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DEVELOPMENT OF A METHOD AND ALGORITHM FOR CALCULATING THE EQUILIBRIUM OF METHANOL SYNTHESIS UNDER MEDIUM PRESSURE

The object of research is the synthesis of methanol under medium pressure, and the subject of research is the calculation of the equilibrium composition. A method and an algorithm for calculating the equilibrium composition of products are proposed. Calculations have been carried out using two mechanisms of methanol synthesis. First of all, the first approach to the mechanism of methanol synthesis was applied, which provides for the formation of methanol from a mixture of carbon oxides and hydrogen by the simultaneous reaction of methanol synthesis from carbon monoxide (IV) and steam reforming of carbon monoxide (II). The poor convergence of the system of two equations on the basis of this interaction mechanism is established, which complicates the selection of initial approximations and the choice of a real solution. At the second stage, the mechanism of the formation of methanol from carbon monoxide (II) with the simultaneous flow of steam reforming of CO in the opposite direction is considered. The degree of the equation describing the dependence of the equilibrium constant on the partial pressures of the reactants decreases in comparison with the first approach and causes much better convergence when solving the system of two equations. The use of a coefficient that takes into account the effect of pressure on the value of the equilibrium constant is proposed, and its dependence on temperature is approximated. The developed algorithm is implemented in MathCAD, while the relative maximum deviation for methanol is 8.53 %. The third approach is to use two coefficients that adjust the equilibrium constant depending on pressure. In this case, the relative maximum deviation of methanol is 16.25 %. The program according to the proposed algorithm takes into account the multiplicity of the initial data, namely the possibility of varying the composition of the initial mixture, pressure and temperature. The calculation was carried out according to the initial data of industrial implementation of methanol synthesis. Comparison of the equilibrium concentration of methanol with the real one at the outlet of the synthesis column established the degree of equilibrium reaching 39.4 %, which indicates the presence of a reserve of methanol synthesis and the possibility of increasing the practical yield of methanol.

Keywords: methanol synthesis, equilibrium constant, equilibrium composition, synthesis column, multivariate calculations.

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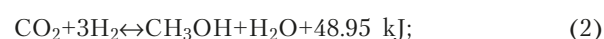
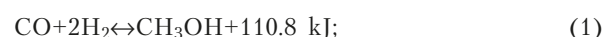
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1. Introduction

The industrial synthesis of methanol from carbon monoxide (II) was initially implemented on a zinc-chromium catalyst under high pressure up to 30 MPa. The study of the regularities of this process is devoted to enough works, for example [1, 2]. At the current stage of methanol production, they work with a mixture of CO, CO₂ and H₂ on a low-temperature catalyst containing copper under a pressure of up to 10 MPa [3, 4]. A complex model of CO₂ integration in the methanol synthesis plant was developed: the reforming product (synthesis gas) is mixed with a high-purity CO₂ stream that comes from the greenhouse gas capture process at the power plant [5]. A method of significantly increasing the yield of methanol during CO₂

hydrogenation by *in situ* sorption of methanol and water in alkaline salt ionic liquids is proposed. The reaction and sorption of the product take place in one reactor vessel, which shifts the equilibrium yield of methanol at 75 bar (H₂:CO₂=3:1) and 250 °C from 25.3 % to 60 % [6]. An analysis of the kinetic aspect of methanol synthesis under medium pressure is provided in [7].

The formation of methanol proceeds by reverse exothermic reactions:



The modern approach to the mechanism of methanol synthesis involves the formation of methanol from a mixture of carbon oxides and hydrogen by the simultaneous course of reactions (2) and (3) [3, 8]. From a thermodynamic point of view, reaction (1) is dependent: (2)+(3)=(1). Therefore, the equilibrium composition is determined by the values of the equilibrium constants of reactions (2) and (3) and the equilibrium partial pressures P_i of the reactants:

$$Kp_2 = \frac{P_{\text{CH}_3\text{OH}} \cdot P_{\text{H}_2\text{O}}}{P_{\text{CO}_2} \cdot P_{\text{H}_2}^3}; \quad (4)$$

$$Kp_3 = \frac{P_{\text{CO}_2} \cdot P_{\text{H}_2}}{P_{\text{CO}} \cdot P_{\text{H}_2\text{O}}}. \quad (5)$$

There are several equations for calculating the equilibrium constant Kp_3 and Kp_2 , provided that the components of reactions (2) and (3) are ideal gases, which is possible at low pressure. The most theoretically justified are the equations given in [1]:

$$\lg Kp_2 = 1581.7/T - 8.7639 \cdot \lg T + 0.0021105 \cdot T - 1.9303 \cdot 10^{-07} \cdot T^2 + 15.0921; \quad (6)$$

$$\lg Kp_3 = 2167/T - 0.5194 \cdot \lg T + 0.001037 \cdot T - 2.331 \cdot 10^{-07} \cdot T^2 - 1.2777. \quad (7)$$

In [3], the method of calculating the equilibrium composition using the following variables is given: x – the number of moles of CO that reacted according to reaction (3) to its equilibrium state; y is the number of moles of CO₂ that reacted according to reaction (2) to its equilibrium state. Thus, the system of equations (4) and (5) is transformed into a system with two unknowns x and y and known initial concentrations of reactants, products, and pressure. The equilibrium composition of the reaction products was calculated using MathCAD and it was established that the results do not coincide with those in the literature. This is explained by the fact that the expressions for the equilibrium constants were obtained for atmospheric pressure. At the same time, it was established that the values of the initial approximations x and y significantly affect the result: the system of two nonlinear equations gives few real results or is not solved at all due to poor convergence. The selection of initial approximations and the choice of a valid solution turn into a rather difficult task. This is a significant drawback of this method and prompts the search for an effective and simpler method of equilibrium calculation.

Thus, *the object of research* was the synthesis of methanol under medium pressure. The relevance of the problem lies in the fact that the equilibrium concentration of methanol determines its maximum possible practical concentration. Therefore, *the aim of research* is to justify the method of equilibrium calculation and to develop an algorithm for its implementation in the MathCAD.

2. Research methodology

It is proposed to change the direction of the calculation in order to replace equation (4) with an equation of a lower order. From a thermodynamic point of view, reaction (2) can be considered as dependent: (1)–(3)=(2). Therefore,

the equilibrium state can be determined by the values of the equilibrium constants of reactions (1) and (3):

$$Kp_1 = \frac{P_{\text{CH}_3\text{OH}}}{P_{\text{CO}} \cdot P_{\text{H}_2}^2}; \quad (8)$$

$$Kp_3 = \frac{P_{\text{CO}_2} \cdot P_{\text{H}_2}}{P_{\text{CO}} \cdot P_{\text{H}_2\text{O}}}. \quad (9)$$

Dependence of the equilibrium constant Kp_1 on the temperature T [9]:

$$\lg Kp_1 = 3971/T - 7.492 \cdot \lg T + 0.00177 \cdot T - 3.11 \cdot 10^{-08} \cdot T^2 + 9.218. \quad (10)$$

The degree of equation (8) decreases compared to equation (4) and this leads to better convergence when solving the system of two equations (8) and (9). Let the initial steam-gas mixture have the following qualitative composition:



The calculation will be carried out for 100 moles of the initial mixture. Let's denote:

- a – number of moles of CO in the initial mixture;
- b – Hydrogen;
- c – CO₂;
- d – Inerts (CH₄+N₂+Ar);
- m – CH₃OH;
- n – H₂O;
- x is the number of moles of CO that reacted according to reaction (3) to its equilibrium state;
- y is the number of moles of CO that reacted according to reaction (1) to its equilibrium state;
- P is the absolute pressure of the equilibrium mixture, atm;
- T is temperature, K.

At the equilibrium of reactions (1) and (3), the composition and partial pressures of the components of the equilibrium mixture will be expressed by the equations given in Table 1.

Table 1

The equation for calculating the composition of the equilibrium mixture

Component	Gas mixture, moles		Concentrations and partial pressures in an equilibrium mixture	
	initial	equilibrium	$[i]^*$, fraction	P_i^*
CO	a	$a-x-y$	$(a-x-y)/\Sigma p$	$(a-x-y) \cdot P/\Sigma p$
H ₂	b	$b+x-2y$	$(b+x-2y)/\Sigma p$	$(b+x-2y) \cdot P/\Sigma p$
CO ₂	c	$c+x$	$(c+x)/\Sigma p$	$(c+x) \cdot P/\Sigma p$
Inerts	d	d	$d/\Sigma p$	$d \cdot P/\Sigma p$
CH ₃ OH	m	$m+y$	$(m+y)/\Sigma p$	$(m+y) \cdot P/\Sigma p$
H ₂ O	n	$n-x$	$(n-x)/\Sigma p$	$(n-x) \cdot P/\Sigma p$
Sum	$\Sigma n = 100$	$\Sigma p = 100 - 2y$	1.0	P

Note: the table is compiled according to the generally accepted method of calculating equilibrium

Substituting the equilibrium partial pressures of the participants in reactions (1) and (3) into the equations of equilibrium constants (8) and (9), let's obtain a system of two nonlinear (relatively unknown x and y) equations:

$$Kp_1 = \frac{(m+y) \cdot (100-2y)^2}{(a-x-y) \cdot (b+x-2y)^2 \cdot P^2}; \quad (11)$$

$$Kp_3 = \frac{(c+x) \cdot (b+x-2y)}{(a-x-y) \cdot (n-x)}. \quad (12)$$

The equilibrium constants Kp_3 and Kp_1 are determined by the above formulas (7) and (10). Solve the system of equations (11) and (12) and determine the values of x and y (moles). The value of x should be negative – this explains the presence of water in the crude methanol. Substituting x and y in columns 3 and 4 of Table 1, determine the number of components (per 100 moles of initial gas) and the composition of the equilibrium mixture, %.

Let's note that for some values of the initial approximations, the system does not have a solution. But in general, the solution of the system of two equations (11) and (12) is not problematic in a sufficiently wide interval of initial approximations of x and y .

3. Research results and discussion

The calculations of the system of equations (11) and (12) using the values of the corresponding equilibrium constants according to equations (7) and (10) showed that the obtained results do not match well with the reference ones. This is explained by the fact that the value of the equilibrium constant Kp_1 was obtained at atmospheric pressure. At the same time, the CO steam conversion reaction takes place without a change in volume, and therefore, a change in pressure does not affect the shift in its equilibrium and the value of the equilibrium constant. In [4], the value of the equilibrium constant of reaction (1) at atmospheric and high pressures at different temperatures is given. This allows entering the coefficient C , approximated in the temperature range of 200–300 °C, which increases Kp_1 , for a pressure of 100 atm:

$$C = 3.584 - 0.0073 \cdot t. \quad (13)$$

By extrapolating this equation to pressures lower than 100 atm, good calculated values of equilibrium concentra-

tions were obtained – calculation 1 in Table 2. Thus, the relative maximum deviation for methanol (pressure 50 atm) is 8.53 %. This approach is simpler and more convenient than the correction for the non-ideality of the gas mixture, as provided by the Soave-Redlich-Kwong equation of state [10] or the use of reagent activity coefficients [11]. An attempt to calculate the equilibrium constant of the reaction (1) according to the equation [12]: $\lg Kp_1 = 5139/T - 12.621$ is unsuccessful, because the values of Kp_1 are even smaller than according to the equation (10).

In the second version of the calculation, in order to take into account the effect of pressure of 50–150 atm on the equilibrium constant of reaction (1), data [13] were used. According to this approach, the dependence of the equilibrium constant of reaction (1) has the form:

$$\lg Kp_1 = 3749/T - 9.28 \cdot \lg T + cc \cdot T + dd, \quad (14)$$

where the constants cc and dd depend on the pressure.

Approximate dependences of these constants on pressure P :

$$cc = 0.00000003 \cdot P^2 + 0.0028;$$

$$dd = 0.0077 \cdot P + 13.969.$$

According to the given initial data in Table 2 [1] the system of equations (11) and (12) was solved using the equilibrium constant Kp_1 according to equation (14). The results are given as calculation 2 in Table 2. For calculation 2, the relative maximum deviation for methanol (pressure 30 atm) is 16.25 %.

In general, calculation 1 using the correction coefficient C according to equation (13) gives results closer to the reference ones than calculation 2 using the two coefficients cc and dd according to equation (14). Satisfactory coincidence of the reference and calculation (calculation 1) data indicates the possibility of using the proposed method for calculations of the equilibrium of methanol synthesis under an average pressure of up to 9.8 MPa.

Table 2

Effect of pressure on the composition of the equilibrium mixture at 260 °C. Initial mixture, moles:
CO₂ $c=6$; CO $a=12$; H₂ $b=58$; Inerts (CH₄+N₂) $d=24$; H₂O $n=0$; CH₃OH $m=0$

Pressure, atm	Value	Equilibrium mixture, %					
	x/y	CO ₂	CO	H ₂	Inerts	H ₂ O	CH ₃ OH
30, [1]	–	6.11	8.47	52.98	26.57	0.53	5.34
30, calculation 1	–0.48/4.85	6.113	8.45	52.958	26.578	0.531	5.37
30, calculation 2	–0.454/4.105	6.042	9.096	53.749	26.146	0.495	4.472
50, [1]	–	6.27	6.38	50.24	28.02	0.73	8.36
50, calculation 1	–0.631/7.679	6.343	5.85	49.633	28.355	0.746	9.073
50, calculation 2	–0.607/7.355	6.323	6.158	50.044	28.139	0.712	8.624
75, [1]	–	6.29	4.24	47.09	29.06	1.11	11.67
75, calculation 1	–0.857/9.723	6.384	3.891	46.797	29.794	1.064	12.07
75, calculation 2	–0.907/10.02	6.37	3.608	46.337	30.016	1.134	12.534
100, [1]	–	6.11	2.93	44.66	30.72	1.57	14.01
100, calculation 1	–1.1/10.94	6.272	2.767	44.83	30.721	1.409	14.002
100, calculation 2	–1.335/11.73	6.094	2.101	43.387	31.353	1.744	15.32

For users, in order to facilitate the implementation of their own research, let's provide an algorithm for calculating the equilibrium state of methanol synthesis.

1. Enter the initial data: $P=30$; $t=260$; $T=t+273=533$; $c=6.0$; $a=12.0$; $b=58.0$; $d=24$; $n=0$; $m=0$.

2. Calculate the equilibrium constants Kp_1 and Kp_3 (first the decimal logarithms according to equations (10) and (7), and only then the constants).

3. Calculate the coefficient C according to equation (13).

4. Adjust the equilibrium constant $Kp_1=Kp_1 \cdot C$.

5. Set initial approximations $x=-0.5$, $y=7$. It is possible to vary these values in MathCad refuses to solve the system, or gives a result outside the real limits. If the system is not solvable, it is necessary to change the initial approximations.

6. Solve the system of equations (11), (12) with respect to x , y using Given and Find operators, between which these equations are entered. To simplify input, use simple identifiers: Kp_3 , Kp_1 .

Equality in equations is written using the «= \Rightarrow » operator (Ctrl and = at the same time). To calculate the values of x , y , assign the Find function to the solution vector: $Z:=Find(x, y)$. Assign $x:=Z_0$; $y:=Z_1$ (enter 0, 1 as indices!) in order to avoid further calculation with the initial approximations of these x and y values given in clause 5.

7. Calculate the equilibrium amount (moles) according to the formulas of Table 1 – the third column: $a_1=a-x-y$; $b_1=b+x-2y$; $c_1=c+x$; $d_1=d$; $m_1=m+y$; $n_1=n-x$; $S=a_1+b_1+c_1+d_1+m_1+n_1$.

Calculate the equilibrium gas composition (%) according to the formulas of Table 1 – the fourth column with multiplication by 100: $A^*=a_1 \cdot 100/S$; $B^*=b_1 \cdot 100/S$; $C^*=c_1 \cdot 100/S$; $D^*=d_1 \cdot 100/S$; $M^*=m_1 \cdot 100/S$; $N^*=n_1 \cdot 100/S$.

The algorithm and the program according to the proposed algorithm take into account the multivariate nature of the initial data, namely the possibility of varying the composition of the initial mixture, pressure and temperature values.

At the second stage of the research, material and equilibrium calculations were performed according to the following initial data (Table 3), which correspond to the conditions of industrial implementation – methanol synthesis: pressure in the synthesis column $P=50$ atm; temperature at the outlet of the synthesis column (SC) $T=553$ K.

Table 3
Output data (column 2) and calculation results

Component	Concentration at the entrance of SC, %	Concentration at the outlet of SC, %	Equilibrium concentration, %
CO ₂	22.17	22.4	24.148
H ₂	45.96	42.7	38.203
CO	21.28	20.7	18.465
Inerts	10.1	10.6	11.39
CH ₃ OH	0.41	2.7	6.849
H ₂ O	0.09	0.9	0.955

A comparison of the equilibrium composition of the gas (column 4) with the actual one at the output of the synthesis column (column 3) shows the degree of reaching equilibrium with methanol $2.7/6.849=0.394$. Note that the authors of [4] note the value of this parameter in the range (0.275–0.44).

The low value of the degree of reaching equilibrium indicates the presence of a reserve of methanol production and the possibility of increasing the practical yield of methanol, which is determined by the process parameters and the activity of the catalyst. The concentration parameters of the source gas for the synthesis of methanol, in turn, are determined by the conditions of steam-carbon dioxide conversion of natural gas and the conditions of methanol synthesis, in particular, the amount of system purging. The yield of methanol in the existing production can be increased by increasing the contact time (reducing the volumetric velocity) and using more active catalysts. But this may reduce the performance of the synthesis column. Therefore, it is necessary to solve the optimization problem: the selection of synthesis parameters that affect the increase in methanol concentration without reducing the productivity of the synthesis column.

4. Conclusions

The relative deviation of the reference and calculation data for methanol in the range of 0.06–8.53 % indicates the complete possibility of using the proposed method and algorithm for calculating the equilibrium of methanol synthesis under an average pressure of up to 9.8 MPa. A low value of the degree of reaching equilibrium in the industrial column indicates the presence of a reserve of methanol production and the possibility of increasing the practical yield of methanol, which is determined by process parameters and catalyst activity. The proposed algorithm takes into account the multivariate nature of the initial data, namely the possibility of variation in the composition of the initial mixture, pressure and temperature values.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this study, including financial, personal, authorship, or any other, that could affect the study and its results presented in this article.

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