Sorption of carbon dioxide in a fixed bed of the molecular sieve Calsit 5. I. Verification of the model of equilibrium adiabatic sorption with the modified Langmuir equation

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The model of equilibrium adiabatic sorption with the modified Langmuir equation is verified by the results of laboratory experiments involving adsorption of carbon dioxide in a bed of the molecular sieve Calsit 5 from the stream of indifferent gas and desorption of carbon dioxide by the stream of nitrogen. The confrontation of experimental course of sorption with the model idea has shown that the equilibrium adiabatic model in intermediate and turbulent flow regime may be used for prediction of the break point time in the stage of adsorption as well as the desorption time and the consumption of indifferent gas in the stage of desorption if kinetic data concerning the investigated system are lacking.

Модель равновесной адиабатической сорбции с модифицированным уравнением Лэнгмюра была применена к результатам лабораторных экспериментов адсорбции углекислого газа в слое молекулярного сита Кальсит 5 из потока индифферентного газа и десорбции углекислого газа током азота. Из сравнения экспериментального хода сорбции с моделируемым вытекает, что в отсутствии кинетических данных можно в случае изучаемой системы использовать равновесную адиабатическую модель в переходном и турбулентном режиме потока для расчета времени проскока, времени десорбции и расхода индифферентного газа в фазах адсорбции и десорбции соответсвенно.

Sorption in a fixed bed of adsorbent is a complex process involving unsteady mass and heat exchange. Its simulation is based on the system of partial differential equations resulting from equations of material and heat balance, rate equations of mass and heat transfer and a convenient equation of adsorption equilibrium. In general, such system of differential equations is not analytically solvable. Analytical solutions proposed by some authors are based on different simplifications among which the most important assumptions are: isothermal conditions of adsorption, linearity of adsorption isotherm, prevailing resistance to mass transfer in the fluid or solid phase. Papers [1—14] are typical studies of this kind. The influence of

adsorption heat on the course of process is taken into account in several papers published in last years [15—37]. The system of simulation equations describing such course of adsorption is numerically solvable, but the consumption of time necessary for calculation is from practical view-point too great even if an efficient computer is used [19]. In this state, asymptotic solutions which represent a compromise between adequacy and applicability are much sought after. Studies of this kind have been also published [17, 20, 27, 28].

The research of adsorption in a fixed bed of adsorbent carried out in our laboratory is governed by the research programme the aim of which is to investigate, step by step, the significance of distinct partial processes that affect the course of the integral process. It is expected that this procedure will enable to find the way in the current boom of information and elaborate the directives useful for common engineering practice.

The first step in this research programme is the study of a physical model which should serve as reference model. As such, the model of adiabatic equilibrium adsorption has been chosen, because of great amount of heat which is released in large-diameter commercial adsorbers especially when the adsorptive concentration in the feed is relatively high. This heat is influencing dominantly the adsorption equilibrium. In this case, the adsorption is neither isothermal nor adiabatic process. It can be qualified as a nonisothermal process. The distinction of a nonisothermal process from adiabatic one is given by the heat loss into surroundings. The adiabatic reference model enables to appreciate the influence of this factor, too.

The equilibrium adiabatic model with the modified Langmuir equation

The separation of adsorptive from a stream of indifferent gas in a fixed bed of adsorbent, or the desorption of adsorbate by a stream of nonadsorbing gas can be described under certain simplifying conditions [38] by the following equations

$$w\left(\frac{\partial c}{\partial z}\right)_\tau + \varepsilon\left(\frac{\partial c}{\partial \tau}\right)_z + \varrho_b\left(\frac{\partial a}{\partial \tau}\right)_z = 0 \tag{1}$$

$$w\left(\frac{\partial T}{\partial z}\right)_\tau + \varepsilon\left(\frac{\partial T}{\partial \tau}\right)_z + \frac{\varrho_b c_{pb}}{\varrho c_p}\left(\frac{\partial T_b}{\partial \tau}\right)_z - \frac{\varrho_b(-\Delta H)}{\varrho c_p}\left(\frac{\partial a}{\partial \tau}\right)_z = 0 \tag{2}$$

The equilibrium in the system is described by Langmuir equation [39] modified to the form

$$a^* = A \exp(-BT_b) \frac{c^*}{\dfrac{\sqrt{T_b}}{K \exp\left(\dfrac{R}{T_b}\right)} + c^*} \tag{3}$$

For equilibrium adsorption, it holds

$$a^* = a, \; c^* = c, \; T_b = T \tag{4}$$

The system of eqns (*1—3*) with conditions (*4*) can be solved numerically for certain initial and boundary conditions.

If a bed of adsorbent is initially saturated to the concentration a_p at the temperature T_p and a gas stream with the adsorptive concentration $c_0 > c_{poc}$ enters the bed of adsorbent at the time $\tau > 0$ (c_p is the equilibrium concentration with respect to a_p at the temperature T_p) the initial and boundary conditions can be expressed as follows

$$\begin{gathered} c(0,z) = c_{poc}, \quad T(0,z) = T_p, \quad a(0,z) = a_p \\ c(\tau,0) = c_0, \quad T(\tau;0) = T_0 \quad \text{for } \tau > 0 \end{gathered} \tag{5}$$

The above initial and boundary conditions may be also used for desorption with a stream of gas. In this case, the condition $c_0 < c_{poc}$ holds for the inlet concentration of adsorptive.

By introducing parameters

$$W = \frac{w}{\varepsilon}, D = \frac{\varrho_b}{\varepsilon}, E = \frac{\varrho_b(-\Delta H)}{\varrho c_p \varepsilon}, F = \frac{\varrho_b c_{pb}}{\varrho c_p \varepsilon} \tag{6}$$

and conditions (*4*) and neglecting accumulation of adsorptive and heat in the fluid phase, the system of eqns (*1—3*) can be transformed into two differential difference equations

$$\frac{dc_i}{d\tau} = -\frac{W}{hF\sqrt{T_i}} \left\{ c_i \left[B\left(\sqrt{T_i} + c_i K \exp\left(\frac{R}{T_i}\right)\right) + \frac{T_i + 2R}{2T_i\sqrt{T_i}} \right] \cdot \right.$$
$$\left. \cdot \left[T_i - T_{i-1} + \frac{E}{D}(c_i - c_{i-1}) \right] + \frac{FK \exp\left(\frac{R}{T_i}\right)\left(T_i \exp\left(\frac{-R}{T_i K}\right) + c_i\right)^2}{DA \exp(-BT_i)} (c_i - c_{i-1}) \right\} \tag{7}$$

$$\frac{dT_i}{d\tau} = -\frac{W}{hF}\left[\frac{E}{D}(c_i - c_{i-1}) + T_i - T_{i-1}\right] \tag{8}$$

which may be solved by simulation on a digital computer.

Computation of concentrations and temperatures in a bed of adsorbent

Calculation of concentrations and temperatures in distinct places along the bed and in time intervals suited for the conditions of experiments was performed on a digital computer Siemens 4004/150 according to the adiabatic equilibrium sorption model. The programmes in the simulation language SIKOS were based on the system of eqns (*7*) and (*8*). In contrast to definition given by eqn (*6*), the heat

capacity of the adsorber was included into the value of parameter F according to the following equation

$$F = \frac{\varrho_b c_{pb} + \left(\frac{S_1}{A_1}\right) \varrho_s c_{ps}}{\varrho c_p \varepsilon} \tag{9}$$

For simulation, the adsorber was divided into 50 parts. The time step chosen for integration was of the decimal order of 10^{-3} h. The error of calculation is given by the error of derivatives of functions and by the error of integration. The error of the Runge — Kutta integration method is of the k^5 order and may be neglected with respect to the step value used. The expression of derivatives in eqns (*7*) and (*8*) by differences and the step in the length coordinate used enable to assume that the error of calculation of derivatives does not exceed 4%. The maximum error of calculation resulting from neglecting the accumulation of adsorptive and heat in the fluid phase is less than 4% under the conditions applied in experiments. Thus, it may be supposed that the error of the results calculated according to the presented model of adiabatic equilibrium adsorption is not greater than 8%.

The constants of equilibrium eqn (*3*) were calculated on the basis of linearization and expansion into the Taylor series [40] including the experimental equilibrium data of carbon dioxide on the molecular sieve Calsit 5 found in the temperature range 273—373 K. The values of constants of eqn (*3*) thus calculated are $A = 5.85$ mol kg^{-1}, $B = 1.57 \times 10^{-3}$ K^{-1}, $K = 3.68 \times 10^{-5}$ m^3 K$^{1/2}$ mol^{-1}, and $R =$ 3889.6 K.

The parameters (*6*) have the following values: $D = 1933.1$ kg m^{-3}, $E =$ 73835.3 kg K mol^{-1}, and $F = 2237.4$ at 298 K for investigated system. The value of parameter W varied according to the flow density used.

Experimental

The apparatus used for investigation of carbon dioxide adsorption from a stream of nitrogen on molecular sieve Calsit 5 in a fixed bed arrangement under adiabatic conditions is schematically presented in Fig. 1. The stream of nitrogen deprived of oxygen by adsorption on the Ronge copper and of moisture by adsorption on the molecular sieve 5A is mixed with a stream of dry carbon dioxide. The mixture enters the adsorber packed with the molecular sieve Calsit 5 after thermostatting. The outlet stream from the adsorber is divided into two parts. In the first part equipped with a catharometer the concentration of carbon dioxide is measured while the second is vented through a needle valve into atmosphere. The reference stream of nitrogen passing the catharometer is purified in the same manner as the feed. The system of valves *21—25* enables to introduce the gas into the adsorber from above, from below or even to bypass it. The particular directions are used for investigating adsorption, desorption or for adjusting the required flow and concentrations of carbon dioxide. As obvious from Fig. 1, a part of the apparatus is placed in a temperature-controlled box for

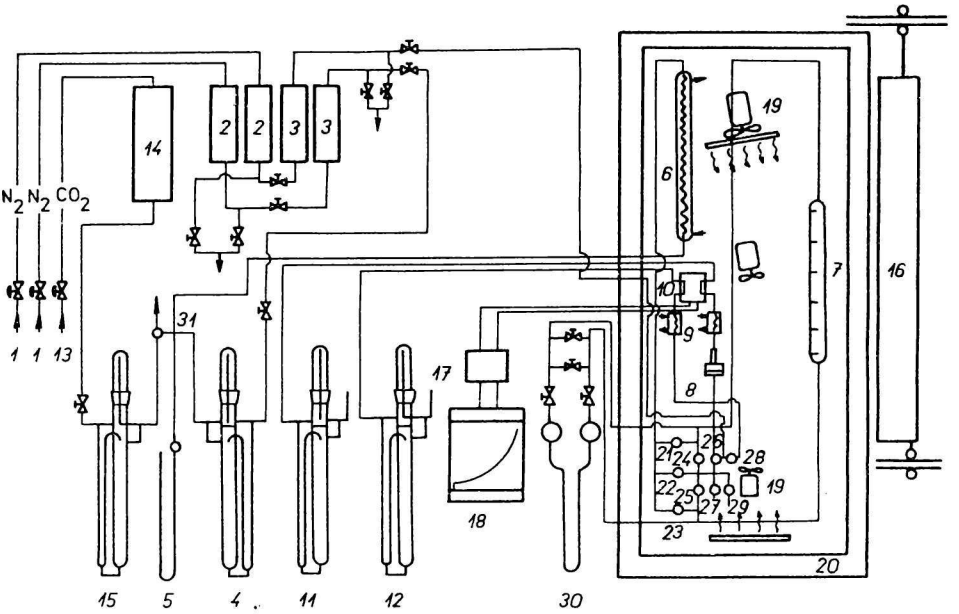

Fig. 1. Scheme of the apparatus. *1.* Inlet of nitrogen; *2.* adsorber packed with the Ronge copper; *3.* adsorber packed with the molecular sieve Calsit 5; *4.* flow-meter for nitrogen; *5.* U-manometer; *6.* heat exchanger; *7.* adsorber; *8.* membrane automatic controller of flow; *9.* heat exchanger; *10.* catharometer; *11.* flow-meter for the processed stream; *12.* flow-meter for the reference stream; *13.* inlet of carbon dioxide; *14.* adsorber packed with the molecular sieve 5A; *15.* flow-meter for carbon dioxide; *16.* furnace for activation of molecular sieve; *17.* high-ohmic Wheatstone bridge; *18.* electronic compensation recorder; *19.* ventilator with heating spiral; *20.* isolated temperature--controlled box; *21.* valve inlet of the mixture during adsorption; *22.* valve, inlet of the mixture during adjustment of conditions; *23.* valve, inlet of nitrogen during desorption; *24.* valve, outlet of the mixture during desorption; *25.* valve, outlet of the mixture during adsorption; *26.* valve, inlet of the measured stream into catharometer; *27.* valve, outlet of the mixture into atmosphere; *28.* valve, inlet of the reference stream into catharometer; *29.* valve, inlet of the mixture into chromatograph; *30.* U-manometer for measuring pressure-loss in adsorber; *31.* mixer of the streams of nitrogen and carbon dioxide.

maintaining the required conditions more precisely. Adsorber 7 having six hollows for thermocouples is enveloped during measurement with a polyurethane foam layer of about 5 cm width. During activation of molecular sieve, the polyurethane coat is taken off and an electric furnace is put on the adsorber. The concentration of carbon dioxide at the exit from the adsorber and the temperatures at the inlet, outlet and in six hollows of the adsorber are recorded with electronic compensation recorders. The positions of hollows in the bed of adsorbent and basic dimensions of the glass adsorber used are given in Fig. 2.

The following materials were used in experiments. Molecular sieve Calsit 5: balls of 2—3 mm diameter, density of balls 1148 kg m^{-3}, bulk density 752 kg m^{-3}, weighed amount of the activated molecular sieve in bed 498 g. Nitrogen of electric bulb quality deprived of

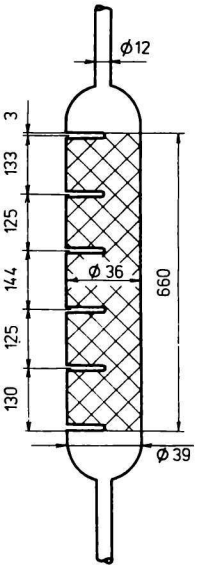

Fig. 2. Adsorber with the bed of molecular sieve.

oxygen rests by chemical sorption on the Ronge copper and of moisture by adsorption on the molecular sieve Calsit 5. Carbon dioxide from gas pressure vessels (content of carbon dioxide 99.9 mole %) was dried by adsorption of moisture on the molecular sieve 5A.

Before adsorption, the molecular sieve was activated in a stream of dry nitrogen for 2 h at 480—500°C. The adsorption was terminated after equalizing concentrations of the adsorbate in the outlet and inlet streams and after balancing the temperatures along the hight of the bed. Afterwards, the desorption with a stream of pure nitrogen followed. During desorption, the flow of nitrogen was oriented through the bed of adsorbent in the direction opposite to that applied in adsorption.

The temperature of the stream entering the adsorber and the initial temperature of the bed of adsorbent were adjusted equal. The carbon dioxide adsorption and desorption was

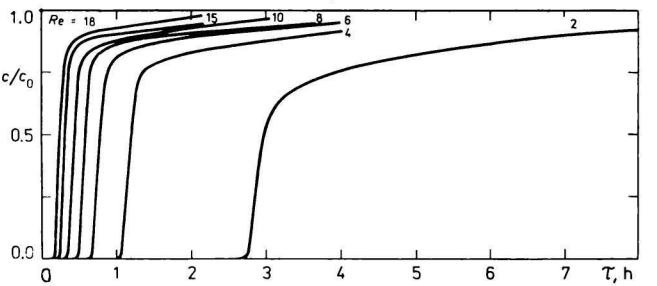

Fig. 3. Experimental break-through curves of carbon dioxide from the bed of the molecular sieve Calsit 5. Height of the adsorbent bed 66 cm, temperature of the feed and adsorbent before adsorption 298 K.

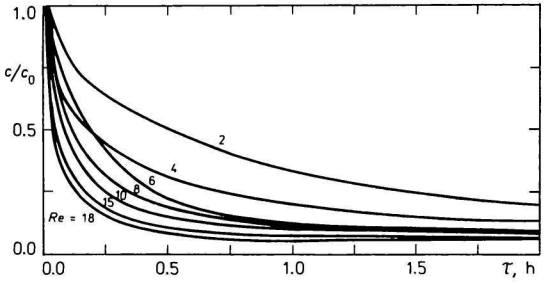

Fig. 4. Experimental relationships between the concentration of carbon dioxide and time in the outlet from the adsorber during desorption with a stream of nitrogen. Height of the bed 66 cm, temperature of entering nitrogen and temperature of the bed before desorption 298 K.

investigated for variant flow rates of nitrogen at temperatures 298 and 323 K. The mean concentration of carbon dioxide in feed during adsorption measurements was 8.55 mol m^{-3}. The concentrations of carbon dioxide at the exit from the adsorber in the course of adsorption from the stream of nitrogen and during its desorption with nitrogen, for different flow rates of nitrogen and for $T_p = T_0 = 298$ K are given in Figs. 3 and 4. The relationships ascertained for $T_p = T_0 = 323$ K exhibit the same character as those shown in Figs. 3 and 4 and are not therefore presented.

Discussion

For confrontation of the experimental and calculated break-through curves, the curves corresponding to values of the *Re* numbers 2, 8, and 18 have been selected from the set of experimental curves represented in Figs. 3 and 4. The pertinent experimental and calculated relationships for adsorption and desorption are represented in Figs. 5 and 6, respectively. The course of measured temperatures in

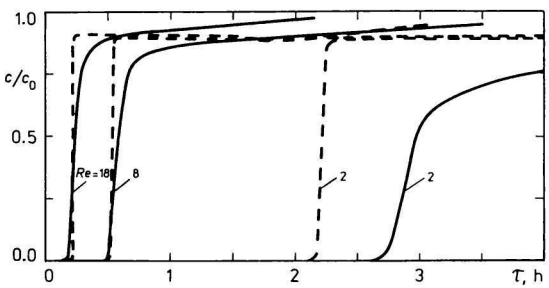

Fig. 5. Break-through curves. $z_0 = 66$ cm, $T_0 = T_p = 298$ K.
——— Experimental curves; – – – curves according to the adiabatic equilibrium model.

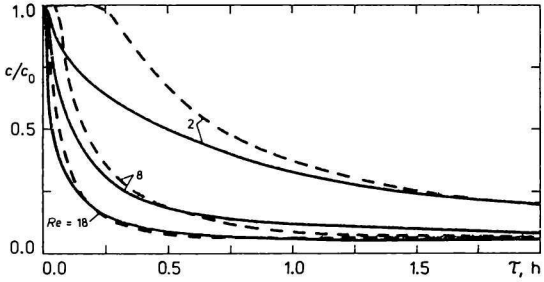

Fig. 6. Variation of carbon dioxide concentration in the outlet from the adsorber during desorption with a stream of nitrogen. $z_0 = 66$ cm, $T_0 = T_p = 298$ K.
——— Experimental; – – – according to the adiabatic equilibrium model.

the bed of adsorbent and the course of temperatures according to the adiabatic equilibrium model at $Re = 8$ are represented for adsorption and desorption in Figs. 7 and 8, respectively.

Fig. 5 shows that the break-through curves according to the adiabatic equilibrium model exhibit equal shape for each flow rate, but they are shifted along the time axis. The shift in these curves corresponds to the flow used. It is obvious from this figure that the greatest differences in the shape and positions of the experimental break-through curve and the break-through curve according to adiabatic equilibrium adsorption appear at the low nitrogen flow rate corresponding to $Re = 2$. These differences and especially the fact that the break-through curves of adiabatic equilibrium adsorption are shifted along the time axis towards the origin

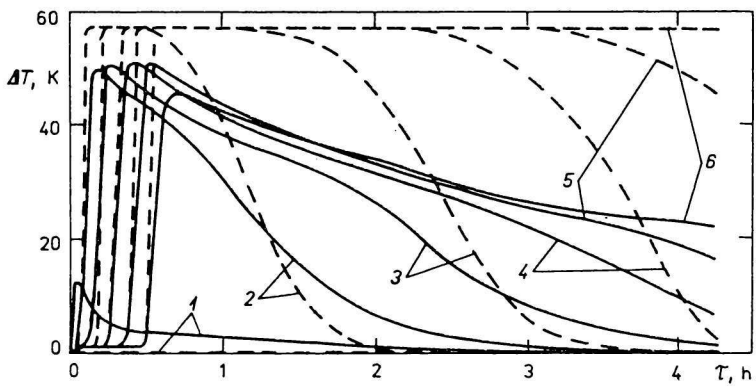

Fig. 7. Variation of temperature in the bed of adsorbent during adsorption. $Re = 8$, $z_0 = 66$ cm, $T_0 = T_p = 298$ K.
——— Experimental; – – – according to the adiabatic equilibrium model. *1, 2, 3, 4, 5, 6* correspond to individual hollows in the adsorber in the order from the inlet of feed (Fig. 2).

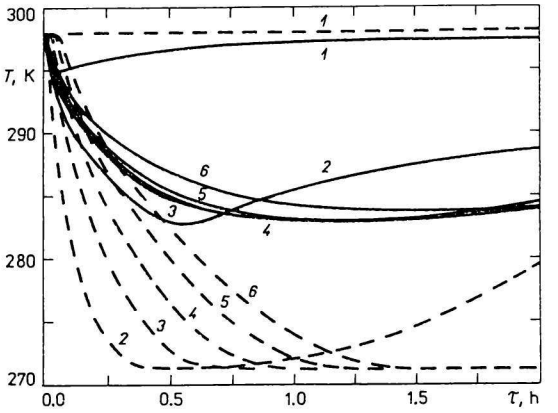

Fig. 8. Variation of temperature in the bed of adsorbent during desorption with a stream of nitrogen. $Re = 8$, $z_0 = 66$ cm, $T_0 = T_p = 298$ K.
——— Experimental; – – – according to the adiabatic equilibrium model. *1, 2, 3, 4, 5, 6* correspond to individual hollows in the adsorber in the order from the inlet of desorbent (Fig. 2).

with respect to the experimental curves cannot be explained by a relatively high and variable resistance to mass and heat transfer under particular conditions. From mutual shift of the curves at $Re = 2$ it results that less amount of carbon dioxide is caught in the bed of molecular sieve in adiabatic equilibrium adsorption than in real experiment in spite of resistance to mass and heat transfer which is effective in this case. Since the equilibrium adsorption with infinitely rapid heat transfer under adiabatic conditions represents the progress of adsorption with the maximum amount of adsorbate caught, this discrepancy indicates that the conditions of the experiment at the minimum flow rate of nitrogen cannot be considered adiabatic. Under these experimental conditions, the rate of heat transfer into surroundings cannot be neglected when compared with the rate of heat release during adsorption. The rate of heat release during adsorption increases proportionally with flow rate of the feed at the same inlet concentration of the adsorptive. Nevertheless, the rate of heat transfer into surroundings does not increase proportionally to the flow rate increase, therefore the conditions of adsorption at higher flow rates approximate to adiabatic conditions. The increase in *Re* thus means an approximation to adiabatic conditions which evidently causes the inversion in the time sequence of the experimental and model break-through curves, respectively. This change can be observed already at $Re = 8$, as obvious from Fig. 5.

Fig. 7 shows that the temperature maxima according to the model idea in the investigated spots of adsorbent bed are during adsorption by about 7°C higher than the corresponding experimental data. The differences are due to inadequacy of the model and to the method of temperature measurement in the bed of adsorbent.

The temperatures of glass wall of the hollow and not the temperatures of gas stream have been measured. It can be seen from Fig. 7 that, after reaching the maximum temperature, the bed of adsorbent rather more rapidly cools in an experiment than it should be according to the model idea. That might be caused by the heat loss into surroundings.

A comparison of the experimental changes of adsorptive concentrations in the outlet stream during desorption with the model ones represented in Fig. 6 shows that the consistence of the experimental curves and curves resulting from the equilibrium model improves with increasing Reynolds number value. Greater differences between the shape of the experimental curve and the shape of the curve according to the adiabatic equilibrium model in the region of laminar flow ($Re < 10$) may be explained, for the conditions of the experiment, on the basis of neglecting the resistance to mass and heat transfer in the model image. It is evident that the resistance to mass transfer in the solid phase does not change owing to variant flow turbulence. For this reason, the part of overall resistance, which varies with desorbent flow rate is to be localized in the fluid phase and shows diminishing influence on the mass transfer rate.

The course of desorption along the bed may be also observed by means of the temperature course shown in Figs. 8 and 9. It can be seen (Fig. 9) that the bed of adsorbent in almost the whole uniformly cools during desorption of carbon dioxide with a stream of nitrogen. This evidences a relatively uniform desorption rate in the whole bed. According to the adiabatic equilibrium model (dashed lines in Fig. 9), the desorption initially takes place in the zone of mass transfer the length of which

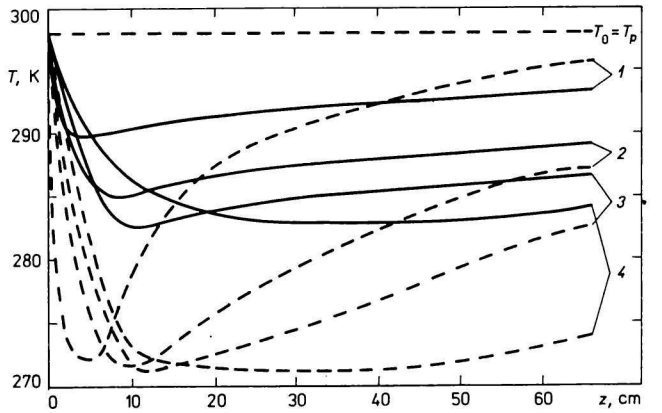

Fig. 9. Temperature profiles in the bed of adsorbent during desorption.
——— According to experimental temperatures; – – – according to temperatures calculated from the equilibrium model.
Time of desorption: *1.* 0.1 h; *2.* 0.3 h; *3.* 0.5 h; *4.* 1 h.

is smaller than the length of the bed of adsorbent and even in time greater than 0.1 h almost the whole desorption zone is in the adsorber. In the course of time, the zone of mass transfer gets washed along the bed hight and after about 1 h the desorption uniformly takes place in the whole bed. The washing of the zone of mass transfer with time resulting from calculations according to the adiabatic equilibrium model has relation solely to the influence of the equilibrium line on the course of desorption. The influence of the shape of equilibrium line on the concentration of adsorptive in the outlet from the adsorber at adiabatic equilibrium adsorption is more complicated than the influence of the shape of isotherm at isothermal adsorption or desorption described by *De Vault* [41] and subsequently by other authors [42—44]. However, it may be stated that the shape of equilibrium line derived for adiabatic equilibrium adsorption and desorption from the so-called favourable shape of isotherm has the same effect on the break-through curve resulting in its expansion as has the favourable shape of isotherm in isothermal sorption. The greater the velocity of desorbent, the greater the influence of equilibrium line on the course of desorption in its initial stage. That is documented by the trend of dashed lines in Fig. 6. The differences between the experimental course of desorption and that according to the adiabatic equilibrium model which manifest themselves considerably in the initial stage of desorption are related to the resistance to mass and heat transfer under particular conditions.

Conclusion

If necessary kinetic data in transient and turbulent flow regime are lacking, the adiabatic equilibrium model may be used for calculating the break point time in the stage of adsorption, desorption time and consumption of indifferent gas in the stage of desorption in the investigated system. Though the calculated values are the maximum or minimum values, they provide, on the basis of correct interpretation, valuable information about processing of adiabatic adsorber in cyclic operation.

Symbols

a	concentration of adsorbate	mol kg^{-1}
a^*	equilibrium concentration of adsorbate	mol kg^{-1}
a_p	initial concentration of adsorbate	mol kg^{-1}
A	constant in equilibrium relation (*3*)	mol kg^{-1}
A_1	cross-section of the adsorber	m^2
B	constant in equilibrium relation (*3*)	K^{-1}
c	concentration of adsorptive	mol m^{-3}

c^*	equilibrium concentration of adsorptive	mol m^{-3}
c_0	concentration of adsorptive in the feed	mol m^{-3}
c_p	mean heat capacity of the fluid phase	J kg^{-1} K^{-1}
c_{pb}	mean heat capacity of the solid phase	J kg^{-1} K^{-1}
c_{ps}	mean heat capacity of glass	J kg^{-1} K^{-1}
c_{poc}	equilibrium concentration of adsorptive with respect to a_p at temperature T_p	mol m^{-3}
d	equivalent diameter of the particles of adsorbent	m
D	parameter defined by relation (*6*)	kg m^{-3}
E	parameter defined by relation (*6*)	kg K mol^{-1}
F	parameter defined by relation (*6*)	—
h	length of axial step in the numerical solution of equations	m
ΔH	enthalpy change in adsorption	J mol^{-1}
K	constant in equilibrium relation (*3*)	m^3 K$^{1/2}$ mol^{-1}
R	constant in equilibrium relation (*3*)	K
S_1	area of cross-section of the wall of adsorber	m^2
T	temperature of the fluid phase	K
T_b	temperature of adsorbent	K
T_0	temperature of the feed	K
w	superficial gas velocity	m s^{-1}
W	interstitial gas velocity	m s^{-1}
z	axial coordinate	m
ε	voidage fraction	—
μ	viscosity of the fluid phase	Pa s
ϱ	density of the fluid phase	kg m^{-3}
ϱ_b	bulk density of the bed of adsorbent	kg m^{-3}
ϱ_s	density of glass	kg m^{-3}
τ	time	s
$Re = \dfrac{dw\varrho}{\mu}$	modified Reynolds criterion	—

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