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MODEL OF ACETYLENE SYNTHESIS FROM METHANE IN A HYDROGEN PLASMA JET[1)]

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The energy consumption in the process of acetylene synthesis is presented in the form of the analytic dependences as a function of reaction temperature, hydrogen/methane volume ratio and reaction chamber efficiency. On the basis of the earlier described quasiequilibrium process those dependences were elaborated by the rotatability Box-Hunter method.

I. INTRODUCTION

The synthesis of acetylene from methane is one of the most frequently modelled processes occurring in an equilibrium plasma jet [1–8]. A large number of thermodynamic as well as kinetic computational models for the evaluation of the final parameters, among them a yield of the acetylene synthesis from methane, were elaborated. A large number of reaction parameters makes difficult a univocal analytical description of the process and the establishing of essential criteria of its effectivity. The statistical methods of the planning of experiments have been applied in the analysis and the optimizing of various physical and chemical processes [9–12].

The aim of the presented work was to test the statistical methods of experiment planning as a way to the energy prediction in the process of the acetylene from methane synthesis in a hydrogen plasma jet. The investigations consist in a numerical simulation of the course of the acetylene synthesis process. The equilibrium model of the process, which has been already elaborated [6], was taken as a basis. In the model the energy consumption was calculated from a known initial reaction temperature, the composition of the methane-hydrogen mixture and the efficiency of both a plasma generator and a reaction chamber.

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II. MODEL CONSIDERATION

The yield of the process of acetylene synthesis from methane is characterized by a substrate in-product conversion degree and an energy consumption for a preparation of 1 m^3 of acetylene. The energy consumption is a main factor of the cost of the plasma method of the acetylene production [1]. A number of the calculation methods of the energy consumption for the production process of C_2H_2 from methane in a plasma jet were elaborated [1–7].

A number and a character of the parameters influencing the energy consumption depend, in a considerable degree, on the accepted calculation model of the process of acetylene synthesis from methane synthesis. On the basis of a model consideration [6] the energy consumption can be described by the following formula:

$$EC = \left[\frac{2E_m.\Delta H^0_{Tq}(C_2H_2)}{E_m.\eta_{rch} - X\Delta H_n(T_q) - \Delta H_m(T_q)}\right]/\eta = \frac{EC_{pj}}{\eta}, \tag{1}$$

where E_m - is the specific energy of methane calculated in relation to on energy of the plasma jet, $\Delta H^0_{Tq}(C_2H_2)$ - is the enthalpy of acetylene synthesis from methane at quenching temperature, $\Delta H_h(T_q)$, $\Delta H_m(T_q)$ - are the enthalpy of hydrogen and methane at quenching temperature, repsectively, EC_{pj} - is the energy consumption calculated relative to plasma jet energy.

It should be emphasized that the specific energy of methane which appears in the formula (1) depends also on the initial reaction temperature [6]:

$$E_m = X\Delta H_h(T_r) + \Delta H_m(T_r), \tag{2}$$

where $\Delta H_h(T_r)$, $\Delta H_m(T_r)$ - are the enthalpy of hydrogen and methane at reaction temperature, respectively.

From the elaborated quasiequilibrium model of the process and the known values of the reaction temperature, a hydrogen /methane molar ratio plasmatron and a reactor chamber efficiency, it is possible to calculate the effective energy consumption. However, the model does not allow to derive a direct functional dependence such as this:

$$EC_{pj} = f(Tr, X, \eta_{rch}). \tag{3}$$

Those dependences were established by the method of the statistical planning of experiments [9] one decided to derive the dependence shown as a fromula (3). The second order Box-Hunter method of the rotability planning was applied [13]. It is a five level multifactor planning. In the presented work the number of the factors was limited to 3.

For three factors the regression equation of the planned experiment can be expressed in the following from:

$$EC_{pj} = b_0+b_1z_1+b_2z_2+b_3z_3+b_{12}z_1z_2+b_{13}z_1z_3+b_{23}z_2z_3+b_{11}z_1^2+b_{22}z_2^2+b_{33}z_3^2. \quad (4)$$

In the equation the dimensionless variables are used. These variables are calculated in the following manner:

$$z_i = \frac{a_i - a_{i,0}}{\Delta a_{i,0}} \quad (5)$$

where

$a_{i,0}$	ground level	$z_i = 0$
$\Delta a_{i,0}$	variation interval	
$a_i = a_{i,0} + \Delta a_{i,0}$	higher level	$z_i = +1$
$a_i'' = a_{i,0} - \Delta a_{i,0}$	lower level	$z_i = -1$

The experiment plans (using dimensionless variables) and the calculated values of an effective energy consumption EC_{pj} are diven in Table 1.

The regression equation (4) calculated in accord with the method presented by Akhnazarova and Kafarov [10] is as follows:

$$EC_{pj} = 54.18 + 13.36z_1 - 14.33z_2 + 13.17z_3 + 9.86z_1.z_2 + 13.89z_1.z_3 + 0.23z_2.z_3 + 14.22z_1^2 + 7.38z_2^2 + 2.23z_3^2. \quad (6)$$

A comparison of dependences of EC_{pj} on the reaction temperature according to the quasiequilibrium model and to the regression equation (5) for:

a) $x = 2$ and $\eta_{rch} = 0.5$

b) $x = 0.5$ and $\eta_{rch} = 0.9$

are presented in Fig. 1.

The regression equation, as shown in Fig., adequately describes the dependence of an energy consumption EC_{pj} on the reaction temperature.

The Table 1 was completed with the methane-to-acetylene conversion degrees U_{ac}. The U_{ac} was calculated from the following formula :

$$U_{ac} = 2E_m/EC_{pj}.$$

The formula results from the definitions of the effective energy consumption as well as the methane to acetylene conversion degree.

$$EC_{pj} = E_{pj}/V_{ac} = E_{pj}/0.5V_m.U_{ac} = 2E_m/U_{ac} \quad (7)$$

Table 1

Plan of experiment for the calculation of parameters of equation (4) calculated values of U_{ac} from equation (6)

Level	Factor		
	z_1 T_r[K]	z_2 η_{rch}	z_3 X
Δz_i	600	0.15	1
+1	3800	0.85	3
0	3200	0.70	2
−1	2600	0.55	1

Number of experiment	Design matrix z_1	z_2	z_3	EC_{pj} [MJ/m^3]	U_{ac} [mol.%]
1	−1	−1	−1	87.8	26.9
2	+1	−1	−1	68.7	83.8
3	−1	+1	−1	41.7	56.7
4	+1	+1	−1	58.3	99.9
5	−1	−1	+1	91.4	42.1
6	+1	−1	−1	124.0	95.3
7	−1	+1	+1	42.5	90.6
8	+1	+1	+1	118.0	99.9
9	−1.682	0	0	70.5	33.6
10	+1.682	0	0	116.0	99.9
11	0	−1.682	0	99.0	50.7
12	0	+1.682	0	50.3	99.9
13	0	0	−1.682	41.0	63.9
14	0	0	+1.682	77.8	95.3
15	0	0	0	55.3	94.2
16	0	0	0	53.6	93.5
17	0	0	0	54.0	92.9
18	0	0	0	54.7	91.7
19	0	0	0	55.1	91.1
20	0	0	0	55.4	90.5

Where V_{ac} - volume of acetylene, V_m - volume of methane.

The effective specific energy of the methane E_m was also defined as the ratio of a plasma jet energy to the volume of a substrate introduced into the reactor.

Then, using the formulas (5) and (2) the influence of a hydrogen/methane ratio and reaction chamber as well as plasma torch efficiences on the energy consumption

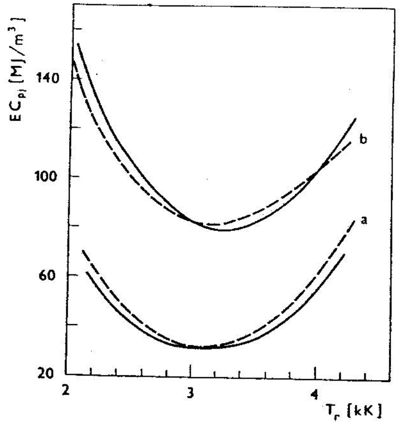

Fig. 1. A comparison of dependences of energy consumption EC_{pj} on the reaction temperature : a) $x = 2$ and $\eta_{rch} = 0.5$; b) $x = 0.5$ and $\eta_{rch} = 0.9$; – quasiequilibrium model, – – – equation (5).

Table 2

The dependence between the energy consumption and the hydrogen-methane ratio x as well as a plasmatron efficiency ($T_r = 3200$K, $\eta_{rch} = 0.7$)

x	EC_{pj}	EC [MJ/m³]				
		η				
	[M/m³]	0.4	0.5	0.6	0.7	0.8
4	90.2	225	180	150	129	113
3	69.7	174	139	116	99.6	87.1
2	54.1	135	108	90.3	77.3	67.7
1	43.5	108	87.0	72.5	62.1	54.4

was analyzed. The results are listed in Tables 2 and 3.

As it is shown in Table 2 and 3 the decrease of the energy consumption is aided when a hydrogen-methane ratio is decreased and a reaction chamber efficiency is increased. The lowering of the energy consumption EC for a constant EC_{pj} value is favored by an increase of a plasmatron efficiency. These conclusions are supported by our earlier experimental works as well as literature data [16].

Table 3

The dependence between the energy consumption and the reaction chambers η_{rch} as well as a plasmatron efficiency ($T_r = 3200$, $x = 2$)

η_{rch}	EC_{pj}	EC [MJ/m³]				
		η				
	[M/m³]	0.4	0.5	0.6	0.7	0.8
0.4	112	280	224	187	160	140
0.55	76.1	190	152	127	109	95.1
0.7	54.1	135	108	90.3	77.3	67.7
0.85	46.3	116	92.5	77.1	66.1	57.8

Table 4

Experimental results obtained using a reactor on a large laboratory scale

Parameters	Units	1	2	3	4
$P^{*)}$	kW	61.2	64.3	96.7	99.8
V_h	m³/per 1h	10	12.5	18	18
V_m	m³/per 1h	12	9	15	15
x	1	0.83	1.33	1.2	1.2
η	%	84.6	88.3	88.4	85.4
T_r	K	3090	3330	3260	3300
E_m	MJ/m³CH_4	15.2	22.4	20.5	20.9
η_{rch}	%	78.7	75.1	78.9	78.9
EC_{pj}	MJ/m³C_2H_2	53.6	59.7	59.6	60.4
EC	MJ/m³C_2H_2	63.3	67.6	67.5	67.5
U_t	%	64.8	76.5	69.8	69.5
U_{ac}	%	58.0	76.1	68.7	63.4
$cCH_4^{**)}$	vol.%	13.9	14.8	8.3	6.7
$cC_2H_2^{**)}$	vol.%	9.7	10.2	9.8	9.8

*) P - power of arc discharge **) cCH_4, cC_2H_2 - concentrations of CH_4 and C_2H_2, respectively, in post-reaction temperature.

The average efficiencies of a plasmatron and a reaction chamber obtained at x=2 in a reactor with an arc power 10-40 kW [6,14] were equal to 0.56 and 0.7, respectively. An occurrence of an optimal range of a reaction temperature (2800–3300 K) in which the effective energy consumption reaches a minimum value of

60–70 MJ/m^3 was indicated. Also the EC obtains a minimum value of 110–130 MJ/cu.m. The U_{ac} reaches 54–85%.

The experiments at an arc power of 60–110 kW [6,8,15] were carried out with a plasmatron operating at high efficiency 80–88%. These conditions allowed us to perform the measurements at x=0.8–1.3. The reaction chamber efficiency was 80%.

As it is shown in Table 4 under the above conditions the effective energy consumption attained 52–60 MJ/m^3. The energy consumption attained 52–60 MJ/m^3. The energy consumption EC was found to be 60–70 MJ/m^3, which is significantly lower than at x=2. The CH_4 to C_2H_2 conversion reaches 54–76%.

Jasko and Laktushin [16] obtained in a reactor with the arc power 0.4–1 MW at x=0.5–0.7 the effective energy consumption EC_{pj} 43–50 MJ/m^3. The reaction products were quenched by means of a water spray.

The considerations mentioned above show that the methods of experiment planning are useful for the prediction of the C_2H_2 from CH_4 synthesis yield on the basis of an adequate quasiequilibrium process model.

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МОДЕЛ СИНТЕЗА АЦЕТИЛЕНА ИЗ МЕТАНА В СТРУЕ ПЛАЗМЫ ВОДОРОДА

Приводится употребление энергии в процессе синтеза ацетилена в форме аналитической зависимости от температуры, отношения объема водорода к метану и эффективности реакционной камеры. Зависимости получены на основе раньше опубликованных квазиравновесных процессов, но также с применением метода ротабельности Бокса-Хунтера.