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Carbon Black Generation in Gliding Discharges

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Carbon blacks (CB), being an important group of industrial carbons, are used chiefly in the rubber tire, plastics, ink and paint industries. Some new concepts of CB manufacturing are based on processing of hydrocarbons in the arc plasmas [1,2] and plasma-torch reactors [3]. However, among different processes being considered the most promising are those using non-equilibrium plasmas generated under gliding discharge ("Glid-Arc") conditions. In that kind of plasmas, due to the high electron energy, even stable methane molecules can be activated, whereas the mean temperature of reactants is much lower than that in the flames.

For studying the conversion of gaseous hydrocarbons into CB, a gliding-discharge reactor has been developed, operated at the frequency of 50 Hz [4,5]. It was made of a quartz-glass tube of 110 mm in diameter with three symmetrically placed steel electrodes. Using the specially shaped main electrodes and the additional ignition electrode, the continuous run of cyclic discharges can be maintained, being an ordinary form of the gliding discharges. Pure gases: methane and argon were the only components of the inlet gas mixture. The outlet gas components were determined using gas chromatography (Hewlett Packard HP 6890).

The results of experiments with CH_4 (5–20%) + Ar mixtures for the overall gas flow rate of 1500 Nl/h are presented (Table 1 and Figs. 1–3) using the following parameters:

Starting flow rate of CH_4	$W[CH_4]$ [mol/h]
Flow rate of CH_4 in the outlet	$W[CH_4]^*$ [mol/h]
Discharge power	P [W]
Overall methane conversion	$X = \frac{W[CH_4] - W[CH_4]^*}{W[CH_4]}$
Overall methane conversion rate	$Y = X\, W[CH_4]$ [mol/h]
Methane conversion rate into CB	Y_C [molC/h]

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Methane conversion into CB $X_C = \frac{Y_C}{W[CH_4]}$

Methane conversion rate into C_2H_n Y_{hc} [molC_2H_n/h]

Overall unit energy consumption $E_O = 3600 \frac{P}{Y}$ [J/mol]

Energy consumption per CB unit (mol C) $E_C = 3600 \frac{P}{Y_C}$ [J/molC]

Table 1.

Methane concentration % vol.	Power kW	Conversion into:				
		Overall X	C X_C	C_2H_2	C_2H_4	C_2H_6
5	1.18	63.1	21.9	39.1	1.5	0.6
7.5	1.0	58.2	17.7	39	1.2	0.4
10	1.22	57.1	15.9	39.5	1.3	0.4
15	1.50	65.6	37.7	26.9	0.8	0.2
20	1.51	73.5	55.4	19.3	0.5	0.1

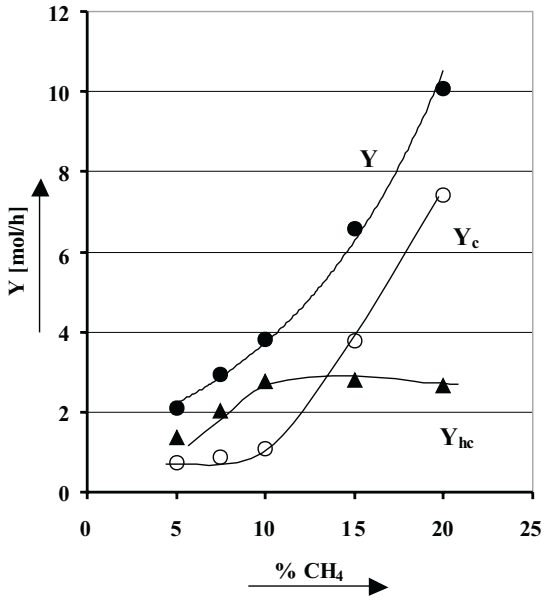

Figure 1. Methane conversion rate: overall – Y, into CB – Y_C, and into C_2H_n – Y_{hc} *vs.* methane concentration in the inlet mixture CH_4 + Ar.

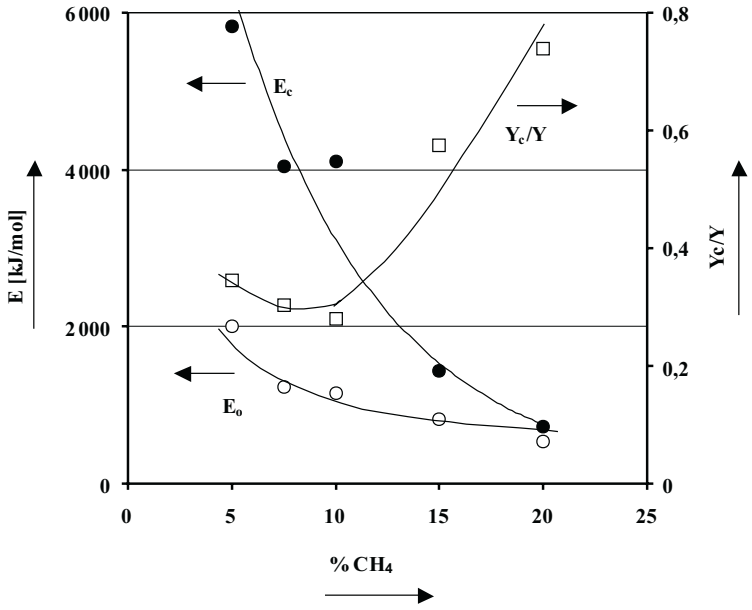

Figure 2. Unit energy consumption: overall – E_O and for the CB generation – E_C, and Y_C/Y ratio *vs*. methane concentration in the inlet mixture CH_4 + Ar.

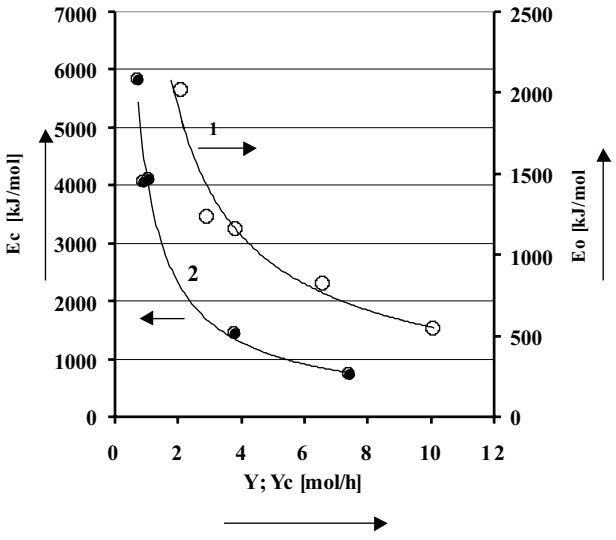

Figure 3. Unit energy consumption: (1) E_O for different values of the overall methane conversion rates Y and (2) E_C for different values of Y_C.

It should be pointed out that, under the fixed parameters of the supply system, the discharge power in the reactor changed depending on the composition of the inlet gas mixture. The power was much greater at high methane concentrations (15–20%) than within 5–7.5%. Carbon black and acetylene were obtained as the main products with small amounts of ethylene and ethane. The overall methane conversion X was high (57–75%) in all the measurement series, whereas the methane conversion into CB X_C varied (within 16–55%) depending on the initial methane concentration.

It has been found that the concentration of methane in its mixtures with argon influenced substantially the rate of CB formation. A 4-fold increase of initial methane concentration (from 5 to 20%) gave a 5-fold increase of the total rate of methane conversion Y. At the same time, a 10-fold increase of the rate of CB formation Y_C was observed and the ratio of Y_C/Y increased from 0.3 to 0.74. It is also interesting that the rate of acetylene and other C_2H_n formation (Y_{hc}) increased markedly with increasing initial methane concentration within 5–10%, whereas it remained nearly constant (2.7–2.8 mole/h) at higher methane concentrations. Therefore, if we assume that the CB formation is a result of the following series of reactions [6,7]

$$CH_4 \rightarrow CH \rightarrow C_2H_2 \rightarrow C$$

it can be concluded that the increase of the initial methane concentration within 5–10% results in a considerable increase of rate of the initial stages in the reaction chain, where acetylene and other C_2H_n hydrocarbons are produced. At the same time the increase of the CB generation rate (the final stage) is much smaller. On the other hand, when higher methane concentrations were used (within 10–20%), the rate of all these stages increases almost equally. For this reason the initial concentration of methane (within 5–20%) has a substantial effect on the unit energy consumption. The amount of energy consumed per unit of methane converted E_O decreases considerably, however the consumption of energy per unit amount of CB produced E_C decreases in a much higher extent. This favourable change is partly due to the decrease of energy absorbed by the stream of argon being the gas component, but the main reason lies probably in the increase of concentration of the reactants (mainly acetylene) involved in the formation of CB. One can state therefore, that the concentration of methane in CH_4 + Ar mixtures is an important factor determining the economy of CB production in the gliding discharges. High methane concentrations can give: 1) higher discharge energy, 2) higher rate of CB formation, 3) smaller unit energy consumption, and 4) lower consumption of argon being the auxiliary gas.

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