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Process of hydrocarbons partial oxidation stimulated by plasma of atmospheric pressure microwave discharge.

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The big interest to process of syn-gas (H_2+CO) production from hydrocarbon fuels is caused by rapid development of hydrogen power in the world.

The main difficulty on a way of wide use Fuel Cell is the problem of storage and transportation of hydrogen. For this reason it is represented expedient to production of hydrogen from hydrocarbon fuels (kerosene and methane) directly on a place of its consumption, on the board of mobile and small-sized technological installations. The specific of this process is that the air is used as oxidant. The future enrichment of production mix by hydrogen usually made by means of shift reaction $CO+H_2O=CO_2+H_2$ and is not a theme of the presented work.

This paper presents the results of an investigation of plasma processes for hydrogen and syn-gas (H_2+CO) production by the method of kerosene ($C_{11}H_{22}$) and methane (CH_4) partial oxidation by air:

$$C_{11}H_{22}+5.5O_2+20.69N_2=11H_2+11CO+20.69N_2+\Delta H_1 \quad (1)$$

$$\Delta H_1=944\text{kJ/mol}$$

$$CH_4+0.5O_2+1.88N_2=2H_2+CO+1.88N_2+\Delta H_2 \quad (2)$$

$$\Delta H_2=35.65\text{kJ/mol}$$

The process of partial oxidation was stimulated by atmospheric pressure microwave discharge.

The purpose of this research is to compare the efficiency of plasma and thermal energy inputs during the process realization. The research of kerosene partial oxidation was performed with the aid of the coaxial and resonator (cavity) microwave plasmatrons. Researches of methane partial oxidation have been performed in the coaxial plasmatron only.

Installation with the coaxial microwave plasmatron consists of the plasmachemical reactor, the block of reagents preparation and the exhaust block. The scheme of the microwave radiation input in the plasmachemical reactor is shown on figure 1. As a source of microwave radiation generator KIE-5 (2,45 GHz) with adjustable output power in a range 1-5 kW is used. Absorption of the microwave power from magnetron by the discharge with the help of adjustment of the matching was possible up to values more than 90% i.e. practically full absorption was achieved. The length of an internal conductor of a coaxial line can be easily changed even then discharge is burning, achieving the discharge zone overlapping with the burning zone. Input mix of fuel and air was carried out through a ring collector and "belt" of 60 apertures in diameter of 2 mm in a wall of the discharge chamber. Typical values of the air flow rate through the reactor in experiments was 6 - 7 (N)m^3/hour (near 2 l/s).

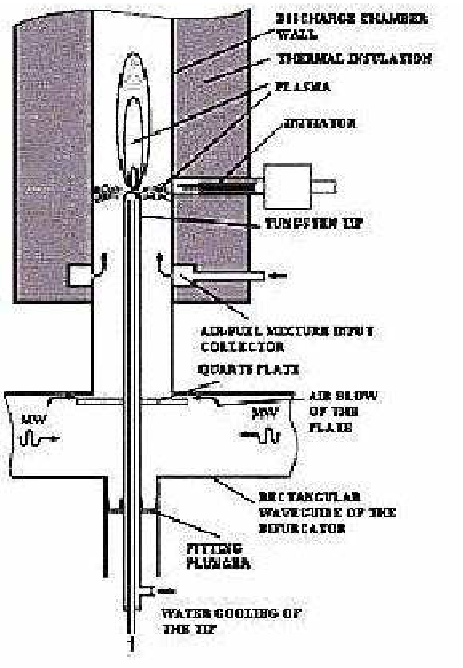

Fig.1 Plasmachemical reactor with coaxial plasmatron.

Installation with the resonator (cavity) microwave plasmatron consists of the same parts. The scheme of the plasmachemical reactor is shown on figure 2. In this case absorption of the microwave power by the discharge was near 80%.

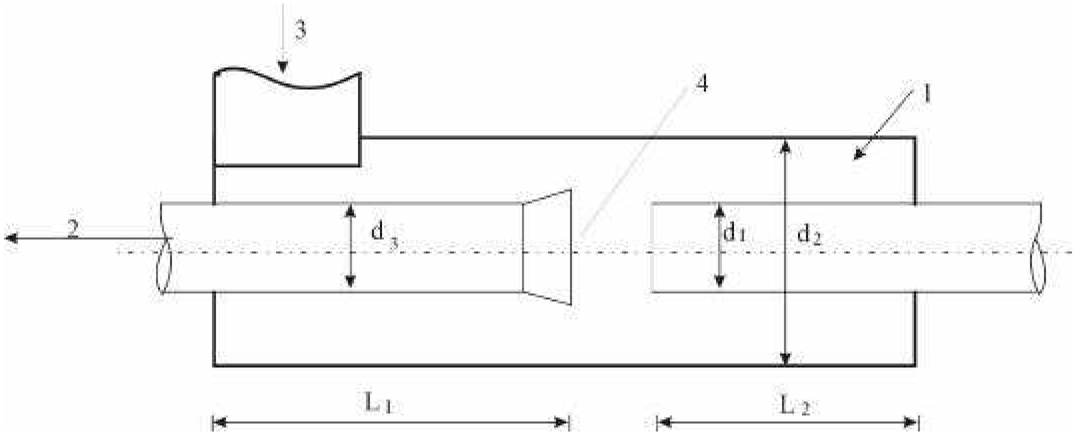

Fig. 2. Draft of construction scheme of resonator (cavity) microwave plasmatron.
1. Input of reagents. 2. Output of products. 3. Input of microwave. 4. Discharge zone.

The total thermal power input was determined as the sum of power inputs from the full combustion of a fuel part and from the reagents preliminary heating.

$$E_{total} = E_{plasma} + E_{thermal} = E_{plasma} + E_{comb} + E_{heat}$$

The dependence of the fuel conversion degree on the plasma and thermal energy inputs was studied. The results is shown on figures 3,5 (kerosene partial oxidation into 2 different types of plasmatrones) and 4 (methane partial oxidation into coaxial plasmatrone). A comparison of the plasma and thermal energy inputs efficiency for the partial oxidation kinetics acceleration was performed.

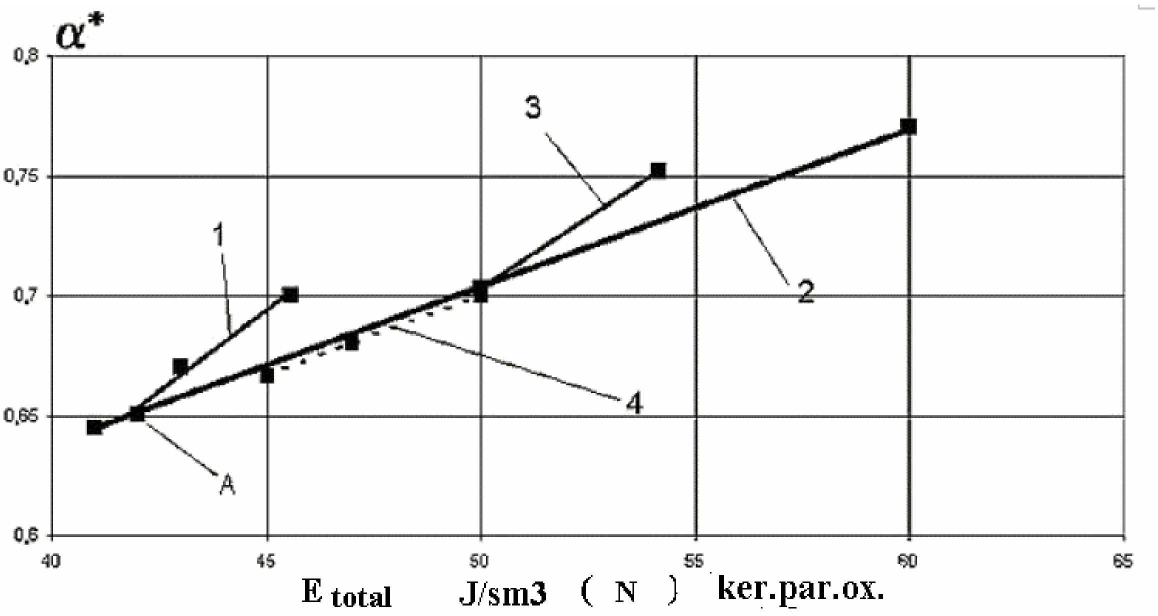

Fig. 3. Experimental dependencies relative degree of conversion of kerosene from total specific energy input in coaxial plasmatron. 1, 3 – variable only E_{plasma} .; 2 – variable only E_{comb} .;4 – variable only E_{heat}

Wide interval of thermal energy input changes in both installations was obtained by full combustion of a fuel part or in other words by increasing of the air/fuel ratio above stoichiometry for partial oxidation (1),(2).

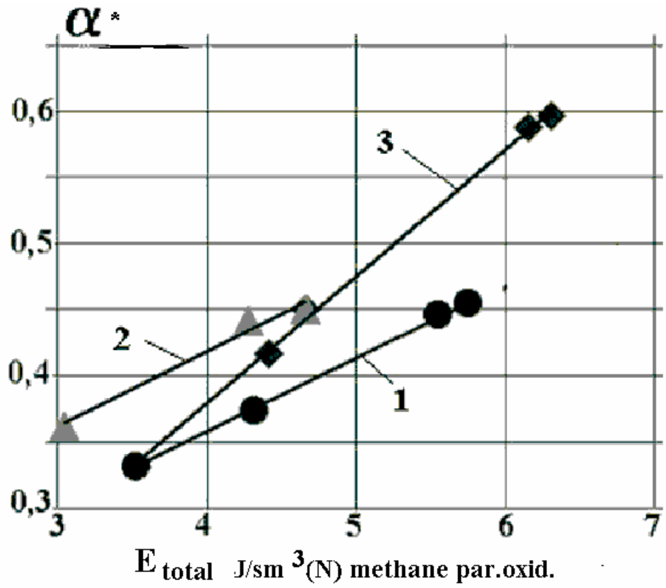

Fig. 4. Experimental dependencies relative degree of conversion of methane in syn-gas from total specific energy input in coaxial plasmatron. 1- variable $E_{thermal}$ at E_{plasma} =1,7 J/sm^3 = const; 2 – variable $E_{thermal}$ at E_{plasma} =3.0 J/sm^3 = const ; 3- variable E_{plasma} at $E_{thermal}$ =1.7 J/sm^3 = const;

During experiment energy input in preliminary heating of reagents, the energy absorbed by the discharge, a ratio between fuel and an oxidant ($S=n[C_nH_m]/2[O_2]$, in a case of partial oxidation S=1) were supervised. In a case when S<1 reaction of partial oxidation goes in plenty of oxidant that results in full combustion of a part of fuel (and to entering additional thermal energy input).

The term a relative degree of conversion - α^* in this case was entered, representing expression for a degree of conversion of that part of fuel which participated in reaction of partial oxidation.

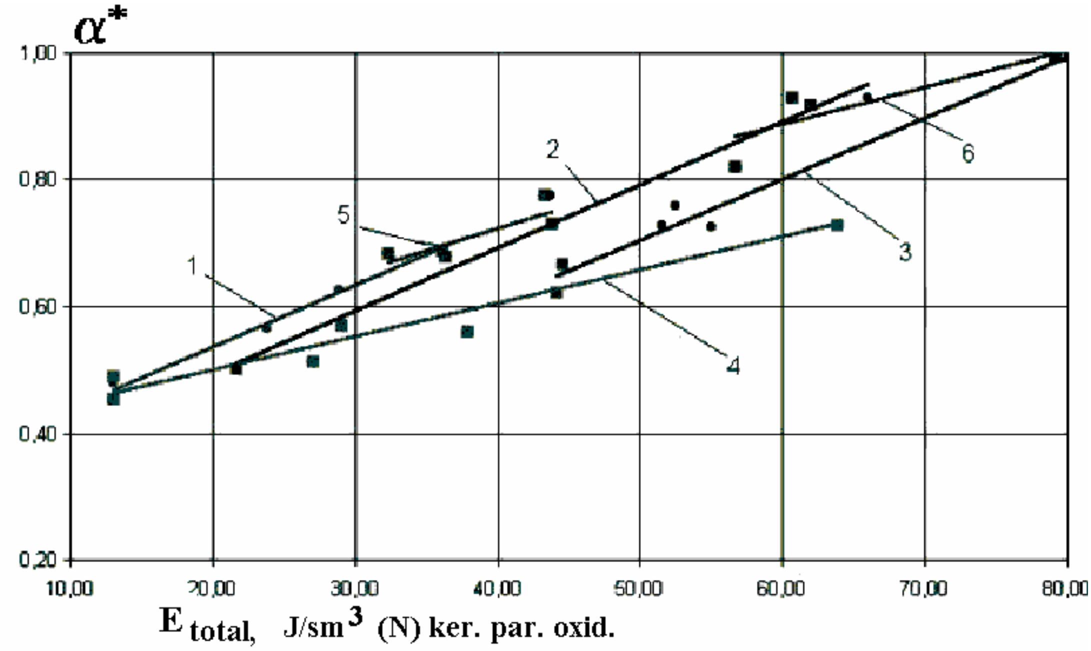

Fig. 5. Experimental dependencies relative degree of conversion of kerosine in syn-gas from total specific energy input in rezonator (cavity) plasmatron. 1,2,3- variable E_{plasma} at $E_{thermal}$ = const; 4,5,6- variable $E_{thermal}$ at E_{plasma} = const.

The structure of a mix on an output received as a result of the gas analysis was processed by specially developed technique. Dependences of a relative degree of conversion α^* were analyzed from plasma and thermal energy inputs and shown on figures.

It is shown that for the coaxial plasmatron the efficiency of the plasma energy input for increasing of the conversion degree is 1,3 times greater than the efficiency of the thermal energy input. For the resonator plasmatron this factor is even greater and reaches 1,6.This effect may be explained by the presence of active particles (ions, radicals) in the discharge plasma. These particles participate in chain chemical reactions resulting in overall acceleration of the process kinetics.