Journal of Engineering Physics and Thermophysics, Vol. 79, No. 6, 2006

PROCESSES OF TRANSFER IN A LOW-TEMPERATURE PLASMA

INFLUENCE OF ENERGY CONTRIBUTIONS TO A HIGH-VOLTAGE ATMOSPHERIC-PRESSURE DISCHARGE ON THE EFFICIENCY OF CONVERSION OF ETHANOL

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UDC 533.951

The results of experimental investigations of the conversion of ethanol in a high-voltage atmospheric-pressure discharge have been presented. The influence of the energy parameters of the discharge on the yield and energy cost of conversion products has been investigated. It has been established that the minimum energy cost of hydrogen is possible in regimes of discharge burning with a maximum degree of nonequilibrium of the plasma.

Modern power engineering based mainly on fossil fuel causes a lot of environmental problems. Furthermore, petroleum, gas, and coal reserves are limited. In this connection, an active search for new alternative power sources is on at present and great expectations are associated with hydrogen power engineering, i.e., with the utilization of hydrogen as the main energy carrier and fuel cells as electric-energy generators [1]. However, the introduction of hydrogen power into daily life is hindered by its relatively high cost and energy consumption. Among the diversity of problems that require solution, a search for new nontraditional methods of producing hydrogen, which are realized in compact apparatuses with a high energy efficiency, is pressing. Great interest has been noted recently in production of hydrogen in plasma reactors in which a high-voltage atmospheric-pressure discharge is realized [2, 3]. A substantial feature of this type of discharge is that a nonequilibrium plasma has a lower integral temperature than a quasiequilibrium isothermal plasma and is characterized by a significant difference between the vibrational and translational temperatures of the gas molecules [2, 4]. Furthermore, a discharge with a wide range of parameters — from a low-current arc discharge to an anomalous glow one — can be realized in the reactor, which is of great interest in selecting the regimes that ensure a minimum consumption of energy to produce hydrogen.

The process of steam-water conversion of methane in a high-voltage-discharge plasma was investigated for the first time in [2]. The minimum specific energy consumption to produce hydrogen in the $H_2O + CH_4$ mixture was $Q_{H_2} \approx 3$ kW·h/m^3.

In this work, we have experimentally investigated the process of conversion of ethanol in the plasma of a high-voltage atmospheric-pressure discharge and have analyzed the influence of the basic parameters of the process on the specific content of hydrogen in the reaction products and its energy cost.

Experimental Setup. To study the process of ethanol–hydrogen conversion in the plasma of a high-voltage atmospheric-pressure discharge we used an experimental setup (Fig. 1) analogous to that described in [2]. It incorporated the following basic units: a plasmachemical reactor, a system for feeding reagents and preparing a working mixture, a high-voltage power supply, and a diagnostics system. The casing of the plasmachemical reactor was made in the form of a quartz tube 1 of diameter 32 mm, in which the gaseous working mixture was processed with the plasma of a high-voltage atmospheric-pressure discharge. The distance between anode 2 (diameter 5 mm) and a hollow cath-

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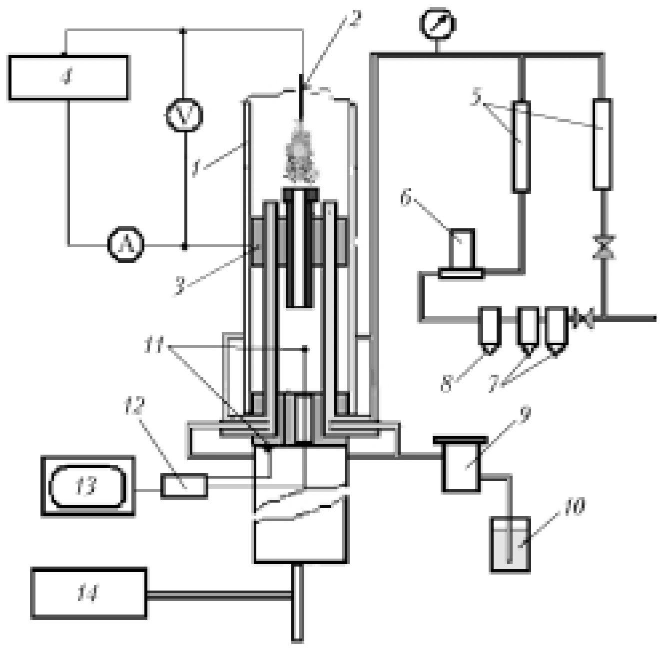


Fig. 1. Diagram of the experimental setup: 1) quartz tube; 2) anode; 3) cathode; 4) high-voltage power supply; 5) rotameters; 6) controlled flowmeter; 7) filters; 8) dehumidifier; 9) diaphragm-type liquid pump; 10) vessel for liquid; 11) thermocouples; 12) analog-to-digital converter; 13) personal computer; 14) chromatograph.

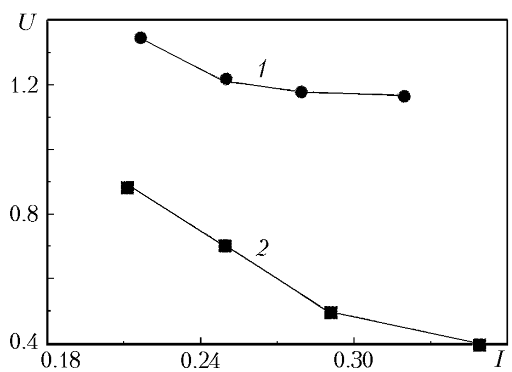
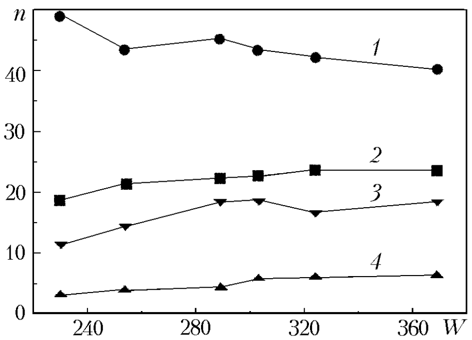


Fig. 2. Typical volt-ampere characteristic of the discharge: 1) ethanol–air mixture (γ = 5.4); 2) air. U, kV; I, A.

ode 3 (channel diameter 7 mm) could be varied within 0–40 mm. The reagents were fed by tangential injection through the holes in the cathode. A rotating ascending flow which stabilized the discharge and was a heat insulator for the discharge zone was formed. The temperature and composition of the mixture going out of the reactor were measured using thermocouples 11 and chromatograph 14. The system for feeding the reagents and preparing the working mixture ensured a supply of air to 1000 liters/h and ethanol in the range 50–200 ml/h. The air from the compressor traversed a system of filters 7 and dehumidifier 8. The gas flow rate was monitored by rotameters 5 and a controlled flow meter 6. Ethanol was fed by a diaphragm-type liquid pump 9 to the high-temperature part of an air line where the ethanol evaporated and mixed with air. Air or the air–alcohol mixture was fed to the zone of burning of the discharge by two metallic tubes in the high-temperature zone of the reactor's lower part. The high-voltage power supply 4 ensured a voltage application in the range 0.5–9 kV for currents of 50 to 500 mA.

Experimental Results and Their Discussion. The volt-ampere characteristics of the discharge for the ethanol–air mixture and air are presented in Fig. 2. The measurements were carried out for the electrode spacing d = 5



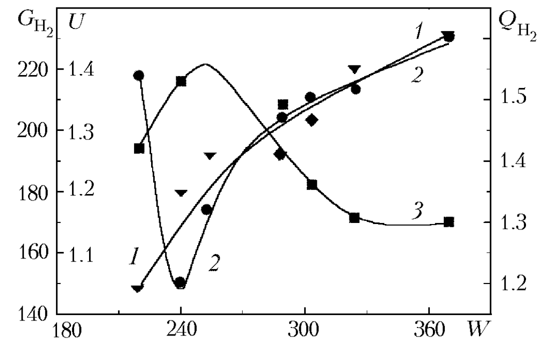


Fig. 3. Concentration of conversion products vs. discharge power: 1) hydrogen; 2) nitrogen; 3) methane; 4) carbon oxide. n, %; W, W.

Fig. 4. Values of the volume yield (1) and energy cost of hydrogen (2) and the voltage (3) at different discharge powers. Q_{H_2}, kW·h/m^3; G_{H_2}, liters/h; U, kV; W, W.

mm; the air flow rate was 0.5 m^3/h and the ethanol flow rate was 320 ml/h, which corresponded to the equivalence factor $\gamma = 5.4$. It is seen that the characteristics are dropping, typical of a high-voltage atmospheric-pressure discharge; the discharge in the mixture burns at a higher voltage than that in air. The discharge voltage is much higher than 100 V for both cases.

The discharge is usually arc-type when the voltage drop on the electrodes is 20 V or lower; if the burning voltage is above 100 V, we have a glow discharge [5]. The plasma in the arc-discharge filament has gas and electron temperatures $T_e \approx T \approx (6–10)\cdot 10^3$ K. In a glow discharge, the gas temperature is about room temperature, and T_e is several thousand degrees. The discharge realized in experiments is intermediate between a strongly nonequilibrium glow-discharge plasma and an arc. A significant difference between the electron and gas temperatures is observed: $T_e \approx$ 1–3 eV and $T \approx$ 2000–3000 K for an electron density of $n_e \sim 10^{13}$–10^{14} cm^{-3}; the electric field is lower than that in a glow discharge but is higher than that in an arc discharge. Furthermore, for electrode spacings d comparable to the diameter of a hole in the cathode, we may have a discharge with its characteristics typical of a hollow-cathode discharge [6]. The drop in the electrode voltage may be ~1000 V for it, but in other characteristics we should classify such a discharge with arc discharges. The discharge in our experiments had the form of a volume discharge for the ethanol–air mixture and a filament one for air. The discharge voltage was 1000–1500 V in conversion of the ethanol–air mixture and 300–900 V in air; the current strength varied within 0.1–0.5 A.

Figure 3 gives the experimental values of the concentrations of the basic components of conversion of ethanol — hydrogen, nitrogen, methane, and carbon oxide — as functions of the discharge power.

Near the lower bound of existence of the discharge, i.e., the minimum voltage and current values for which we were able to maintain a self-sustained discharge, the content of hydrogen was about 19%. An increase from 230 to 300 W in the discharge power led to an increase in the concentration of hydrogen to 24% followed by a weak growth to 27%. As the discharge power increased, the concentration of nitrogen in the outgoing gaseous mixture decreased and that of methane and carbon oxide increased. The temperature of the outgoing gases, measured at a distance of 40 mm from the section of the cathode's lower part, increased from 790 to 840°C with an increase from 230 to 370 W in the power input.

One of the most important characteristics of the process of conversion of hydrocarbons is the energy cost, which is determined by the consumption of energy to obtain a unit volume of hydrogen. Figure 4 plots the volume yield of hydrogen (curve 1) and its energy cost (curve 2) versus the discharge power. It is seen that the value of the hydrogen yield grows with power, but the optimum values of the energy cost are realized for discharge powers of 240 to 260 W. The existence of the optimum is attributable to the fact that the discharge voltage attains its maximum in this power range. As the energy contributions increase, this voltage drops, whereas the current strength grows. Also, Fig. 4 gives the values of discharge voltages (curve 3). It is seen that the minimum values of the energy cost are realized at maximum voltages, and, as has been noted above, the higher the electric-field strength, the higher the degree of nonequilibrium of the plasma and the more efficient the processes of ethanol–hydrogen conversion. For powers of the discharge of 220 to 240 W, it burns in an unstable regime and the efficiency of conversion decreases as a consequence.

Thus, as part of the experimental investigations of the process of ethanol–hydrogen conversion in the plasma of a high-voltage atmospheric-pressure discharge, we have obtained the following values of the basic parameters: maximum hydrogen yield $n \leq 27\%$, hydrogen output $G_{H_2} = 290$ liters/h, and minimum energy consumption to produce hydrogen $Q_{H_2} \approx 1.2$ kW·h/m^3. The minimum energy consumption to produce hydrogen is realized near the lower bound of stable burning of the discharge at the maximum discharge voltages.

NOTATION

d, electrode spacing, mm; G_{H_2}, hydrogen output, liters/h; I, current strength, A; n, concentration of gases, %; n_e, electron density, cm^{-3}; Q_{H_2}, specific energy consumption to produce hydrogen, kW·h/m^3; T, gas temperature, K; T_e, electron temperature, K; U, voltage, kV; W, power, W; γ, equivalence factor.

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