SELECTION OF THERMAL OPERATING REGIMES FOR FUEL CELL REACTOR—CONDENSER SYSTEMS

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UDC 536.248.2:541.136.8

The effect of reactor—condenser system operating regime on fuel cell efficiency is studied.

The contemporary state of theoretical and experimental studies on mass output permits the calculation of mass transfer devices (condensers, evaporators) with satisfactory accuracy.

However, if the condenser operates with an evaporator in a closed system in which a complete change of aggregate state of the working substance does not occur, the two devices affect each other according to the inverse feedback principle. This inverse feedback is especially well manifested in the operation of the oxygen—hydrogen fuel cell in which the moisture formed by electrochemical reaction G_r is evaporated from the membrane surface into the vapor—hydrogen mixture circulating in the reactor—condenser system and then led to the condenser. Thus we have the mass transfer process

$$G_r = mF_c \overline{\Delta p}, \tag{1}$$

where $\overline{\Delta p}$ is the mean integral partial pressure head, determined in the first approximation in analogy with the heat exchange function; for the condenser

$$\overline{\Delta p}_c = \frac{p_c^{in} - p_c^{out}}{\ln \dfrac{p_c^{in} - p_{sc}}{p_c^{out} - p_{sc}}}, \tag{2}$$

$$m = \frac{1}{\dfrac{1}{\beta_c} + \dfrac{1}{\beta_r} \cdot \dfrac{F_c}{F_r}}. \tag{3}$$

To ensure mass transfer the mixture must circulate through the circuit at a definite flow rate which is characterized by the multiplicity of the circulation:

$$k = \frac{G^{out,c} + G_h}{G_h}. \tag{4}$$

An additional relationship between the partial vapor pressures at the condenser input and output and the multiplicity of the circulation may be obtained from the equation of state

$$p_c^{out} = \frac{p^2 - kpp_c^{in}}{8p_c^{in} - p(k+7)}. \tag{5}$$

To ensure stationary operation of the fuel cell the mass expenditures must be balanced:

$$G_c = G_r = 9G_h. \tag{6}$$

The area of the reactor surface F_r is chosen from the permissible current density and the required power level. Usually $F_r \gg F_c$ and for $\beta_r \geq \beta_c$ we have $\beta_r F_r \gg \beta_c F_c$ which means $m \approx \beta_c$ and

$$G_r = G_c = \beta_c F_c \overline{\Delta p}_c. \tag{7}$$

This also means that the difference between the partial vapor pressure in the mixture p_r and the pressure on the surface p_{sr}

$$\Delta p_r = p_r - p_{sr} = \frac{j_r}{\beta_r}$$

Odessa Technological Institute of the Refrigeration Industry. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 34, No. 4, pp. 642-647, April, 1978. Original article submitted March 10, 1976.

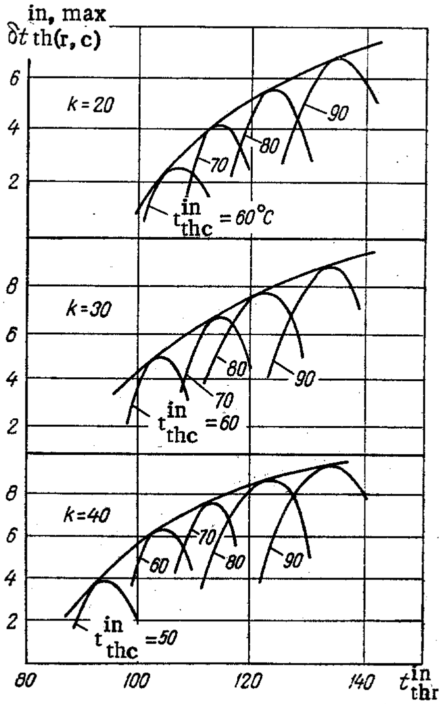

Fig. 1. Admissible deviation of temperature (°C) at reactor input from reactor temperature for various condenser temperatures. I = 100 A, $\beta_c F_c = 3 \cdot 10^{-4}$ m²/h.

in any desired section of the reactor is extremely small (because of the low flux density of the evaporating water j_r for sufficiently large section β_r); i.e., it can be assumed that any given section $p_r = p_{sr}$ and, in particular, $p_r^{in} = p_{sr}^{in}$, and $p_r^{out} = p_{sr}^{out}$. Since because of the closed nature of the system the quantities p_r^{in} and p_r^{out} are uniquely determined by the mass exchange parameters $\beta_c F_c$, the temperature of the condenser surface t_{sc} and the reactor surface t_{sr}, the current density i, and the multiplicity of the circulation k, these same parameters uniquely determine the quantities p_{sr}^{in} and p_{sr}^{out} and the intermediate values of p_{sr}. By virtue of the function

$$f(p_{sr},\, t_{sr},\, c_{sr}) = 0 \tag{8}$$

the mass exchange parameters uniquely determine the values of electrolyte concentration on the surface of the hydrogen electrode. This concentration c_{sr}, determined by solution of the system of equations (2), (4)-(8), should serve as a boundary condition for finding the concentration distribution over thickness of the electrochemical group.

The quantity t_{sr} is larger than the temperature of the thermostating liquid t_{thr} due to thermal resistance to heat transfer in the reactor. The value of t_{thr} increases with motion of the thermostat liquid and depends on the flow rate of the latter, G_{thr}, the load on the fuel cell, and the peculiarities of operation of the thermostat system.

Moisture withdrawal in the condenser depends on the value of the complex $\beta_c F_c$, the flow rate of the thermostat liquid G_{thc}, and its temperature t_{thc}, on the thermal resistance offered to the heat transfer process in the condenser, on the construction of the condenser, and on the operation of the thermostat system. The coefficient β_c in turn depends on the mixture flow rate, i.e., on the multiplicity of the circulation k.

The thermal resistance to heat transfer in the condenser was calculated for a full-scale fuel cell, and the mean (over area) condenser surface temperature was determined as a function of the current load for constant pumping rate of the thermostat liquid. Analogous information for the reactor was obtained experimentally.

Changes in the temperature of the thermostat liquid at the reactor and condenser inputs produced by nonuniformity in thermostat system operation play an important role, since other conditions being equal these changes cause a redistribution of partial water vapor pressures in the system, which can lead to critical situations due to limitations on buffering and crystallization.

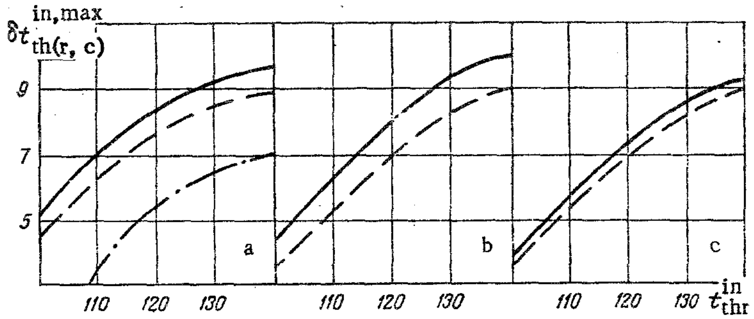

Fig. 2. Optimum admissible temperature deviation (°C) at reactor input versus reactor temperature at I = 100 A for various $\beta_c F_c$ (m^2/h) and circulation multiplicities: solid curve, k = 40; dashes, k = 30; dash—dot curve, k = 20; a) $\beta_c F_c = 3 \cdot 10^{-4}$; b) $5 \cdot 10^{-4}$; c) $1 \cdot 10^{-3}$.

We have attempted to determine the possible admissible variation in the temperature of the thermostat liquid at the reactor and condenser inputs $\delta^{in,max}_{th(r,c)}$. For convenience in computer calculation these variations were specified in steps identical in size and direction for reactor and condenser, since it was assumed that the fuel cell would be provided with automatic temperature regulators of identical characteristics.

With consideration of the above, the system of Eqs. (2), (4)-(8) was solved by sampling of values of the following quantities:

1) the complex $\beta_c F_c = 3, 5, 10, 20 \cdot 10^{-4}$ m^2/h;
2) current load I = 25, 50, 75, 100 A (corresponding current density values i = 50, 100, 150, 200 mA/cm²);
3) thermostat liquid temperatures: at reactor input t^{in}_{thr} 80-140°C and at condenser input t^{in}_{thc} = 40-100°C in 10°C steps;
4) changes in thermostat liquid temperature at reactor input δt^{in}_{thr} = 10, 8, 6, 4, 2°C; changes in thermostat liquid temperature at condenser input δt^{in}_{thc} = 10, 8, 6, 4, 2°C;
5) circulation multiplicity k = 20, 30, 40.

Calculations were performed for a full-scale fuel cell with matrix chemical groups at a membrane thickness δ = 1 mm.

The range of parameter variation was made significantly broader than that of the full-scale fuel cell in order to determine general principles of parameter interaction for possible thermal readjustments of the fuel cell.

Using the values of p^{in}_r, p^{out}_r, t^{in}_{thr}, t^{out}_{thr} the concentrations c^{in}_s and c^{out}_s on the membrane surface on the hydrogen side were determined. Then using these as boundary conditions in the equation for concentration distribution over electrode thickness, values of the concentrations $\bar{c}^{in}$ and $\bar{c}^{out}$ averaged over membrane section were obtained. Then with consideration of limitations on buffering and crystallization in the full-scale electrodes, variants ensuring fuel cell efficiency were chosen.

The functional relationships in Eq. (8) were approximated by polynomials and power functions, and the boundary conditions were set in the form of inequalities, then used to compose an algorithm for computer calculation of variants.

The processing results were obtained in the form of graphical functions $\delta^{in,max}_{th(r,c)} = f(t^{in}_{thr})$ for each combination of $\beta_c F_c$, I, k values. A characteristic example for $\beta_c F_c = 3 \cdot 10^{-4}$ m^2/h and I = 100 A is shown in Fig. 1.

Analysis of the results confirmed the presence of a relationship between reactor and condenser thermal regimes outside the region of concentration value criticality. This function has an optimum point; i.e., for each selected reactor temperature t^{in}_{thr} there corresponds an optimum simultaneous deviation of t^{in}_{thc} for the condenser.

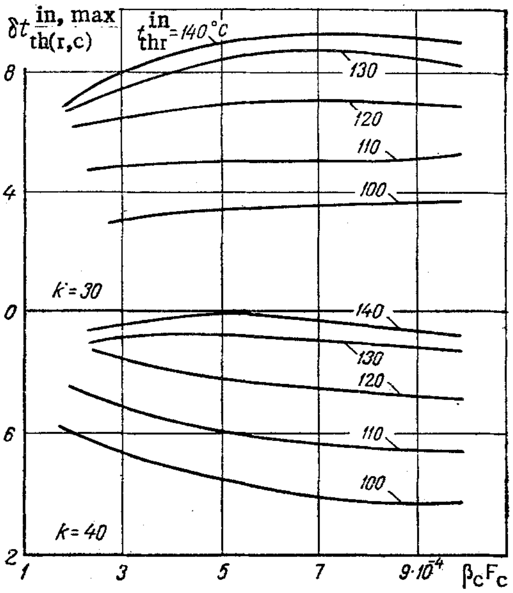

Fig. 3. Optimum permissible temperature deviation (°C) at reactor input versus complex $\beta_c F_c$ (m²/h).

It can be maintained that a similar type of inverse feedback should appear for any type of fuel cell with a closed circuit and extraction of excess moisture by use of heat—mass transfer processes. In the type of fuel cell considered here the feedback appears more clearly due to limitations on buffering.

The temperature of the thermostat liquid at the reactor input should then be chosen as high as possible with consideration of conditions of electrolyte existence and capabilities of the construction materials, as was done for the full-scale fuel cell.

It was established that the optimum thermostat liquid temperature at the condenser input t^{in}_{thc}, selected from t^{in}_{thr}, was practically independent of the multiplicity of circulation k (Fig. 1). However, with increase in k the maximum admissible temperature deviation $\delta^{in,max}_{th(r,c)}$ increases, as is evident from Fig. 2, which shows the envelopes of the optimum thermostat liquid temperatures at the condenser input. The greatest increment $\delta t^{in,max}_{th(r,c)}$ is observed in the region from k = 20 to k = 30.

At low tract resistances these coefficients can be provided by an injector, as shown by additional experimental studies [1].

Construction of the dependence of maximum admissible deviation $\delta t^{in,max}_{th(r,c)}$ on the complex $\beta_c F_c$ (Fig. 3) makes it possible to conclude that at low k the optimum value of the complex $\beta_c F_c$ is independent of t^{in}_{thr} and k, but that with increase in t^{in}_{thr} and k an optimum appears in $\beta_c F_c$; at t^{in}_{thr} = 140°C the optimum value of $\beta_c F_c$ = 5–7 at k = 30–40. These data permit structural calculation of the optimum area of the condenser using the method of [2, 3, 4]. However, any change in condenser construction or thermostat liquid pumping rate will produce corresponding changes in the condenser surface temperature and require a new calculation.

In the present study the goal in condenser area optimization was simultaneous attainment of the maximum admissible deviation of the temperature of the thermostat liquid at the input of the reactor and condenser, but one could also consider problems of temperature deviation by any other prespecified rule. Moreover, condenser optimization for other goals could be considered, for example, with respect to weight and size considerations, or with respect to some complex of goals. The existence of various methods of affecting concentration distribution in the membrane electrolyte makes it possible to optimize not only the condenser, but also the reactor, with respect to goals chosen beforehand.

The method on which the calculations were based and the subsequent analysis used here suffer from a number of inaccuracies and are based on some assumptions.

1. The definition of the mean integral pressure head as a mean logarithmic quantity, destroying the analogy between the processes of heat and mass exchange, can lead to significant errors.

2. The method of averaging wall temperatures and membrane concentrations as mean arithmetic quantities is only approximate.

3. The method of calculating concentration distribution over membrane thickness does not take into account the convective component of transfer.

However, despite the shortcomings connected with the approximate nature of the calculations, the method used is correct in principle, and the results obtained do give a qualitatively correct picture, without a doubt. A more refined technique will naturally produce more accurate results.

NOTATION

G, weight flow rate; F, surface area; m, mass transfer coefficient, referred to condenser surface; $p_{r,c}$, partial vapor pressure in mixture; p, total mixture pressure; β, mass transfer coefficient, referred to partial pressure; Δp, partial pressure heat; j, vapor transverse mass flow density; c, electrolyte concentration; t, temperature; δt, temperature variation; i, current density; I, load current. Subscripts and superscripts: r, in reactor; c, in condenser; s, on surface; h, hydrogen in reaction; m, mixture; th, thermostat liquid; in, input; out, output.

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EFFECTIVE CONDUCTIVITY OF HETEROGENEOUS SYSTEMS WITH DISORDERED STRUCTURE

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UDC 536.24

A model reflecting the disordered character of the structure of a heterogeneous system is proposed, together with a method for calculating the coefficients of generalized conductivity* of compounds, eutectics, composites, and solutions.

We will consider the simplest possible two-component heterogeneous system with chaotic structure, formed, for example, by the pressing of a mixture of two different powders of compact particles. We will limit our consideration to the case of mechanical mixture of noninteracting components which preserve their original properties both within the volume and at the phase boundary.

Use of the concept of "disordered" structure assumes that in regions of the system with dimensions significantly exceeding the dimensions of the original particles the values of the volume concentrations of the components m_i, m_j are practically indistinguishable from the average values of those quantities over the entire system volume, while the system properties are isotropic within the limits of the region and over the system as a whole, although local deviations from mean values may occur.

We will attempt to develop a model reflecting the random (probability) character of the distribution of the individual components within the volume of the two-component system with disordered structure, and to determine its effective conductivity.

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*The coefficients of generalized conductivity are the coefficients of thermal conductivity, electrical conductivity, dielectric permittivity, and magnetic permeability, the determination of which is the object of the theory of generalized conductivity.

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 34, No. 4, pp. 648-655, April, 1978. Original article submitted April 25, 1977.