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SIMULATION OF PURIFICATION OF COMPOUNDING GASES MIXTURES IN MICROBUBBLING EQUIPMENT WITH A CONSIDERATION OF CHEMISORPTION OF CARBON DIOXIDE

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Key words: microbubbling equipment, tubular ceramic membranes, highly concentrated methane, unconventional energy sources, mathematical simulation, mass transfer, liquid, gas, microbubbles.

Abstract. The presented paper describes processes of absorption and chemisorption during microbubbling in mobile liquid phase are studied. Mass transfer coefficients are identified, specific interphase surfaces and interphase flows in microbubbling device during absorption of carbon dioxide from its mixtures with methane and water suspension of CaO are identified. The adequacy of model is proved by experiments of the authors and comparisons with reference experimental data. It is demonstrated, that value specific interphase surface during microbubbling is 8-30 times higher, than in the normal bubbling, which leads to a significant reduction in working volume of a device with the same efficiency.

All presented conclusions are based on the provisions of film model of substance's transfer. However, penetration model can also be used for a description of membrane microbubbling method. Therefore, a comparison of relationships obtained by film and penetrations model is of big interest. Existing theories regarding chemisorption, even though they cannot be considered complete, provide sufficiently reliable results for a case of fast nonreversible reactions of first and pseudo-first order. At the same time, in fact, all models of mass transfer are demonstrating similar values for mass transfer coefficient for chemisorption. On the basis of the obtained data it will possible to calculate values of mass transfer coefficient in liquid phase during physical absorption k_x . In addition the study physical absorption of carbon dioxide may allow a verification of a satisfaction of the condition. As a result of the study optimal process parameters are defined, key factors affecting mass transfer characteristics of membrane microbubbling method are also established. Efficiency of membrane microbubbling equipment from the point of view of interphase mass transfer is assessed.

Introduction

Use of well-known methods of gas separation [1-4] in that case is ineffective and requires a development of new methods of purification. An alternative way of solving that problem is a combination of membrane and absorptive processes.

Recently, scientific community of developed countries of the West [5-7] produced a number of publications, reporting that during a dispersion of a gas through porous membranes microbubbles are formed [8-11] with dimensions of 0.5-150 μm . As a result of such small sizes, microbubbles have a number of unique properties, such as increased contact surface of interacting phases, they can be widely applied in chemical [12,13], food and pharmaceutical [14] industries, as well as in biotechnology, medicine and unconventional energy production.

Mathematical simulation of dispersion mechanics of gas components in the developed technology of separation and a definition of a relationship of main mass transfer parameters, i.e. specific interphase surface, interphase flow of absorbed substances (CO_2 , H_2S , NH_3 etc.), mass transfer coefficients, and speed of fluid and concentration of active part of absorbent in microbubbling processes are not established so far, which makes it a topical problem.

Theoretical analysis and results studies

Analysis of features of mass transfer processes in microbubbling equipment allows to conclude, that in a case of a design of an equipment by a type of shell-and-tube module, average time of presence of microbubbles in apparatus is quite small. In that case, considering that the flow's structure in an equipment is close to perfect displacement model (PDM), average time of presence can be calculated as follows:

$t_{pr} = \frac{l}{2\omega}$. Considering standard length of ceramic membranes of 0.8 m and fluids' speeds of 1-3 m/s, time of presence is 0.1-0.4 s, the time limit might be not enough for effective physical absorption. However, in a case of chemisorption, time of gas-liquid reaction is of 0.01 s magnitude [15]. Thus, from a point of view of effectiveness of mass transfer processes in membrane module, the most prospective are chemisorption processes in a case of comparatively fast chemical reaction. Moreover, membrane microbubbling equipment can be used not only as absorbers, but also as chemical reactors. Thus, from a point of view of study of interphase mass transfer in microbubbling contactor, it is necessary to discuss main existing chemisorption theories.

Two directions in a development of chemisorption theory can be marked out, they are based on two models of mass transfer. The first direction is based on film theory of Uiten and is developed in works of Hatta[16] and Van Krevelen[17]. The second direction is based on a penetration theory, it discusses non-stationary process of absorption during continuous renovation of interphase surface. That direction is mainly developed in works of Danckwerts[18]. It's worth mentioning, that all existing models cannot be considered complete and possess a number of disadvantages and number of studies, which would allow to select one of the directions, is insufficient. At the same time, in some practical cases, results, obtained by means of different models, are virtually the same [19].

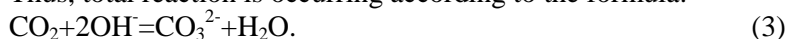
So far there is no data on studies of interphase mass transfer during membrane dispersion of gas. That data would allow to compare effectiveness of mass transfer equipment. Hence, the presented study is dedicated to experimental study of mass transfer in membrane microbubbling contactor on an example of CO₂ absorption by CaO suspension. CaO content in the suspension is 10-20% (mass).

Study of interphase mass transfer in the presented study was carried out on an example of chemisorption of carbon dioxide by alkali solutions of various concentrations, in order to do that the method, proposed by Danckwerts and Sharma was used [20].

The process comprises two consecutive reactions:



Thus, total reaction is occurring according to the formula:



In a case of sufficient excess of alkali, the second reaction is occurring almost momentarily, that's the process is limited by the first reaction, which is speed is defined by means of the expression:

$$N_R = k_2 [\text{CO}_2][\text{OH}^-] \quad (4)$$

Thus, in a case of excess of OH⁻ ions, the reaction can be considered as a reaction of pseudo-first order [20]. According to the main mass transfer equation, molar flow of CO₂ from phase to phase can be defined as follows:

$$M = k_y (C_{0,G} - C_{i,G})F = k'_x (C_{i,L} - C_{0,L})F, \quad (5)$$

where k_y , k'_x – coefficients of mass transfer in gas and liquid phases respectively, $C_{0,G}$ – CO₂ concentration in a volume of gas phase, $C_{i,G}$ – CO₂ concentration in gas phase in interphase border, $C_{0,L}$ – CO₂ concentration in a volume of liquid phase, $C_{i,L}$ – CO₂ concentration in liquid phase in interphase border, F – interphase surface area. If chemical reaction is fast enough, it can be accepted that $C_{0,L} = 0$, considering that concentrations in interphase surface are related by equilibrium – $C_{i,G} = mC_{i,L}$, the following can be obtained from expression (5):

$$M = \frac{G_{0,G}F}{\left(\frac{1}{k_y} + \frac{m}{k'_x}\right)} \quad (6)$$

In the following, let's discuss the solution, leading to expression for mass transfer coefficient for chemisorption. The discussed reaction is very fast nonreversible chemical reaction in liquid phase of the following type [16]:



At the same time, it's considered that reaction zone starts directly in a vicinity interphase surface and, depending on speed of reaction and concentration of components, is extended on a different depth. The process is considered stationary. The solution can be applied to reaction of first (of $A \rightarrow v_d D$ type) and pseudo-first type.

In a case of nonreversible reaction of first order n by substance A , equation of convective diffusion will take the following form [21]:

$$D_A \frac{d^2 C_A}{dy^2} = k_p C_A^n \quad (8)$$

where reaction speed constant k_p is defined according to expression $k_p = k_{n+m} C_B^m$, in which k_{n+m} — reaction speed constant of order of $n+m$. Boundary conditions:

$$\begin{aligned} y = 0 & \quad C_A = C_{Ai} \\ y = \infty & \quad C_A = 0 \end{aligned}$$

By means of specifying $\frac{dC_A}{dy} = q$, expression (8) can be presented in the following form:

$$q \frac{dq}{dC_A} - \frac{k_p}{D_A} C_A^n = 0. \quad (9)$$

Integration of that expression considering boundary conditions leads to the expression:

$$\left(\frac{dC_A}{dy}\right)^2 = \frac{2}{n+1} \frac{k_p}{D_A} C_A^{n+1}. \quad (10)$$

Because $\frac{dC_A}{dy}$ is negative:

$$\left(\frac{dC_A}{dy}\right) = -\sqrt{\frac{2}{n+1}} \sqrt{\frac{k_p}{D_A}} C_A^{\frac{n+1}{2}}. \quad (11)$$

Thus, interphase chemisorption speed can be defined as follows:

$$N'_A = -D_A \left(\frac{dC_A}{dy}\right)_{y=0} = \sqrt{\frac{2k_p D_A}{n+1}} C_{Ai}^{\frac{n+1}{2}}. \quad (12)$$

In a case chemical reaction of first (or pseudo-first) order is not fast enough, $C_{A0} \rightarrow 0$, and from expression (12) it follows, that chemisorption speed can be calculated as follows:

$$N'_A = \sqrt{k_p C_{Ai} D_A}, \quad (13)$$

Then, considering, that in a case of fast enough reaction $C_{A0} = 0$, the following expressing for coefficient of mass transfer for absorption, which is accompanied by reaction of pseudo-first order (also, considering that the reaction has the first order by substance B):

$$k'_x = \frac{N'_A}{(C_{Ai} - C_{A0})} = \sqrt{k_p D_A} = \sqrt{k_2 C_B D_A} \quad (14)$$

In all relationships presented above it is presumed, that substance B is in excess and $C_B = \text{const}$. In the study [17] an investigation into influence of concentration of C_B on ration of values $\frac{k'_x}{k_x}$ and $\frac{\sqrt{k_2 C_B D_A}}{k_x}$ (as C_B an average value is used). The studies were carried out using carbon dioxide – sodium hydroxide, results are presented in figure 1.

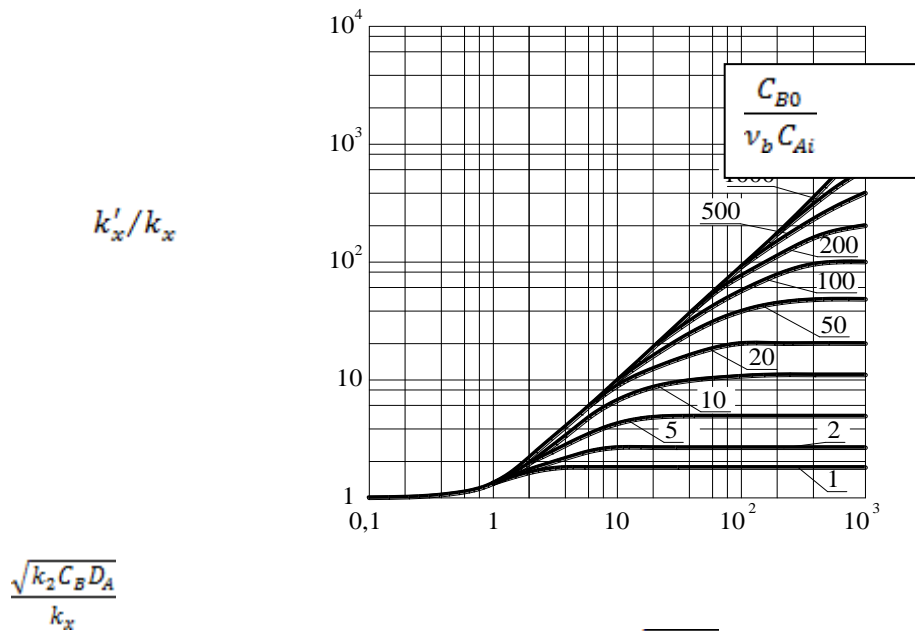


Figure 1 – Relationship of value $\frac{k'_x}{k_x}$ from $\frac{\sqrt{k_2 C_B D_A}}{k_x}$ with different values $\frac{C_{B0}}{v_b C_{Ai}}$.

Analysis of the diagram shows that even in a case of slow reaction, independently of concentration B , value of $\frac{k'_x}{k_x} \approx 1$ and process are close to physical absorption. Increase of reaction speed leads to that value of $\frac{k'_x}{k_x}$ becomes approximately equal to $\frac{\sqrt{k_2 C_B D_A}}{k_x}$, which demonstrates a possibility of application of expression (14) for calculation of mass transfer coefficient. In that area the process is limited by speed of chemical reaction. With a further increase of speed value of $\frac{k'_x}{k_x}$ becomes constant, which is close to $\frac{C_{B0}}{v_b C_{Ai}}$, at the that, the process is limited by mass transfer B to reaction zone.

According to aforementioned points, expression (5) can be used with acceptable accuracy in the following range:

$$5 < \frac{\sqrt{k_2 C_B D_A}}{k_x} < \left(\frac{C_B}{v_b C_{Ai}} \right) \tag{15}$$

where k_x – mass transfer coefficient for physical absorption. Substitution of expression (14) in expression (6), considering that $F = aV_{work}$, produces the following:

$$M = \frac{C_{0,G} V_{work}}{\left(\frac{1}{k_y} + \frac{m}{\sqrt{k_2 C_B D_{CO_2}}} \right)} \tag{16}$$

Expression (16) can be transformed in the following form:

$$\frac{C_{0,G} V_{work}}{M} = \frac{1}{k_y a} + \frac{m}{a \sqrt{k_2 C_B D_{CO_2}}} \tag{17}$$

In coordinates $y = \frac{C_{0,G} V_{work}}{M}$, $x = \frac{m}{\sqrt{k_2 C_B D_{CO_2}}}$ expression (17) describes line with angle of inclination tangent $\frac{1}{a}$ and crossing with coordinate axis in point $\frac{1}{k_y a}$. Thus, by means of experimental study of relationship of y from x specific interphase surface area in equipment can be defined, as well as mass transfer coefficient in gas phase, which allows to define effectiveness of interphase mass transfer in membrane microbubbling equipment.

Following conclusion can be made on a basis of analysis of mass transfer with chemical reaction: existing theories regarding chemisorption, event though they cannot be considered complete, provide

sufficiently reliable results for a case of fast nonreversible reactions of first and pseudo-first order. At the same time, in fact, all models of mass transfer are demonstrating similar values for mass transfer coefficient for chemisorption. However, because an implementation of expression (4) requires conforming with condition (15), it is necessary to carry out additional experimental studies in order to define value of k_x . For conditions, specified by inequality (15), coefficient of mass transfer may be calculated according to expression $k'_x = \sqrt{k_p D_A} = \sqrt{k_2 C_B D_A}$, which is conforming both with film and penetration theories. The expression shows, that in a case of fast chemical reaction, mass transfer coefficient doesn't depend on hydrodynamic conditions in flow core, but it is defined by speed of reaction and speed of diffusion of absorbed component.

Methodology

For the study of interphase mass transfer during chemisorption of carbon dioxide by CaOH solution in membrane contactor, the method proposed by Sharma and Danckwerts was selected [20].

Molar flow of absorbed CO₂ in that study was experimentally defined by means of a change of concentration of alkali in solution, the following expression was used for calculations:

$$M = \frac{(C_B^H - C_B^K) V_L}{\nu_b}, \quad (18)$$

where C_B^H and C_B^K – initial and final alkali concentration, V_L – consumption of solution, ν_b – stoichiometric coefficient in overall equation, which is equal to 2.

Reaction speed constant of second order k_2 can be calculated using the following expression [22]:

$$\lg \left(\frac{k_2}{k_{2,\infty}} \right) = 0.221I - 0.016I^2. \quad (19)$$

Value of speed constant in infinitely diluted solution is defined as follows:

$$\lg(k_{2,\infty}) = 11.895 - \frac{2382}{T} \quad (20)$$

Expression (20) can be used in temperature range 290-314 K. Ionic force is calculated according to the following expression [23]:

$$I = \frac{1}{2} \sum_{j=1}^n C_j Z_j^2 \quad (21)$$

where C_j – concentration of Ca⁺, HCO₃⁻, OH⁻, CO₃²⁻ ions, Z_j – valence of those ions. At the same time, it is noted [24] that, because of HCO₃⁻ and CO₃²⁻ ions' concentration are quite small, their values can be neglected. Thus, value of I becomes equal to concentration of alkali in a solution.

Diffusion coefficient CO₂ in alkali solution is defined according to the following equation [25]:

$$D_{CO_2} = D_{CO_2}^{aq} \left(\frac{\mu_{aq}}{\mu_s} \right)^{0.85}, \quad (22)$$

where μ_{aq} – viscosity of water, μ_s – viscosity of alkali solution (was defined according to the data [25]), CO₂ diffusion coefficient in clear water is defined as follows [26]:

$$D_{CO_2}^{aq} = 2.35 \cdot 10^{-6} \exp \left(-\frac{2119}{T} \right). \quad (23)$$

Distribution coefficient m was defined on a basis of experimental data on solubility of carbon dioxide in solutions of CaOH, presented in [24]. In experiments on membrane with average pore diameter of 0.5 μm, in a case of alkali concentrations of 0.030-0.070 kmole/m³, value of $m=10.3$ was used, in experiments on membrane with average pore diameter of 2.6 μm, in a case of alkali concentrations of 0.014-0.030 kmole/m³, value of $m=9.9$ was used,

Analysis of adequacy of the obtained data

Analysis of main mechanisms of mass transfer with chemical reaction allows to conclude, that during chemisorption mass transfer coefficient in liquid phase both in plate type and membrane type membrane microbubbling device can be calculated using expression (14), thus, values of k'_x will be comparable.

Mass transfer coefficients in plate-type device, calculated on a condition of equal k'_x and value $k_y = 1 \cdot 10^{-3}$ m/s are $4.6 \cdot 10^{-5} - 7.5 \cdot 10^{-5}$ m/s and $2.9 \cdot 10^{-5} - 5.1 \cdot 10^{-5}$ m/s respectively. Therefore, mass transfer coefficients in plate-type device in the discussed conditions will be 1.1-2.9 times higher than in membrane contactor.

From expression (17) with known values of a and k_y , interphase flow can be described as follows:

$$M = \left(\frac{1}{k_y} + \frac{m}{\alpha \sqrt{k_2 c_B D_{CO_2}}} \right)^{-1} C_{0,G} V_{work} \quad (24)$$

Transformation of expression (24) gives the following:

$$M = \left(\frac{1}{k_y} + \frac{m}{\alpha \sqrt{k_2 c_B D_{CO_2}}} \right)^{-1} \alpha C_{0,G} V_{work} = k_y \alpha C_{0,G} V_{work} \quad (25)$$

it can be concluded, that because value of k_y membrane device is in average 1.5 times smaller and specific interphase surface is in 8-30 times higher, than in a case of the same working volumes amount of absorbed substances will increase in 5-20 times. It allows to conclude that in order to reach the desired level of absorption, membrane microbubbling device must have 5-20 smaller working volume than plate type device. At that, the following recommendations can be given. Mass transfer coefficient obtained during experiments with both membranes are quite close, but in the same time values of specific interphase surface for a membrane with $d_0=0.5 \mu\text{m}$ are in 2-2.5 times bigger. Thus, it can be presumed, that implementation of microfiltering membranes with pores less than $1 \mu\text{m}$ will give more significant effect in decreasing sizes of a devices and, consequently, will reduce capital spendings. At that, the most optimal range of speeds of liquid, both from point of view of mass transfer coefficients and specific interphase surface is a range 1.5-2.5 m/s. At the same time, in a case of use of membrane with $0.5 \mu\text{m}$ pores necessary gas pressure is three times higher, as compared to a membrane with $2.6 \mu\text{m}$ pores. It can lead to significant energy expenses for gas blow off, especially with their expenses. Thus, for design of membrane device for a specific process technical and economic analysis should be carried out in order to find a compromise decision between a selection of type of membranes with smaller pores to reduce sizes of device from one side, and a selection of membranes with bigger pore to reduce necessary pressure from another side.

Calculations of mass-transfer coefficient, which were carried out on a basis of experimental data, shows, that for conditions, studied in the presented paper, mass-transfer coefficient in membrane contactor (calculated for gas phase) has values of $1.6 \cdot 10^{-5} - 4.5 \cdot 10^{-5}$ m/s for a membrane with $0.5 \mu\text{m}$ pores and $2.3 \cdot 10^{-5} - 4.5 \cdot 10^{-5}$ for a membrane with $2.6 \mu\text{m}$ pores.

Conclusion

Processes of absorption and chemisorption during microbubbling in mobile liquid phase are studied. Mass transfer coefficients are identified, specific interphase surfaces and interphase flows in microbubbling device during absorption of carbon dioxide from its mixtures with methane and water suspension of CaO are identified. The adequacy of model is proved by experiments of the authors and comparisons with reference experimental data. It is demonstrated, that value specific interphase surface during microbubbling is 8-30 times higher, than in the normal bubbling, which leads to a significant reduction in working volume of a device with the same efficiency. A comparison with membrane hollow fiber contactors shows that using of ceramic membranes allows to substantially increase value specific interphase flow in the microbubbling device and it is comparable or higher than in a case of hollow fiber contactor.

As it was noted, all presented conclusions are based on the provisions of film model of substance's transfer. However, penetration model also can be used for a description of membrane microbubbling method. Therefore, a comparison of relationships obtained by film and penetrations model is of big interest. Existing theories regarding chemisorption, even though they can't be considered complete, provide sufficiently reliable results for a case of fast nonreversible reactions of first and pseudo-first order. At the same time, in fact, all models of mass transfer are demonstrating similar values for mass transfer coefficient for chemisorption. For the verification of the conditions on pseudo-first order reaction (condition 15) it is planned to conduct experiments on physical absorption of pure CO_2 by water, on the same membranes and in the same speed range of liquid and gas consumptions as in experiments on chemisorption. On the basis of the obtained data it will possible to calculate values of mass transfer coefficient in liquid phase during physical absorption k_x . In addition the study physical absorption of carbon dioxide may allow a verification of a satisfaction of the condition (15).

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Моделирование очистки многокомпонентных газов в микробарботажном аппарате при хемосорбции диоксида углерода

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Ключевые слова: микробарботажный аппарат, керамические мембраны, высококонцентрированный метан, нетрадиционная энергетика, математические моделирование, массообмен, жидкость, газ, микробарботаж

Аннотация. В данной статье описаны процессы абсорбции и хемосорбции при микробарботаже в подвижную жидкую фазу. Определены коэффициенты массоотдачи, удельные поверхности контакта фаз и межфазные потоки в микробарботажном аппарате при поглощении диоксида углерода из его смесей с метаном и водной суспензией СаО. Адекватность модели доказана собственными экспериментами и сравнением с литературными экспериментальными данными. Показано, что величина удельной поверхности контакта фаз при микробарботаже в 8-30 раз больше, чем при обычном барботаже, что приводит к существенному уменьшению рабочего объема аппарата при одинаковой

эффективности.

Все представленные выводы основаны на положениях пленочной модели переноса вещества. Однако для описания массообмена при мембранном микробарботаже может подходить и пенетрационная модель. Поэтому представляет большой интерес сравнение данных зависимостей с зависимостями, полученными на основе пенетрационной модели. Существующие теории хемосорбции, хотя и не являются достаточно полными, но дают достаточно надежные результаты для случая быстрых необратимых реакций первого и псевдо-первого порядка. При этом фактически все модели переноса предсказывают сходные значения для коэффициента массоотдачи при хемосорбции. Однако в силу недостатков, свойственных существующим моделям переноса, для точного предсказания k'_x необходимо иметь экспериментальные данные по k_x . В результате исследования определены оптимальные параметры процесса, выявлены основные факторы, влияющие на массообменные характеристики мембранного микробарботажа. Дана оценка эффективности мембранных микробарботажных аппаратов с точки зрения межфазного массообмена.

Микробарботажды аппаратта көміртегі диоксидінің хемосорбциялану кезінде көпкомпонетті газдарды тазалауды модельдеу

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Түйін сөздер: микробарботажды аппарат, түтікшелі керамикалық мембрана, жоғарыконцентрациялы метан, дәстүрлі емес энергетика, математикалық моделдеу, массаалмасу, сұйық, газ, микробарботажа.

Аннотация. Бұл мақалада микробарботажа кезінде қозғалмалы фазадағы абсорбция және хемосорбция процестерінің жүруі сипатталған. Микробарботажды аппараттағы көмірқышқыл газының метанмен қоспаларында СаО сумен суспензиясының сіңірілуі кезінде массаалмасу коэффициенті, фазалардың меншікті жанасу беттері және фазалар аралық ағындары анықталды. Алынған модельдердің анықтылығы тәжірибелік мәліметтер және әдебиеттегі ақпараттармен салыстырылып дәлелденген. Микробарботажа кезінде фазалардың меншікті жанасу беттері қарапайым барботажа кезіндегіден 8-30 есе көп екендігі көрсетілген, ол бірдей тиімділік кезінде аппараттың жұмыс көлемінің елеулі төмендеуіне мүмкіншілік береді.

Барлық ұсынылған қорытындылар зат алмасудың қабыршақты моделіне негізделген. Бірақ, мембраналы микробарботажада массаалмасуды сипаттау үшін пентерациялық модель де дұрыс келуі мүмкін. Сондықтан, алынған мәліметтерді пентерациялық модель негізінде алынған мәліметтер арқылы да салыстыру қызығушылық тудырады. Осы күнге дейінгі белгілі хемосорбция теориялары толық болмаса да, бірінші және жалған-бірінші ретті өте жылдам қайтымсыз реакциялар үшін сенімді нәтижелер береді. Бұл жағдайда зат алмасудың барлық модельдері хемосорбция кезінде массаалмасу коэффициенттері үшін ұқсас нәтиже береді. Бірақ зат зат алмасудың барлық модельдеріне тән кемшіліктеріне байланысты k'_x дәл анықтау үшін k_x бойынша тәжірибелік мәліметтер қажет. Зерттеу барысында процестің тиімді параметрлері, мембраналы микробарботаждың көрсеткіштеріне әсер етуші негізгі факторлар анықталды. Фазалар аралық массаалмасу тұрғысынан микробарботажды аппараттардың тиімділігіне баға берілген.

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