Journal of Physics: Conference Series

PAPER • **OPEN ACCESS**

Mathematical modeling of technological process in formalin production

To cite this article: T V Botirov *et al* 2021 *J. Phys.: Conf. Ser.* **2094** 022052

Mathematical modeling of technological process in formalin production

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Abstract. Methods and algorithms of the sustainable synthesis of adaptive control systems for the technological process of formalin production have been proposed in this article. Parameters of the regulator are estimated in the developed adaptive control system, i.e. the purpose of adaptation is reduced to the estimation of coefficients of the regulator providing the specified behavior of the system. The results obtained and the analysis of transition processes based on the modeling of the control system showed that with adaptive control, mass fractions of formaldehyde and methanol in formalin are held within 36.5÷37.5% and 0.7÷0.9% respectively, while the specific consumption of the methanol decreases by 3÷4%, which allows the process in mode close to the optimal.

Mathematical models of chemical and technological processes can be divided into two groups: theoretical physical-chemical models and empirical statistical models.

We will consider the characteristic features of mathematical models of the first group regarding the production of formalin. A mathematical model of the formaldehyde synthesis process is described in [1, 2], which is a system of equations for material and heat balances on the catalyst grain:

$$\begin{aligned} &-\chi\frac{\partial\overline{C}}{\partial l}+b(\overline{Y}-\overline{C})=e\frac{\partial\overline{C}}{\partial t}\\ &-\chi\frac{\partial T}{\partial l}+\frac{a}{C_p}S_{УД}\left(T_з-T\right)=e\frac{\partial T}{\partial t};\\ &b(\overline{Y}-\overline{C})-\overline{W}=e_k(1-e)\frac{\partial\overline{Y}}{\partial t}\end{aligned} \tag{1}$$

$$\frac{\lambda_k}{C_p}\frac{\partial^2 T_з}{\partial l^2}+\frac{a}{C_p}S_{sp}+\frac{1}{C_p}\sum_{j=1}^{m}\left(-\Delta H_j\right)W_j=\frac{\lambda_k}{C_p}(1-e)\frac{\partial T_з}{\partial T}$$

where χ is the linear speed of the gas flow; $\overline{C}$ is the concentration of components; $\overline{Y}$ is near-surface concentrations; $\overline{W}$ is transformation speed of components; T is temperature in the flow; T_3 is temperature of the catalyst grain; ΔH_j, $j=\overline{1,m}$ is thermal effect of the *j*-th reaction; C_k is heat capacity of the catalyst; C_p is heat capacity of gas; a is coefficient of heat transfer from the grain surface to the gas flow; λ_k is coefficient of thermal conductivity of the catalyst layer; S_{sp} is specific surface of the granular layer; b is the coefficient of mass transfer; e_k is the porosity of the catalyst grain; e is the porosity of the catalyst layer.

The system of equations (1) was used to optimize the process of incomplete oxidation of methanol to formaldehyde in a reactor with a fixed layer of a pumice-silver catalyst [3]. In [4], a model of a combined reactor for the oxidation of methanol to formaldehyde has been developed, considering the presence of uneven profile of the speed, temperature and concentration of gas flow components at the entrance to the contact layer, as well as the presence of heterogeneities in the permeability and catalyst activity in the layer:

$$
\begin{gathered}
\frac{\partial P}{\partial z} = ueB; \frac{\partial P}{\partial x} = veB; \frac{\partial}{\partial z}(upe) + \frac{\partial}{\partial z}(vpe) = 0; \\
\lambda_z \frac{\partial^2 T}{\partial z^2} + \lambda_x \frac{\partial^2 T}{\partial x^2} - C_p \frac{\partial}{\partial z}(upet) - C_p \frac{\partial}{\partial x}(vpet) + (1-e)\sum_{j=1}^{m}\left(-\Delta H_j\right)W_j = 0 \\
D_z \frac{\partial^2 C_i}{\partial z^2} + D_x \frac{\partial^2 C_i}{\partial x^2} \frac{\partial}{\partial z}(ueC_i) - \frac{\partial}{\partial x}(veC_i) + (1-e)\sum_{j=1}^{m}\left(V_{ij}W_j\right) = 0 \\
p = \frac{P_0}{\left[1 + 0{,}0036\,(T - T_0)\right]}; M = M_0 \frac{T_0 - 111}{T + 111}\left(\frac{T}{T_0}\right)^{\frac{3}{2}}; \\
B = \frac{150\,M\,(1-e)^2}{d_3^2 e^3} + \frac{1{,}75\,p(1-e)}{d_3}\sqrt{(ue)^2 + (ve)^2}
\end{gathered}
\tag{2}
$$

where i is the index of the substance; j is the reaction index; P is the pressure; z, x are longitudinal and transverse coordinates of the layer; u, v are longitudinal and transverse speeds; p_0, p are the densities of the reaction mixture, the initial and in the catalyst layer; D, λ_z are effective coefficients of diffusion and thermal conductivity, respectively; T_0, T are temperatures of the reaction mixture, the initial and in the catalyst layer; c_i is concentrations of the components of the reaction mixture in the catalyst layer; V_{ij} is the stoichiometric coefficient of the i-th substance in the j-th reaction; M_0, M are viscosities of the reaction mixture, the initial and in the catalyst layer; d_3 is the diameter of catalyst grain.

The system of equations (2) was used to calculate the parametric sensitivity of tubular and combined reactors, as well as to assess the influence of a wide class of spatial inhomogeneity on the characteristics of catalytic processes in apparatus of this type.

Analysis of the theoretical mathematical models described above [5,6] from the point of view of their applicability for the development of controlling algorithms showed the following:

The models fully reflect the basic physical and chemical patterns of formalin production processes and, in the absence of disturbances, are adequate to the described processes.

Because of the fact that perturbations, i.e. decrease in catalyst activity, changes in methanol quality indicators, etc. are constantly affect the process of formalin production, the model must either take them into account explicitly, which significantly complicates the mathematical description of the chemical-technological processes of formalin production, or use the algorithm for adapting the model parameters according to experimental data. These computational operations are very laborious both because of the high dimension of the problem and because of the nonlinear dependence of the parameters of the model, therefore, theoretical mathematical models, in the general case, are inappropriate to use in control algorithms, they should be used for the design of devices, optimization of technological schemes, development simplified management models [7].

The characteristic property of empirical mathematical models of technological processes of the formalin production is the following. In connection with the above-mentioned complexity of chemical-technological processes for the purposes of control and optimization of production, they are often limited to their simplified description in the form of finite equations. In this case, a mathematical description of the static modes of production is considered. In the general case, the target criterion for optimality and control of the formalin production process is that the *Y* – formaldehyde yield depends on a number of process variables $X_1, X_2,\ldots,X_n$.

There is a general functional connection between them:

$$Y = f\left(X_1, X_2,\ldots, X_n\right), \tag{3}$$

which admits expansion in Taylor series

$$Y = b_0 + \sum_{i=1}^{n} b_i x_i + \sum_{i,j=1}^{n} b_{ij} x_i x_j + \sum_{i,j=1}^{n} b_{ij} x_i^2 , \tag{4}$$

where Y is the objective function; X_i, X_j are process parameters; b_0, b_1, b_{ij}, b_{ii} are regression coefficients. When optimizing and controlling technological processes of formalin production, both empirical equations based on expression (3) and regression dependences based on the expansion of the objective function in a Taylor series (4) are used

Among the works using regression models of the form (4), the work [8] should be noted, in which a model is proposed that relates the formaldehyde yield to the oxygen/methanol ratio at the inlet to the contact apparatus:

$$Y = K_0 + K_1 X , \tag{5}$$

where *Y* is the formaldehyde yield; *X* is the oxygen/methanol ratio; K_0 , K_1 are regression coefficients. To adapt model (5) to a decrease in catalyst activity and other disturbances of the technological process, the Kachmazh algorithm [3] has been used:

$$\begin{aligned}
K_0(n) &= K_0(n-1) + \frac{Y(n) - (K_0(n-1) + K_1(n-1)X(n))}{(Y(n) - \overline{Y}(n))^2 + 1 + X(n)^2} \\
K_1(n) &= K_1(n-1) + \frac{Y(n) - (K_0(n-1) + K_1(n-1)X(n))}{(Y(n) - \overline{Y}(n))^2 + 1 + X(n)^2} \\
\overline{Y}(n) &= \frac{n-1}{n}\overline{Y}(n-1) + \frac{1}{n}Y(n),\ n = 1,2,3,\ldots,
\end{aligned} \tag{6}$$

where *n* is the number of the control cycle; *Y* is the current value of the formaldehyde yield; *Y* is the average value of the formaldehyde yield. Model (5) together with the adaptation algorithm of coefficients (6) was applied for optimal control of formalin production through the channel “oxygen/methanol ratio - formaldehyde yield” [9,10].

Analysis of empirical mathematical models of chemical-technological processes from the point of view of the possibility of their application in automated control systems for the production of formalin shows that:

- the models do not reflect the physical-chemical laws of the formalin production process as fully as the theoretical physical-chemical models. Empirical models are adequate to the described process only in a narrow range of parameter variation.

- the adaptation of the parameters of these models is not difficult, for which, for example, regression methods of identification are used. In addition, the observed indicators of the quality of raw materials and catalyst can be included in the model in an explicit form without significantly complicating it.

Thereby, empirical models of chemical technological processes can be directly used in adaptive algorithms for optimization and control of formalin production, while theoretical mathematical models are advisable to use to improve the hardware and technological design of processes and obtain simplified control models when developing such algorithms.

The main parameters of technological processes for the production of formalin are [11]:

- at the stage of preparation of the alcohol-air mixture: methanol consumption; air consumption; gas consumption; blowing consumption; the ratio of air and methanol in an alcohol-air environment; volume fraction of oxygen in the blowing; the level of methanol in the alcohol evaporator; the temperature of the alcohol-air mixture after the alcohol evaporator; temperature of the alcohol-air mixture at the entrance to the contact apparatus;
- at the stage of catalytic synthesis: temperature in the tubular of the contact apparatus; temperature in the adiabatic layer of the contact apparatus; thermolan temperature at the inlet of the contact apparatus; thermolan temperature at the outlet of the contact apparatus;
- at the stage of formaldehyde absorption: consumption of cooling water in the shell space of the refrigerator; distilled water consumption; supply of reaction gases; the volume fraction of formaldehyde in the blowing; formalin yield with a given formaldehyde and methanol content; temperature of formalin in bubbling refrigerator.

The research results of technological process of formalin production, carried out in the conditions of “Navoiazot” JSC, show that their static and dynamic characteristics, both in control and disturbing channels, vary within wide limits in an unforeseen manner (due to changes in the quality of initial raw material, intensity of modes, etc.). Qualitative regulation of these objects requires the use of a control system that includes an adaptation loop to the changed parameters of the object, which compensates for the influence of the latter. For many technological processes, such laws are sought in the form of functions of time corrected in the course of the process [12].

Based on a preliminary study and formalization of the process under consideration from the point of view of control tasks, it has been determined that the main indicators characterizing the process of obtaining formalin are the following variables:

- Control parameters_$\mathrm{U}=(u_1,u_2)$, where u_1 is the cooling water consumption; u_2 is the temperature of the thermolan at the inlet to the contact apparatus;
- Output parameters $\mathrm{Y}=(y_1,y_2)$, where y_1 is the formalin yield with a given formaldehyde and methanol content; y_2 is the formalin temperature in the bubbling refrigerator;
- Uncontrolled disturbing influences $\mathrm{W}=(w_1,w_2,w_3)$, where w_1 is fluctuations in the composition of raw materials; w_2 is the decrease in the activity and selectivity of the catalyst;
- w_3 is the enthalpy of the coolant and heat loss. In this case, the ultimate objective of control is to obtain a given amount of formalin with a content of 37±0.5% weight of formaldehyde.

Below are the dynamic characteristics of the process of obtaining formalin through the main channels of transmission of influences based on experimental research methods:

1. $u_1 \to y_1$: $(192\,p^2+63{,}8p+1)y_1(t)=0{,}021u_1(t)$;

2. $u_1 \to y_2$: $(211{,}2\,p^2 + 72{,}5\,p + 1)\,y_2(t) = (0{,}005\,p + 1)\,u_1(t)$;

3. $u_2 \to y_1$: $(131\,p^2 + 28\,p + 1)\,y_1(t) = (0{,}017\,p + 1)\,u_2(t)$;

4. $u_2 \to y_2$: $(148{,}9\,p^2 + 67{,}4\,p + 1)\,y_2(t) = 0{,}0035\,u_2(t)$.

Analysis of the obtained dynamic models shows that the most significant channel of the object under consideration is $u_2 \to y_1$ “temperature of thermolan at the inlet to the contact apparatus - formalin output with a given content of formaldehyde and methanol” channel. This is conditioned that this channel has the highest gain coefficient and the lowest lag effect. For this reason, the synthesis of the control system for the formalin production process will be carried out based on the control parameter, i.e. the temperature of the thermolan at the inlet to the contact apparatus.

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