$, their yield under the action of external gamma rays is G(n,p) ≈ 4-5 pairs /100 eV expected. According to the above scheme, the yield of hydrogen with the participation of the n,p pair should be $G(H_2) \approx 2–2.5$ molecules/100 eV. The observed values of $G_{gen}(H)$ show that the efficiency of internal and external processes of conversion of radiation energy into hydrogen during radiolysis of water in the presence of radium-containing silicates at T = 300 K is relatively low. One of the main reasons for this may be the emission of secondary electrons into the contact medium. To take this factor into account, we studied the kinetics of hydrogen formation during the radiolysis of a silicate system in the form of an emulsion in water. Research was carried out comparing primary SiO_2 and water adsorbed on the surface of the silicate with the highest activity of radium and emulsion (m=0.2 g H_2O). Based on the obtained kinetic curves, the kinetic parameters of the formation of molecular hydrogen in the mode of gamma radiation with an intensity of $\dot{D}$=0.28 Gy/s at T = 300 K were determined, which are given in Table. 7.

Table 7.

Comparative values of initial SiO2 with the highest activity radium-containing silicate adsorbed on the surface of water and in emission cases

Catalyst samples and the state of water under radiolysis conditions	$\dot{D}$ Gy/sec	T, K	$W(H_2)$, Molecule $.g^{-1}.sec^{-1}$	$G_{ads}(H_2)$, molecule / 100 eV	$G_{gen}H_2)$, molecule / 100 eV
Silicate + $H_2O_{ads.}$ (m_{H2O}= 5 $\cdot 10^{-3}$ g)	0.28	300	$9.1 \cdot 10^{11}$	10	0.005
Silicate + $H_2O_{emulsion}$ (m_{H2O}= 0.2 g)	0.28	300	$0.33 \cdot 10^{13}$	0.9	0.18
Radium containing silicate, A= 6100 Bq/g·sec + H_2O_{ads}. (mH_2O= 5 ·10-3 g)	0.28	300	$2.44 \cdot 10^{12}$	28	0.14
Radium containing silicate A= 6100 Bq/g·sec + $H_2O_{emulsion}$ (m_{H2O}= 0.2 g)	0.28	300	$6.9 \cdot 10^{12}$	2	0.40

Based on the results obtained, it is possible to evaluate the role of secondary electron emission emitted into the contact medium as a result of external radiation in the catalyst + water system and internal radiation in the heterogeneous radiolysis of water. In the silicate system, 3.6 times more energy is transferred to the contact medium by emitting electrons than the energy remaining in the volume and used at the surface level.

$$\frac{\text{G gen.emis.(H2)}}{\text{G gen.vol.(H2)}} = 3.6 \qquad (28)$$

During the radiolysis of water with the presence of radium-containing silicate (A = 6100 Bq/g), with the help of electrons, about 3 times more energy is emitted into the contact medium than the volume. For radium-containing silicate (A = 6100 Bq/g) and initially pure silicate, during radiolysis of water, the radiation-catalytic activity increases approximately 2.2-2.8 times due to internal irradiation.

In order to determine the role of non-equilibrium charge carriers in the radiolytic decomposition of water in the presence of a radium-containing silicate, the kinetics of the formation of paramagnetic centers as a result of irradiation with an intensity of $\dot{D}$=0.38 Gy/s at T=77K was studied using the EPR method. Electronic and hole centers and lines characteristic of H were observed in the EPR spectrum of samples of radium-containing silicate partially removed from the hydrate coating.

In the presence of water molecules, electron and hole centers on the surface, along with lines belonging to the OH group, a doublet characteristic of H atoms is also observed in the spectrum. In the presence of water molecules on the surface, the intensity of the line characteristic of hole centers decreases sharply and after a while becomes unobservable. The kinetics of the formation of hole centers during gamma irradiation of a radium-containing silicate at 77 K was studied and the value of the radiation-chemical yield G(DM)=0.31 particles/100 eV was determined.

Based on the results obtained during the action of gamma rays on the radium-containing silicate + water system, the following mechanism for obtaining paramagnetic particles was proposed.

$(RaO)_x(SiO_2)_y \rightarrow (RaO)_x(SiO_2)_y + n + p$ (29)

$n + L_A \rightarrow L_n$ — electronic center (30)

$P + L_D \rightarrow L_P$ — hole center (31)

$H_2O_s + p \rightarrow H_2O_s^+$ (32)

$H_2O_s^+ + e \rightarrow H_2O_s^* \rightarrow H + OH$ (33)

$H + S \rightarrow H_s$ — H atoms (34)

$OH + S \rightarrow OH_s$ — OH group (35)

In the fifth chapter the kinetic energy of nuclear fission fragments is mainly converted into thermal energy in the environment. Therefore, in the processes of radiolysis of fragments, it is necessary to take into account the temperature factor of the reaction medium. For this purpose, the influence of temperature on the processes of formation of molecular hydrogen during the radiolysis of water with the participation of a radium-containing silicate system in the range T=300-673K was studied.

It has been determined that, under statistical conditions, the process of obtaining molecular hydrogen by radiolytic decomposition of water in the presence of a radium-containing silicate in the presence of a constant amount of a catalyst depends mainly on such factors as temperature (T), dose radiation absorption (D) and vapor density in the reaction medium of water (ρ_{H2O} = mg/cm^3). The influence of the absorption dose at a constant gamma-ray intensity was characterized by the irradiation time and expressed in kinetic curves. The results obtained show that after a certain temperature, thermal catalytic processes of water decomposition proceed in the presence of spent catalysts (T ≥ 473K):

$$\text{Catalysis} + H_2O \xrightarrow{T \geq 473K} H + OH \quad (36)$$

Therefore, in the temperature range where the catalysts have thermocatalytic activity, the radiation-catalytic activity was determined by the difference in the rates of radiation-thermal and thermal processes carried out under the same conditions (Fig. 11).

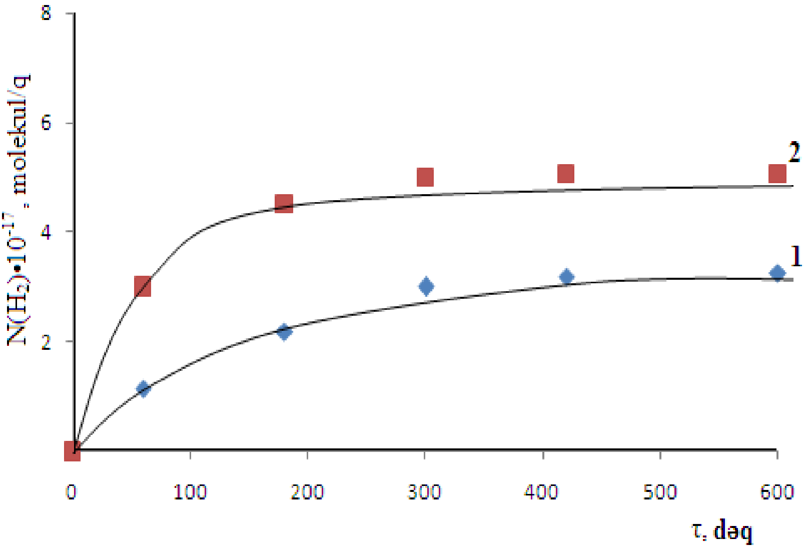

Figure 11. Kinetics of hydrogen generation as a result of radiation-thermal (2) and thermal (1) decomposition of water (H_2O vapor) in the medium $(RaO)_x(SiO_2)_y$, T=573K, ρ_{H2O}=5mg/cm^3, D=0.28Gy/sec

$$W_R(H_2)= W_{RT}(H_2) - W_T(H_2) \qquad (37)$$

Radiation-chemical yields of hydrogen were determined from the values of the rates of

radiation-catalytic processes. The effect of temperature on the formation of hydrogen during the radiation-catalytic decomposition of water in the presence of radium-containing silicates was studied on radium-containing silicate with activity A=6100 Bq/g. All kinetic parameters were determined based on the kinetic curves of the processes and are shown in table 8.

Table 8.
Effect of temperature on the formation of molecular hydrogen during the radiation-catalytic decomposition of water in the presence of SiO_2 and radium-containing SiO_2.

	Research objects	T, K	ρ_{H2O} mg/cm^3	$\dot{D}$ Gy/s	$W_{RT}(H_2)$, molecule. $g^1.sec^{-1}$	$W_T(H_2)$, molecule $g^{-1}.sec^{-1}$	$W_R(H_2)$, Molecule $g^{-1}.sec^{-1}$	$G_{ads}(H_2)$ molecule/ 100 eV	$G_{gen}(H_2)$ molecule/ 100 eV
1	$(RaO)_x(SiO_2)_y$ $+H_2O$ A=6100 Bq/g	373	5	0.28	-	-	$1.67\cdot10^{13}$	191	0.96
2	$(RaO)_x(SiO_2)_y$ $+H_2O$ A=6100 Bq/g	473	5	0.28	$5.5\cdot10^{13}$	$2.2\cdot10^{13}$	$3.3\cdot10^{13}$	379	1.89
3	$(RaO)_x(SiO_2)_y$ $+H_2O$ A=6100 Bq/g	573	5	0.28	$8.3\cdot10^{13}$	$2.5\cdot10^{13}$	$5.8\cdot10^{13}$	667	3.33
4	$(RaO)_x(SiO_2)_y$ $+H_2O$ A=6100 Bq/g	673	5	0.28	$1.03\cdot10^{14}$	$0.28\cdot10^{14}$	$7.5\cdot10^{13}$	862	4.31
5	SiO_2+H_2O	673	5	0.28	$3.8\cdot10^{14}$	$3.30\cdot10^{14}$	$5.0\cdot10^{13}$	560	2.8

As it can be seen, increasing the temperature in the region T=300÷673K increases the yield of hydrogen approximately 38 times. Considering that during thermoradiolysis of water at T ≥ 673 K, intermediate hydrogen atoms can be converted into hydrogen molecules by the following reaction, and the yield can reach $G_{hom}(H_2)$ = 8 molecules/100 eV, at T = 673 K G_{ads} (H_2) /G_{hom} We can conclude that (H_2)=108 times more:

$$H + H_2O \longrightarrow H_2 + OH \qquad (38)$$

In this table, for comparison, the kinetic parameters of obtaining molecular hydrogen from the radiation-catalytic decomposition of water, carried out under the same conditions in the presence of pure SiO_2, are given. As can be seen, the inclusion of the radioactive isotope Ra^{226} in the composition of the silicate increases the radiation-chemical yield of hydrogen production from the radiation-thermocatalytic splitting of water by ~1.5 times.

Considering that the temperature in the active zones of existing nuclear power reactors is close to T ≈ 673 K, the results obtained can be attributed to real processes of fragment radiolysis.

The influence of the water density in the reactor at T=373 K and T=473 K on the kinetic parameters of the process of obtaining molecular hydrogen from the radiation-thermocatalytic decomposition of water in the presence of silicates containing Ra^{226} has been studied. It was found that the radiation-chemical release of hydrogen occurs according to the Langmuir kinetics and is expressed by the following equation:

$$G(H_2) = \frac{kb\rho_{H2O}}{1+b\rho_{H2O}} \quad (39)$$

here k - is the rate constant of the process, b - is the adsorption equilibrium coefficient of water under the given conditions, and ρ_{H2O} is the density of water vapor in the reactor.

As a result, taking into account the electrical properties of the silicate system, a mechanism for the formation of energy carriers, the processes of localization, recombination and diffusion during heterogeneous radiolytic splitting of water under the action of external and internal radiation are proposed.

MAIN RESULTS

1. A silicate system containing Ra^{226} in various concentrations has been synthesized. The activity of radium in silicate systems synthesized by the gamma spectrometric method was determined, and the structural properties were studied. Ra^{226}, which is a part of silicate systems, its fission products and the energies released by them as a result of internal radiation processes under secular equilibrium conditions have been determined

2. Fourier-IR spectroscopy and derivatography were used to study the processes occurring in the hydrated coating of a radium-containing silicate system under the action of internal and external

gamma-radiation. It has been established that the activity of Ra^{226} in silicates sharply increases due to molecular water adsorbed in the excess hydration layer. Under the action of external gamma radiation in radium-containing silicates, violent processes of dehydration and decomposition occur. The processes of dissociation and desorption of adsorbed water molecules of various nature are stimulated by internal hydration and the formation of additional OH groups occurs.

3. It has been found that the electrical conductivity of hydrate coated silicate increases under the action of internal radiation caused by radioactive radium. With an increase in temperature, the electrical conductivity decreases as a result of dehydration processes and recombination processes of unbalanced charge carriers formed under the action of internal radiation, and becomes equal to the electrical conductivity of the silicate at $T \geq 550^0C$. Under the action of external radiation, the increase in electrical conductivity characteristic of the silicate system is stimulated by internal radiation.

4. Radium-containing silicate systems have radiation-catalytic activity in the radiolytic decomposition of water. The activity of Ra^{226} in silicates is already observed in their presence, the rate of hydrogen formation during the radiolytic splitting of water and an increase in radiation-chemical yields. This increase is due to the stimulating effect of internal exposure.

5. The role of the second electron beams emitted from a radium-containing silicate system under the action of external gamma-radiation in the processes of hydrogen formation during the radiolytic splitting of water is determined. It has been determined that the yield of hydrogen with the presence of electrons emitted into the contact medium during heterogeneous gamma-radiolysis of water with the presence of a silicate system with A=6100 Bq/Gy·sec is approximately 3-4 times higher than the yield of hydrogen obtained as a result of radiolytic decomposition at the surface level. That is, the main part of the second electron beam, formed when gamma rays act on a radioactive silicate system, radiates into the contact medium.

6. Regularities of the effect of temperature and water vapor density in the reaction medium on the process of obtaining hydrogen from heterogeneous radiolysis with the participation of the radioactive

silicate system of water were revealed. It was determined that the radium-containing silicate system has thermocatalytic activity in water splitting in the T≥473K temperature range, and the role of these processes increases as the temperature increases. With the increase of temperature in the range of T=300÷673K, the presence of silicate with activity Ra^{226} in the water content of A=6100 Bq/Gy·sec, the output of molecular hydrogen from thermo-radiation catalytic decomposition increases approximately ~30 times. When Ra^{226} with activity A=6100 Bq/Gy·sec is included in SiO_2, the yield of hydrogen from thermo-radiation catalytic decomposition of water at T=673K increases by ~1.5 times compared to non-radioactive SiO_2, which is due to the stimulating effect of internal radiation can be explained. Based on the obtained results, the mechanism of radiation- and radiation-thermocatalytic decomposition processes of water with the presence of radioactive silicates was given.

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