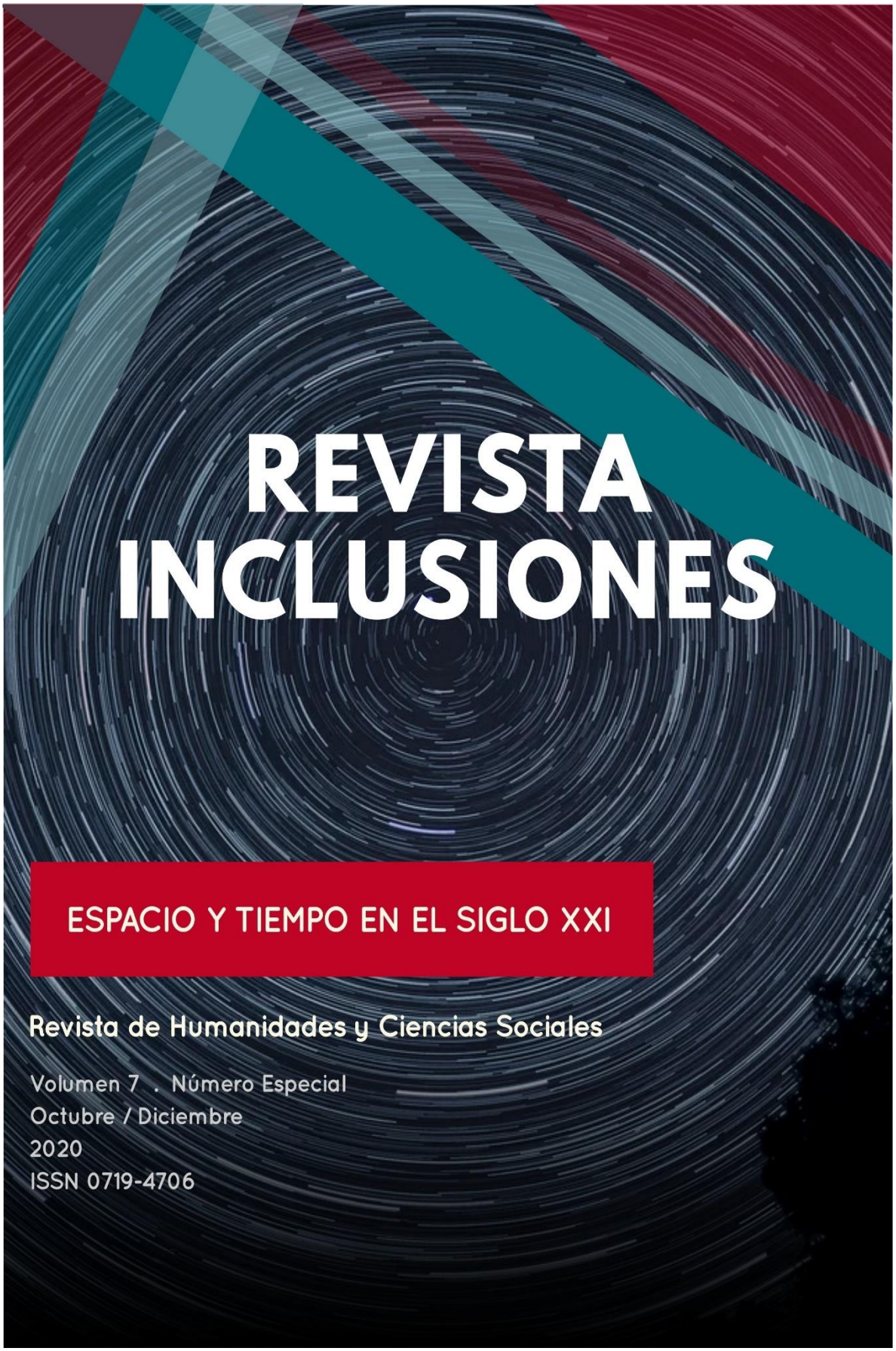
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LOW-TEMPERATURE PLASMA DECOMPOSITION OF LIQUIDS UNDER ULTRASONIC ACTION

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Abstract

In this work, a low-temperature plasma initiated in liquid media between electrodes has been shown to be able to decompose hydrogen containing organic molecules leading to obtaining gaseous products with volume part of hydrogen higher than 90% (up to gas chromatography data). Preliminary evaluations of energetic efficiency, calculated from combustion energy of hydrogen and initial liquids and electrical energy consumption have demonstrated the efficiency about 60-70% depending on initial liquids composition. Theoretical calculations of voltage and current values for this process have been done, that is in good agreement with experimental data.

Keywords

Hydrogen – Plasma – Ultrasonic cavitation

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Introduction

In our previous work, it has been shown that in intensive ultrasonic field above the cavitation threshold in the liquid a new form of electrical discharge with volumetric glow between the electrodes and increasing the current - voltage characteristic similar to that of abnormal glow discharge in a gas can occur[1].

Such kind of a discharge with large surface area of microbubbles may be of substantial interest in view of creation of new plasma chemical processes since large surface area between plasma and liquid may lead to increase of the diffusion fluxes of reactive plasma particles from a liquid. In this discharge, it can be possible to carry out a number of interesting chemical reactions[2].

Preliminary experiments have shown that as a result of acoustoplasma reactions in liquid hydrocarbons, there are chemical conversions in the liquid phase and also solid phase carbon and the hydrogen-containing gas are formed.

Physical and chemical processes in acoustoplasma

Physical principle of the approach is the decomposition of complex hydrogen-containing molecules in plasma and their ionization followed by recombination with the formation of simple molecules: H2, H2O, C, CO2, MOX, where M - the electrode material.

Plasma discharge initiated in the reactor between metal and graphite electrodes is powered by a specially designed source of DC or AC voltage, which allows to investigate the effect of the plasma characteristics on the reaction rate and the chemical composition of its products.

Optical spectroscopy observations allowed to evaluate the composition of reaction mixture (Fig. 1).

[1] N. A. Bulychev y M. A. Kazaryan, “Optical Properties of Zinc Oxide Nanoparticles Synthesized in Plasma Discharge in Liquid under Ultrasonic Cavitation”. Proceedings of SPIE, Vol: 11322 (2019): article 1132219.

[2] N. A. Bulychev; M. I. Danilkin; N. Yu. Vereshchagina y M. A. Kazaryan, “Luminescent Properties of ZnO Nanoparticles Doped by W Obtained in Plasma Discharge in Liquid under Ultrasonic Cavitation”. Proceedings of SPIE, Vol: 11322 (2019): article 113221S y N. A. Bulychev; A. I. Erokhin y M. A. Kazaryan, “A Comparative Study of Anti-Stokes Shift under Stimulated Rayleigh-Mie Scattering in Suspensions of Ag Nanoparticles Obtained in Plasma Discharge in Liquid under Ultrasonic Cavitation”. Proceedings of SPIE, Vol: 11322 (2019): article 113222G.

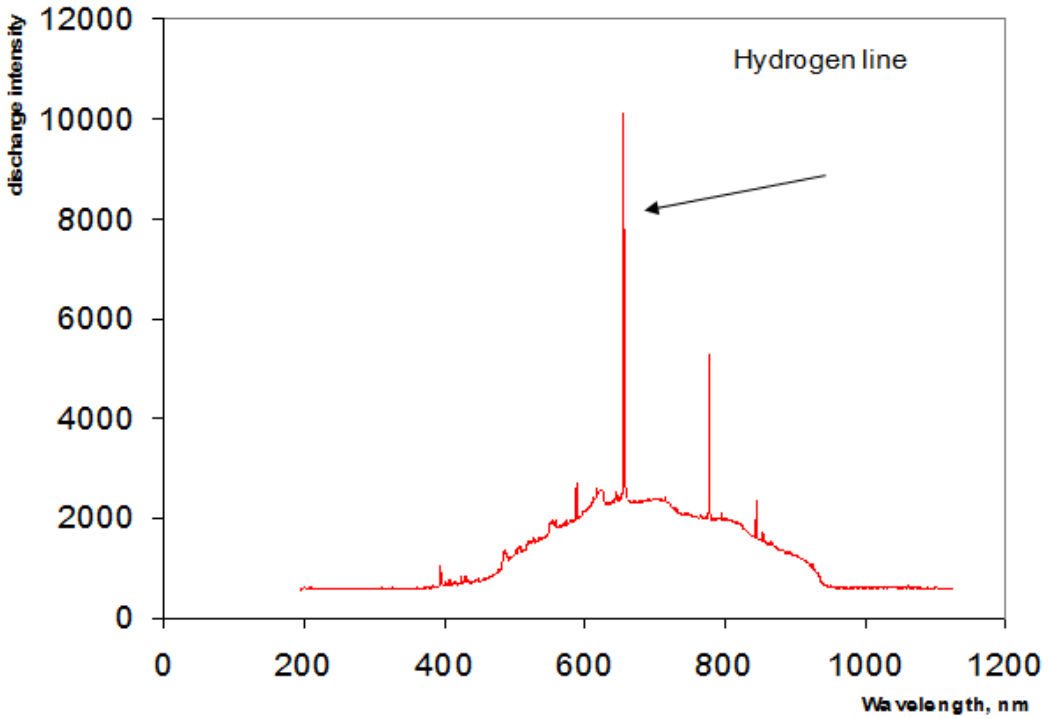

Figure 1
Optical spectra of plasma discharge in water

The analysis of the gaseous products of reactions in acoustoplasma discharge in liquid media confirmed the optical spectroscopy data and revealed that the main product gas was hydrogen. Therefore, the possibility of hydrogen synthesis from different liquids using acoustoplasma discharge was studied. This was carried out by measuring the current and voltage discharge, quantity of gas formed and the gas composition analyzed by gas chromatography[3]. The values of current and discharge voltage were necessary for calculation of the amount of energy consumed in the decomposition of the initial liquid and, finally, to calculate the amount of energy consumed per weight unit of hydrogen produced[4].

[3] N. A. Bulychev y M. A. Kazaryan, "Application of Optical Spectroscopy for Study of Hydrogen Synthesis in Plasma Discharge in Liquid under Ultrasonic Cavitation". Proceedings of SPIE, Vol: 11322 (2019): article 113221A; M. N. Kirichenko; L. L. Chaikov; I. S. Burkhanov; N. A. Bulychev y M. A. Kazaryan, "Interaction of aluminum oxide nanoparticles with human blood plasma thrombin (according to light scattering)". Proceedings of SPIE, Vol: 11322 (2019): article 113221Y y M. N. Kirichenko; L. L. Chaikov; I. S. Burkhanov; N. A. Bulychev y M. A. Kazaryan, "Effect of the pH of iron oxide nanoparticles solution on the rate of fibrin gel formation (according to light scattering data)". Proceedings of SPIE, Vol: 11322 (2019): article 113221E.

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Results of the gas mixture analysis demonstrate that the decomposition of water in acoustoplasma discharge leads to formation of almost pure hydrogen (98%) (Fig. 2); decomposition of organic liquids produces gas that also comprises carbon oxides, but their concentration does not exceed 5-6%, as a major amount of carbon during the decomposition of organic liquids forms solid particles[5].

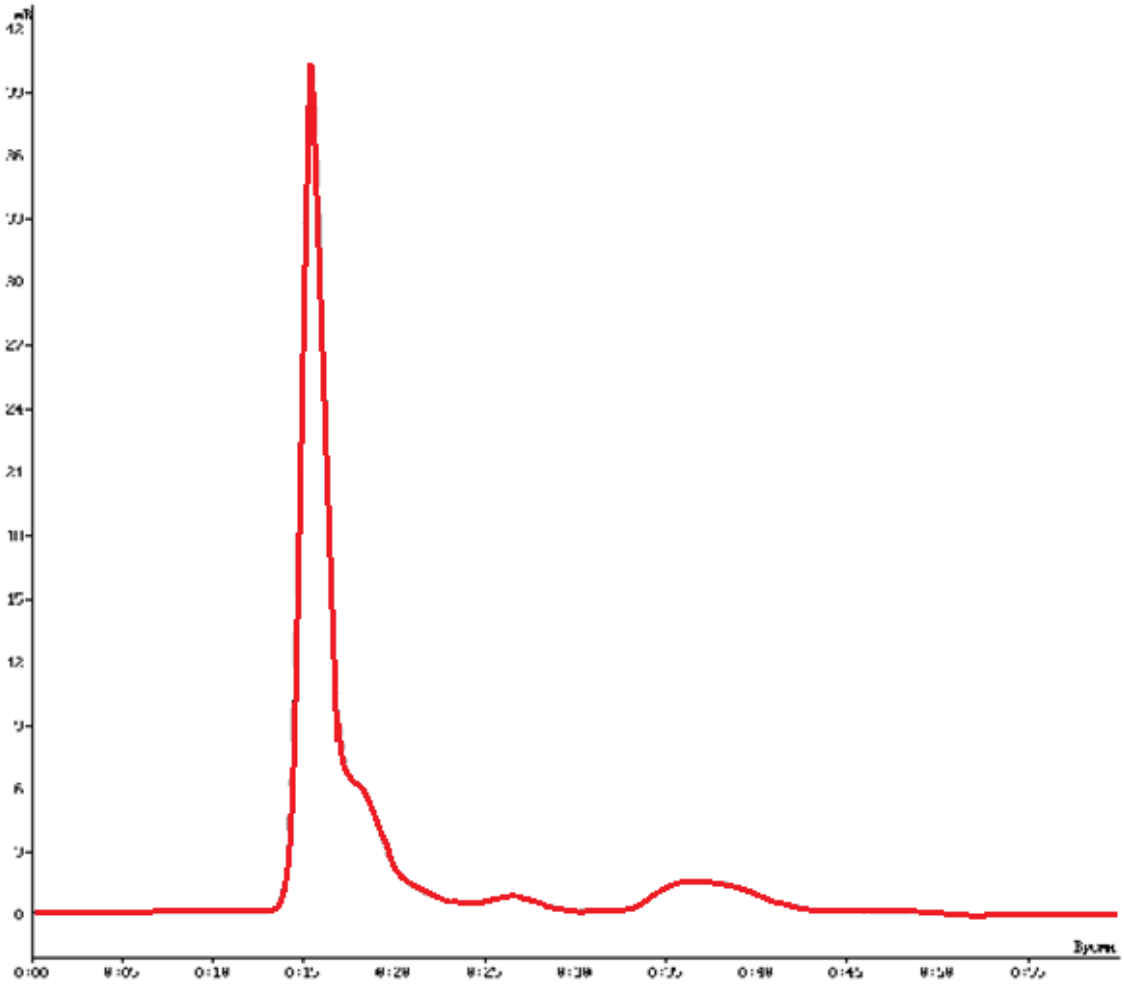

Figure 2
Gas chromatography analysis of gas mixture composition obtained from ethanol in plasma. Main peak belongs to hydrogen.

The following raw materials were used: water, alcohols, hydrocarbons and their mixtures (Table 1). Initiation of plasma discharge in these media leads also to formation of solid products: carbon nanoparticles and metal oxide nanoparticles from electrode material. The results of analyzes and stechiometric calculations of the formation of these by-products allowed to conclude that most of oxygen and carbon contained in the molecules of the original liquid consumed for nanoparticles, thus forming the gaseous mixture substantially enriched with hydrogen.

[5] M. N. Kirichenko; N. A. Bulychev; L. L. Chaikov; M. A. Kazaryan y A. V. Masalov, “Effect of iron oxide nanoparticles on the concentration-versus-sizes relation of proteins in the blood plasma and serum, and in model solutions”. Proceedings of SPIE, Vol: 10614 (2018): article 10614OM.

Feedstock	Concentration of H_2 in outlet gas %	Production L/min
Water	98	0,5
Hexane	85	1
Toluene	80	1
Ethanol/ water 50:50	95	2
Isopropyl alcohol/ water 50:50	93	1,7
Emulsion bitumen/ water	80	1,5

Table 1
Results of acoustoplasma reactions in various media

The amount of gas mixture obtained by the decomposition of organic liquids indicates that the process is strongly dependent on the discharge current, and the volume of the discharge zone varying depending on the distance between the electrodes in the reaction chamber. In the experiments, the discharge current was from 4A to 8A, the discharge voltage depending on the type of the liquid was 30-45 V.

Other product of the acoustoplasma decomposition of organic liquids is the solid carbon in the form of agglomerates of nanoparticles of various structures. It precipitates during the reaction at the bottom of the reaction chamber[6]. Analysis of these nanoparticles by scanning and transmission electron microscopy showed that carbon fibers, nanotubes, plates etc. can be synthesized by acoustoplasma[7].

[6] N. A. Bulychev; M. A. Kazaryan; A. D. Kudryavtseva; M. V. Kuznetsova; T. F. Limonova; N. V. Tcherniega y K. I. Zemskov, “Anti-Stokes luminescence in nanoscale systems”. Proceedings of SPIE, Vol: 10614 (2018): article 106140N; A. S. Averyushkin; A. N. Baranov; N. A. Bulychev; A. D. Kudryavtseva; M. A. Strokov; N. V. Tcherniega y K. I. Zemskov, “Stimulated low-frequency Raman scattering in aqueous suspension of nanoparticles”. Proceedings of SPIE, Vol: 10614 (2018): article 106140K y A. S. Averyushkin; A. N. Baranov; N. A. Bulychev; A. I. Erokhin y M. A. Kazaryan, “Ag nanoparticles suspensions for stimulated Rayleigh backscattering of single frequency 0.5 micron pulsed laser radiation”. Proceedings of SPIE, Vol: 10614 (2018): article 106141L.

[7] Y. A. Dyakov; M. A. Kazaryan; M. G. Golubkov; D. P. Gubanova; N. A. Bulychev y S. M. Kazaryan, “Laser-induced dissociation processes of protonated glucose: dehydration reactions vs cross-ring dissociation”. Proceedings of SPIE, Vol: 10614 (2018): article 1061417; A. A. Asratyan; S. A. Ambrozevich; O. S. Andrienko; N. A. Bulychev; A. G. Grigoryants; M. A. Kazaryan; S. M. Kazaryan; N. A. Lyabin; R. G. Mkhitaryan; G. A. Tonoyan; I. N. Shiganov y V. I. Sachkov, “Comparative analysis of parameters of pulsed copper vapour laser and known types of technological lasers”. Proceedings of SPIE, Vol: 10614 (2018): article 1061402 y K. V. Pushkin; S. D. Sevruk; N. S. Okorokova y A. A. Farmakovskaya, “The most efficient corrosion inhibitors for aluminum anode of electrochemical cell used as a controlled hydrogen generator”. Periodico Tche Quimica, Vol: 15 num 1 (2018): 414-425.

The obtained nanoparticles and the agglomerates may also be applied as fillers, colorants, components, composite materials[8]. Theoretical explanation as well as examination of such materials is referred[9].

Conclusions

Thus, it was shown that acoustoplasma initiates a range of plasmachemical reactions in liquid media. Application of this method for obtaining hydrogen has advantages over the most commonly used steam reforming of methane and electrolysis. Having comparable to steam reforming of methane efficiency (60-80%), the proposed method does not require bulky and expensive equipment, and is superior in speed and efficiency to electrolysis. The significant advantage of the proposed method is the possibility of using wide variety of raw materials.

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[8] Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova; N. P. Zharova y V. Kohlert, “Surface modification of organic pigments by isobutyl vinyl ether copolymers under the action of ultrasonic”. Revista Inclusiones, Vol: 7 num Especial (2020): 11-21; Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova,; N. P. Zharova y V. Kohlert, “The effect of ultrasonic treatment on the stability of aqueous dispersions of inorganic and organic pigments in the presence of surfactants”. Revista Inclusiones, Vol: 7 num Especial (2020): 387-397 y Yu. P. Aleksandrova; N. S. Budanova; A. A. Farmakovskaya; N. S. Okorokova; G. N. Ustyuzhaninova; N. P. Zharova y V. Kohlert, “Theoretical and experimental studies of the spectral characteristics of doped semiconductors on the example of zinc oxide and sulfide”. Revista Inclusiones, Vol: 7 num 3 (2020): 453-463.

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