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Hydrogen Generation During Thermal Processes of Water Decomposition on the Surface of Nano-ZrO_2+3mol.%Y_2O_3

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Abstract

The study of physicochemical properties of nano-ZrO_2+3mol.%Y_2O_3 was determined. The X-ray diffraction spectrum of the nano-ZrO_2 and system nano-ZrO_2+3 mol.%Y_2O_3 compound was drawn by the Ritveld method and the crystal structure was determined at different temperatures (T = 373, 473, 573, 673 K) and under normal conditions. The energy of the arrangement of H_2 as a result of the decay of H_2O, on the surface of nano-ZrO_2+3mol.%Y_2O_3 was considered. Impacts of adsorption and desorption preparation of ZrO_2+3mol.%Y_2O_3 nanoparticles were considered at diverse (T = 373, 473, 573, 673 K) temperatures. The thought of H_2 in warm forms at nano-ZrO_2+3mol.%Y_2O_3 framework expanded. This tells about hydrogen era by water part in close future. Nano-ZrO_2 and system nano-ZrO_2+3mol.%Y_2O_3 with the temperature range of T = 373, 473, 573, 673 K have been studied by SEM analysis. These results are promising of hydrogen generation by water splitting in near future.

Keywords: Nano-ZrO_2+3mol.%Y_2O_3, Hydrogen generation, Kinetics, Thermal processes, Adsorption, desorption

Introduction

Recently, in various researches conducted by us and other researchers around the world, the study of products obtained from the radiolysis process of liquids, especially water in contact with metal or metal oxides under the influence of ionizing rays (γ-quanta, electrons, protons, neutrons, α-particles, high-energy ions, etc.) is of great importance both scientifically and energetically. The dependence of the radiation-chemical yield of the products obtained from the experiments on the particle size of metal or metal oxides (size effect), on their mass in suspended systems (mass effect) and on their type was observed. This effect is more pronounced in nanoscale metal or metal oxides. Now, special attention needs to be on production of new technologies, of zirconium dioxide, which is paid. Zirconium dioxide is used in metallurgy for receiving zirconium, which is applied in nuclear reactors as constructional material. The development of new electricity generation techniques is one of the most relevant tasks, especially nowadays under conditions of extreme growth in energy consumption. The exothermic heterogeneous electrochemical energy conversion to electric energy through the interaction of the ZrO_2 based on nanopowder system with atmospheric moisture, is one of the ways of electric energy obtaining.

Recently, due to the rapid development of industry, the demand for energy has increased sharply, and the use of nuclear energy [1] 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 [2-7].

In the last years, the awareness of climatic change has been increasing, leading to the tempting of exploration of alternative sources of energy. Therefore, the request for green energy is the chief objective for the scientists in 21 century. Among various kinds of safe, green and ecological energy, hydrogen is

measured as the best one, owing to its low cost and no toxicity to the environment. The richness of water on the earth is a big benefit for us to generate hydrogen fuel. Hydrogen is created from water by water splitting using dissimilar methods. The significant methods include photocatalytic, photo-electrochemical, thermal decomposition, and photo-biological radiolysis. Among these, photocatalytic of water is measured as the best one due to green, efficient, inexpensive with comfort process and with good amount of hydrogen formed [8-10].

For a long time, dissimilar groups of catalysts are being developed and utilized to split water in the light, but a good crop of hydrogen could not be attained at good scale. Nanocatalysts have been produced and utilized in water splitting with good achievements. Among these nano, zirconium oxide is measured as the best material due to its suitable band gap. Also, the gamma radiation irradiation of the water on the surface of nanocatalyst increases the production of hydrogen. Zirconium dioxide (ZrO_2) has unique properties of refractoriness, low volatility, high chemical resistance, mechanical strength, wear resistance, low thermal conductivity, wide band gap, oxygen conductivity, and high refractive index. Also, zirconium dioxide has complex polymorphisms, including high pressure phases. The unique properties have provided a wide and varied application of materials based on ZrO_2 in various fields of science and technology. Currently, the development of new technologies for the production and obtaining of nanodioxide zirconium is of particular importance. Nanoscale systems differ in many respects from ordinary single-crystal systems, therefore, the study of their interaction with water under the influence of γ-radiation is of great practical and scientific interest in the field of high-energy chemistry, as well as in solving environmental problems [11-15].

The formation of H_2 in the radiolysis of liquid water containing nanometer-sized ZrO_2 particles was found to be dependent on the crystalline structure of the particle. Zirconia particles of a few tens of nanometer in diameter may be formed with the tetragonal crystalline structure at room temperature rather than the more stable monoclinic form for bulk zirconia. Radiolysis of liquid water containing tetragonal ZrO_2 particles exhibits a significant increase in the decomposition of water to H_2 as compared to the monoclinic form. Annealing the tetragonal particles to the monoclinic structure results in the loss of excess H_2 production above that found with water alone. The results showed that surface morphology is extremely important in the decomposition of liquids at solid interfaces, which may have many consequences ranging from nuclear waste storage to the H_2 economy. The presented work is devoted to the kinetics and mechanism of the formation of hydrogen as a result of the decomposition of water on the surface of nano-ZrO_2. It is known that the characteristic incorporation of hydrogen era shapes in radiation-heterogeneous systems is the alteration of warm and ionizing radiation imperativeness into more effective shapes in solid materials and, at final, hydrogen, which is the foremost essentialness carrier in physicochemical shapes. Nano assistant materials have a made surface and extended distortion ness on the boundary of particles, which is of unimaginable noteworthiness in radiation-heterogeneous shapes with their back, as well as in the midst of the progression of significantly tricky locators of ionizing radiation. To alter physicochemical properties, materials based on the blends of nano-measured oxides are created [16-25].

Radiation-chemical yields of molecular products (H_2, O_2, H_2O_2, etc.) from the radiation-heterogeneous decomposition of water by metals or metal oxides used in each of these research methods are varies depending on their type, band-gap, particle size, saturation degree of adsorbed water on the particle surface, the temperature of the general system, the strength of the absorption dose, and the mass of metal or metal oxides suspended in the water [26-32].

In the presented work, molecular hydrogen obtained from thermocatalytic processes and its formation rate were studied in the nano-ZrO_2+mol.3%Y_2O_3 system under the influence of different temperatures (T = 373, 473, 573, 673 K).

Materials and methods

For the production of nano-ZrO_2+mol.3%Y_2O_3 used chemical technology of local sedimentation using physical effects [28]. Firstly, hydrated zirconium hydroxide was precipitated from a chloride raw material. Then, the obtained material undergoes dehydration in a specialized microwave oven (T = 120 °C, t = 0, 4 h). After which, the amorphous powder was subjected to crystallization annealing at a temperature of 400 °C for 2 h.

The X-rays were done in the X-Ray Diffractometer D2 PHASER (model number-"Bruker D8 Advance") developed. For this reason, the scattered nanoparticles were arranged. These nano-particular chips were set within the goniometer of the diffractometer and the X-ray diffraction range of the test was

drawn within the extent of diffraction point 20 < 2θ < 100. The cage parameters are calculated based on the square equations of crystallography.

Microscopic Analysis (SEM): The number of active centers formed on the surface of the catalyst and the number of elements concentrated in them were studied using an electron microscope brand SEM (Scanning Electron Microscope, Zeiss, SIGMA / VP).

Analysis gases: The amount of the obtained molecular hydrogen was analyzed on the "Agilent-7890" chromatograph.: Column: Carboxen-1010 PLOT, 30 m × 0.53 mm I.D. (25467)
Detector - TCD, "Constant Flow", Flow-3 mL / min,
20Hz / 0.01 min, T (head) = 50 °C, T (detector) = 230 °C
Makeup Flow = 0 mL / min,
gas carrier - Ar
oven: 50 °C (7.0 min), 20 °C / min to 230 °C
injector temperature.: 230 °C

Investigation of hydrogen and hydrogen-containing gasses in a vacuum adsorption gadget was carried out beneath static conditions. Exploratory thoughts about it were performed in uncommon quartz ampoules with a volume of V = 1 cm^3 beneath static conditions. A sample of 10 nm nano-ZrO2+mol.3%Y2O3 was taken as the object of research. Thermo-vacuum processing of samples is carried out with a zeolite pump at T = 300 K, P = 10^{-3} Pa for 2 h. The gases generated were inhaled from each adsorbed and desorbed ampoule to the chromatograph directly.

Results and discussion

The results of the XRD analysis are shown in **Figure 1**. **Figure 1** shows the X-ray diffraction spectrum of the nano-ZrO_2 and nano- ZrO_2+3mol.%Y_2O_3 system compound at different temperatures (T = 373, 473, 573, 673 K) and normal conditions. Analysis of the X-ray diffraction spectrum by the Ritveld method revealed the crystal structure of the nano-ZrO_2+3mol.%Y_2O_3 system compound corresponds to the tetragonal symmetry P42/nmc, A = 3,6 Å; C = 5,18 Å; V = 67,33; Z = 2-phase group (T = 373 K), monoclinic symmetry P_{21}/A, A = 5,31 Å; B = 5,2125 Å; C = 5,1471 Å; β = 99,218, Z = 4- phase group (T = 473 K), monoclinic symmetry P_{21}/A, A = 5,31 Å; B = 5,2125 Å; C = 5,1471 Å; β = 99,218, Z = 4-phase group (T = 573 K), monoclinic symmetry P_{21}/A, A = 5,31 Å; B = 5,2125 Å; C= 5,1471 Å; β = 99,218, Z = 4-phase group (T = 673 K).

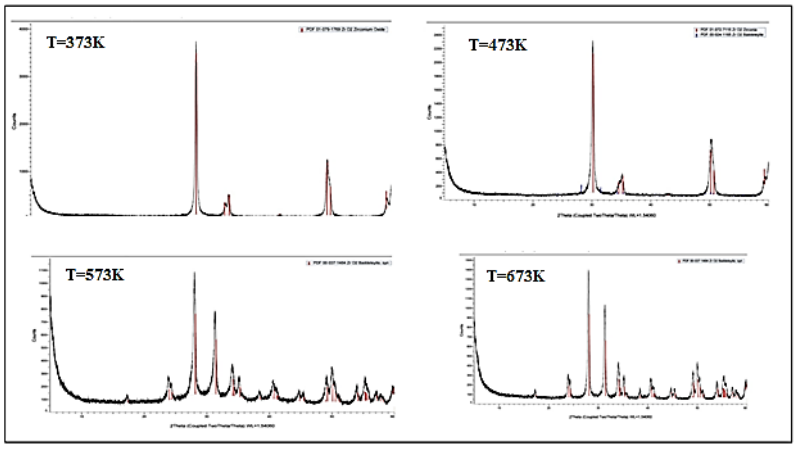

Figure 1 X-ray diffraction spectrum of the nano- ZrO_2+3mol.%Y_2O_3 system compound taken at different temperatures (T = 373, 473, 573, 673 K).

Microscopic Analysis (SEM)

SEM analysis of the nano-ZrO_2+3mol.%Y_2O_3 system compound at different temperatures (T = 373, 473, 573, 673 K) are shown in **Figure 2**. **Figure 2(a)** shows a surface description of nano- ZrO_2+3mol.% Y_2O_3 system. **Figure 2(b)** shows the corresponding energy spectra of nano-ZrO_2+3mol.%Y_2O_3 system and examines the elemental composition of the sample taken. It was found that the purity of nano-ZrO_2 is high (97,10 % (nanoZrO_2) + 2,90 % (3mol.%Y_2O_3)), (T = 373 K), nano-ZrO_2 is high (96, 90 % (nano-ZrO_2) + 3,10 % (3mol.%Y_2O_3), (473 K)), nano-ZrO_2 is high (96, 70 % (nano-ZrO_2)+ 3,30 % (3mol.% Y_2O_3), (T = 573 K)), nano-ZrO_2 is high (97, 00 % (nano-ZrO_2) + 3,00 % (3mol.%Y_2O_3), (T = 673 K)).

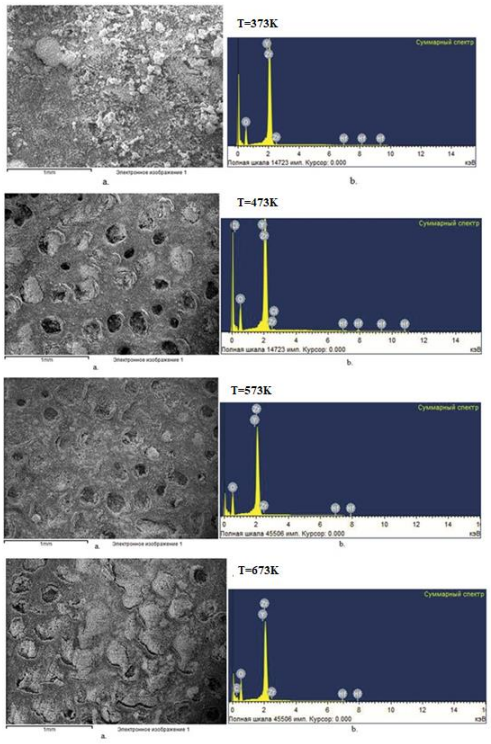

Figure 2 Scanning electron microscopy images (a) and corresponding energy spectra (b) with nano-ZrO_2+3mol.%Y_2O_3 system compound taken at different temperatures (T = 373, 473, 573, 673 K).

Molecular hydrogen generation

In this work for the purpose of identification of nano-ZrO_2+mol.3%Y_2O_3 system influence on the water surface, the kinetics of accumulation of molecular hydrogen at thermal processes of the at different temperatures T = 373, 473, 573, 673 K is investigated. **Figure 3** shows both: adsorption and desorption processes have been carried out in the nano-ZrO_2+mol.3%Y_2O_3 system and kinetic curves have been constructed.

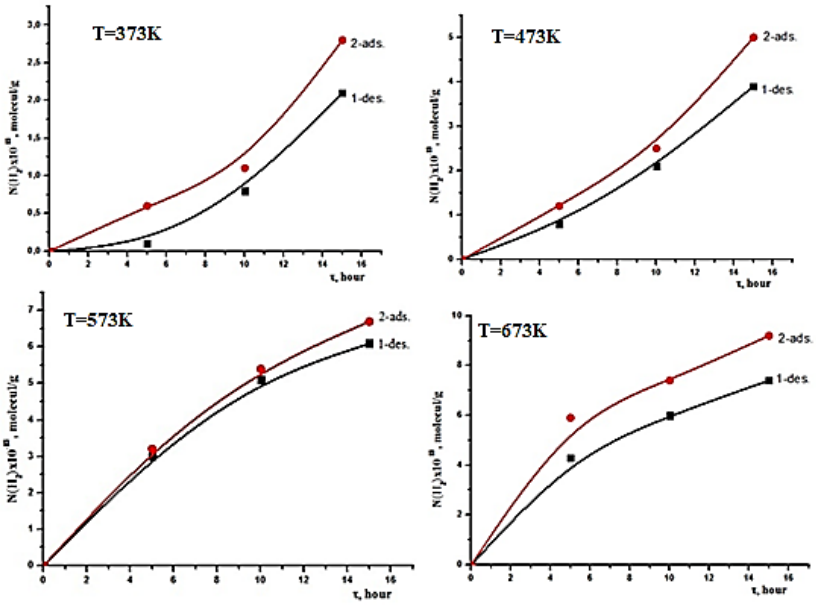

Figure 3 Kinetics of molecular hydrogen formation as a result of molecular thermal processes of adsorbed and desorbed water on nano-ZrO_2+mol.3%Y_2O_3 system, at different temperatures (T = 373, 473, 573, 673 K).

The velocity of hydrogen was determined in the investigated system based on the values obtained from the kinetic curves. The same results are given in **Table 1**.

Table 1 The velocity of hydrogen was determined in the investigated system based on the values obtained of the kinetic curves (nano-ZrO_2+mol.3%Y_2O_3) at different temperatures (T = 373, 473, 573, 673 K).

N	**nano-ZrO_2+mol.3%Y_2O_3**					
	Sample weight (g)	**Particle size, (nm)**	**Temperature (K)**	**Hour (τ)**	**The velocity of the process, $W_T(H_2)\cdot10^{18}$, molecule/g**	
					desorption	**adsorption**
1	$1\cdot10^{-3}$	10	373	5 10 15	0,11	0,7
2			473	5 10 15	0,23	1,3
3			573	5 10 15	0,5	1,5
4			673	5 10 15	0,6	1,8

The values of adsorption and desorption processes carried out in a thermal process at different temperatures (T = 373, 473, 573, 673 K) are shown in **Table 1**. **Table 1** shows that the values obtained in both processes are the values formed by the formation of active centers on the surface.

Depending on their kinetic energy, the Compton electrons pass from the nanoparticle several times into the liquid phase or vice versa, gradually losing their kinetic energies in both elastic and inelastic collisions and becoming thermal electrons in nano-ZrO_2+3%mol.Y_2O_3 system.

Mechanism

The study of structure-activity relationship of adsorbents plays a significant component in the adsorption and desorption process. The adsorption affinity between the adsorbent and the adsorbate determines adsorption and desorption behavior. Under the influence of temperature, positively charged ions are formed in the catalyst as a result of the adsorption of water and the transfer of charges to water molecules. These positively charged ions cause the water molecules to disintegrate as a result of their recombination with the electrons formed on the surface. In this process, the production of hydrogen is determined by the cost of electrons and charges formed on the surface. As the temperature increases, the mobility of the particles on the surface of the catalysts increases. On the other hand, since part of the electron-hole pair involved in the decomposition of water is regenerated, these induced particles (by temperature) participate in the decomposition of water. Thus, the production of hydrogen at higher temperatures is greater.

On the other hand, the Compton electrons shaped the interior of nanoparticle beneath the impact of light, and with each unused era of δ-electrons they make steadily lose their dynamic energies in versatile and inelastic collisions interior of the molecule, a few are captured by basic surrenders interior of the molecule and a few are transported to the molecule surface. The electrons thermal from the surface of the strong to the fluid stage slowly lose their active vitality by versatile and inelastic collisions in water and firstly change over into warm electrons, and after that can be rescued within the water:

$$\left(e^- + nH_2O \rightarrow e_{aq}^-\right) \quad (1)$$

The production of molecular hydrogen (1−3) by the thermocatalytic decomposition between salvaged electrons $\left(e_{aq}^-\right)$ and water molecules, as well as protonated water molecules (H_3O^+) in the intergranular liquid phase can be described as follows [4,5-7]:

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

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

$$e_{aq}^{-}+H_3O^{+} \rightarrow H+H_2O \quad (4)$$

$$H+H \rightarrow H_2 \quad (5)$$

Here it becomes clear that 1 pair of electron-hole pairs or 2 excitons are used to obtain 1 molecule of hydrogen. Reactions (3−4, 5) play a major role in the production of molecular hydrogen from the thermocatalytic decomposition of water adsorbed on the surface of nano-ZrO_2 under the influence of high temperature. Here in this study, the results of exploring the prospects of the hydrogen economy are presented. It is shown that the comparative analysis of the economic competitiveness of these chains with each other with solutions based on the use of alternative fuels has been performed, respectively. The analysis has established the most promising directions in the development of the hydrogen economy in order to justify the economic value of this research.

Future perspective of thermal process's role in hydrogen generation

The literature is a witness for many publications on water splitting for hydrogen generation but still, there is no method, which is feasible at a large scale economically. It means that there is a further need to do more research on water-splitting to explore the practical and feasible methods. It means that there is a great demand for research to explore the utility of thermal processes in water splitting. Consequently, there is a good call for complete research on water splitting; particularly on optimization to attain an extreme amount of thermal processes hydrogen production. The amount of hydrogen production may be increased by condensed thermal processes under the constant string. The future is very positive for water splitting by using different temperatures.

Conclusions

This study is the X-ray diffraction spectrum of the nano-ZrO_2+3mol.%Y_2O_3 system compound at different temperatures (T = 373, 473, 573, 673 K). The surface images of nano-ZrO_2+3mol.%Y_2O_3 system and the corresponding energy spectra were captured at different temperatures (T = 373, 473, 573, 673 K) by the SEM method. This study the corresponding energy spectra of nano-ZrO_2 and examines the elemental composition of the sample taken. In this process, water molecules are absorbed on the surface of nano-ZrO_2+3%mol.Y_2O_3. This showed when nano-ZrO_2+3%mol.Y_2O_3 was covered with water, the energy carriers (electrons, holes, excited states-exactions) formed under the influence of thermal effect in the same system carried processes. The checking of warm impact started changes inside the surface and the choice of the execution characteristic of a heat-resistant catalytic texture based on these nanomaterials. The changes in their physical and chemical properties made it conceivable to anticipate the working modes of the catalytic materials. Nano-ZrO_2+3mol.%Y_2O_3 system with nano-size opens a new actual direction in thermocatalytic processes and the thermoactive study of materials. It is clear from the presented results that nanostructured materials become an important role in nuclear-power engineering as structural and functional materials practically in all stages of the nuclear fuel cycle. It is extremely important for the formation of nanostructured materials after exposure to ordered nanostructure from the new phase with a period of a few nanometers, promoting the preservation of the properties of materials under high temperatures.

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