American Journal of Engineering Research (AJER)	2016
American Journal of Engineering Res	earch (AJER)
e-ISSN: 2320-0847 p-ISS	N:2320-0936
Volume-5, Issue	e-12, pp-37-45
	www.ajer.org
Research Paper	Open Access

The Modeling of Absorption in two-Layer System "Gaseous Mixture-Liquid" Under Ultrasonic Vibrations Influence

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ABSTRACT: The paper presents theoretical studies of absorption gas mixture separation under ultrasonic vibrations influence, which provides cavitation and acoustic process intensification. The theoretical studies based on diffusion equations with non-isotropic effective diffusion coefficients determined by acoustics flows and natural diffusion. The proposed model takes into account increasing of interface "gas-liquid" area, caused by surface capillary waves provided by shock waves from cavitation bubbles. In result of the studies, it is evaluated, that ultrasonic intensification increases absorption productivity up to 2 times and more with amplitude of vibrations 1...2 µm. The obtained results can be used for development of high-efficiency absorption apparatuses that are supplemented by ultrasonic influence sources.

Keywords: Ultrasonic, absorption, gaseous mixture, diffusion, cavitation

I. INTRODUCTION

The problem of artificial separation of gas mixtures caused by the rapid development of techniques and technologies is for over 100 years [1]. Until today, in connection with the development of production, technology, transportation and medicine, demand for the separation of gas mixtures for separation of finished products, and to remove undesirable impurities, is constantly growing. One good example is the increasing demand for chemically pure hydrogen. Today, due to avalanche increase in the number of exhaust gases emitted into the atmosphere, in fact it is necessary to organize a network of hydrogen filling stations to carry out a wide spread of environmentally friendly kinds of autos. Obviously, this will require a manifold increase in the hydrogen production volumes to values which are comparable with today's volumes of produced fossil fuels based on crude oil and natural gas. In this case, it is necessary to provide a low cost of the product. It naturally follows the need to create high-performance and low-cost gas separation plants, because as the primary method of hydrogen production is steam reforming of natural gas/methane [2]. The steam reforming results a gaseous mixture of H2, CO2, CH4, H2O and CO, from which it is necessary to separate the H2 only.

Another example is the growing need for oxygen regeneration from CO2 to organize underwater expeditions and long manned space flight to study extraterrestrial objects. The life support systems for such flights and expeditions need high performance, cost-effective and compact device for the O2. In one way or another, the process will be reduced to the separation of oxygen from a mixture of carbon compounds [3]. High demand on the performance and efficiency of these systems is due to the need for the greatest possible number of crew and severe restrictions on the amount of energy produced on board the spacecraft or submarine vessels. You can lead a lot of other examples that show the need to increase productivity and reduce energy gas separation mixture. However, despite the breadth of application of the separation of gas mixtures in a variety of spheres of human activity, and at the same time high energy efficient gas separation methods [1, 3–7] have limited capacity and are now working virtually at maximum capacity.

The potential effectiveness of the cryogenic and membrane separation technology gases today can be considered as exhausted. For cryogenic technologies it is true because of a long start-up period and the need to cool the gas mixture to the boiling point of the lowest boiling point component, for the membrane technologies it is true due to the high compressed gas flow resistance of the membrane. Thus, today, it is the most promising division sorption methods [6–8]. Among sorption methods the absorption has the greatest practical interest. The absorption performance can be up to 200 000 m³/h due to the fact that the absorption of the gas mixture separation, today this performance is insufficient. Attempts to improve the performance of absorption by increasing the absorbent flow rate is a liquid leads to a disproportionate increasing the cost of the process due to

the high cost of the regeneration of the absorbent. Increasing of the flow of the gas mixture obviously leads to decrease the purity of the gases due to limited diffusion rate. Therefore, one of the possible ways to increase the absorption capacity is the use of external influence leading to increase the interface "liquid-gas" that leads to increase gas absorption rate. Known absorbers is base on creating of the larger interface area using spray absorbing liquid (atomizing absorbers), eg, patent RU 2,380,143 by Znyatdinova *et al.* [9]. However, in this case, gases that are little soluble in the absorbent is practically not absorbed due to occurrence of turbulent micropulsations. Thus, the effectiveness of atomizing absorbers for these gases is reduced to nothing.

Therefore, to solve this problem, it is encouraged by applying of acoustic (ultrasonic) cavitation, the impact of which increases the interfacial surface of the "liquid-gas" [8]. The acoustic and cavitation influence is effective for the intensification of the processes of absorption that is experimentally proven by domestic and foreign researchers. Acoustic cavitation influence has the following advantages:

- 1. The possibility of intensification of absorption, regardless of chemical composition of the separated gas and liquid absorbents due to the possibility of choose of the optimal ultrasonic influence mode and exposure conditions (frequency, intensity, location, and direction of the input of ultrasonic vibrations, radiating area) determined by the physical properties of gaseous mixture and absorbing liquid.
- 2. Installation of ultrasound sources does not require changes in the design and functionality of the absorbers.
- 3. Low power consumption. According to the results of laboratory tests, the intensification of gas absorption occurs even at low intensity exposure, if you select a certain resonance frequency of the oscillation.
- 4. The possibility of simultaneous intensification of chemisorption because cavitation contribute to breakage of chemical bonds and the formation of new ones.
- 5. Compact sources of ultrasonic vibrations and electronic generators for their power.

However, despite a number of advantages, in practice, today the acoustic cavitation effects is not been implemented due to lack of knowledge of the physical mechanisms of the process and the lack of scientific data of the best modes of ultrasonic action providing maximum efficiency of the process as consequence.

Due to time consuming and expensive experimental studies it is necessary to develop a mathematical model of the effect of ultrasonic influence on the interfacial surface area of the "liquid-gas" and the kinetics of absorption, allowing evaluating optimal modes of ultrasonic intensification of this process.

II. PROBLEM STATEMENT

According to the method of acoustic cavitation intensification of absorption (Fig. 1), into the thin liquid film (absorbent) powerful acoustic or ultrasonic (US) vibrations (frequency of 20...250 kHz) are introduced by solid-state radiator [8, 10].



Fig. 1. Scheme of the acoustic cavitation intensification of absorption.

Ultrasonic vibrations in fluid create acoustic and cavitation field [11, 12]. It is the cavitation bubbles, which periodically expand and collapse into non-spherical nuclei forming microscopic shockwaves with pressure pulses up to 1000 atmospheres [11, 13]. Shock waves lead to the formation of stable capillary waves on the interface "liquid-gas" [14].Capillary waves allow to increase the area of the interface. Also ultrasonic vibrations and cavitation lead to acoustic flows which cause to increase effective diffusion coefficient. All of this increases the speed of absorption. The model allowing evaluating optimum modes providing maximum of interface area "liquid-gas" was proposed early and described in paper [14]. However, the model doesn't allow to determine concentrations of target gaseous component in liquid and gas layers and to determine optimum modes productivity. Determination of absorption productivity requires to analyze diffusion coefficients and the interface "liquid-gas" under ultrasonic influence. To determine the optimum modes providing maximum absorption productivity, the flow of two-layer system is considered in following calculation area (Fig. 2).



Fig.2. The area of calculation of flow in two-layer system "liquid-gas".

In the area gaseous mixture and liquid flow have velocities u_g and u respectively which are directed along axis x.Near the input boundary (x=0) concentrations of target component in gaseous mixture layer $C_g(x,y)$ (mol/m³) and in liquid layer C(x,y) (mol/m³) essentially depend on x. Wherein at x=0 $C_g(0,y)=C_{gin}$ and C(0,y)=0. On the infinity at $x \to \infty$ the two-layer system achieves the equilibrium state (without diffusion flows). Here is $\lim_{x\to\infty} C_g(x, y) = \lim_{x\to\infty} C(x, y) = C_{\infty}$ and the maximum of gas absorption is achieved. However, real absorption setups have finite sizes. It is means, that the absorption efficiency is restricted by channel length L. In this case, concentrations of target component at output of absorption setup equal to $C_g(L,y)$ in gaseous layer and C(L,y) in liquid layer.

Thus, total absorption productivity (number of target component absorbed by liquid during unit of time) is determined as

$$P = S_{ch} \int uC(L, y) \partial y ; \qquad (1)$$

where S_{ch} is square of channel cross-section which is perpendicular to axis x.

Obviously, at the ultrasonic influence the productivity depends on influence modes which are frequency f (Hz) and amplitude A (m) of solid surface (y=0) oscillations along axis y (see Fig. 1, 2)

$$P(A, f) = S_{ch} \int_{0}^{h} uC_{A, f}(L, y) \partial y \cdot$$

The final goal of ongoing studies is determining of ultrasonic influence modes providing maximum increase of absorption productivity:

$$\beta = \frac{P(A, f)}{P_{NOUS}} = \frac{\int_{0}^{C} C_{A,f}(L, y) \partial y}{\int_{0}^{h} C_{NOUS}(L, y) \partial y} \to \max ; \qquad (2)$$

where P_{NoUS} is absorption productivity without ultrasonic influence, mol/s; P(A, f) is productivity at ultrasonic influence with amplitude A and frequency f, mol/s; $C_{A,f}(x, y)$ is concentration of target component in liquid layer at ultrasonic influence with amplitude A and frequency f; mol/m³; $C_{NoUS}(x, y)$ is concentration of target component in liquid layer without ultrasonic influence, mol/m³.

To determine the modes providing maximum increase of absorption productivity the model of diffusion in "gaseous mixture-liquid" two-layer system in area shown in Fig. 2 was proposed. Further proposed model is described.

III. MODEL OF ABSORPTION IN TWO-LAYER SYSTEM "GASEOUS MIXTURE-LIQUID" UNDER ULTRASONIC INFLUENCE

The model is based on diffusion equation with non-isotropic coefficients taking into account influence of acoustic vortices:

$$u \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2} + 2D_{xy} \frac{\partial^2 C}{\partial x^2};$$
(3)

h

h

$$u_{g} \frac{\partial C_{g}}{\partial x} = D_{g} \left(\frac{\partial^{2} C_{g}}{\partial x^{2}} + \frac{\partial^{2} C_{g}}{\partial y^{2}} \right);$$
(4)

$$D_{y} \frac{\partial C}{\partial y}\Big|_{y=h} = D_{g} \frac{\partial C_{g}}{\partial y}\Big|_{y=h} = K_{s} (C - C_{g})\Big|_{y=h};$$
(5)

$$\frac{\partial C}{\partial y} = 0; (6)$$

$$\frac{\partial C_s}{\partial y}\bigg|_{y=h+h_s} = 0$$
 (7)

$$C(0, y) = 0;$$
 (8)

$$C_{g}(0, y) = C_{gin};$$
 (9)

$$\lim_{x \to \infty} C = \lim_{x \to \infty} C_g = C_{\infty} = \frac{u_g h_g C_{gin}}{u h + u_g h_g};$$
(10)

where D_x , D_y , D_{xy} are non-isotropic diffusion coefficients taking into account ultrasonic influence, m²/s; K_s is the coefficient of concentration interaction between liquid and gaseous mixture, m/s; α is the coefficient of relative increasing of interphase surface determined from paper by Golykh R.N. [14].

The boundary condition (5) including the coefficient of relative increasing of interphase surface α is substantiated by following scheme (Fig. 3).



Fig. 3. Scheme of diffusion of target gaseous component from gaseous mixture to liquid at capillary waves formed by ultrasonic influence.

At considering of small area ∂s_0 , which is perpendicular to axis y, the input target component flow

 $-\partial S_0 D_y \frac{\partial C}{\partial y}$ projected to ∂S_0 equals to output flow $-\partial S_0 D_y \frac{\partial C_y}{\partial y}$ at small distances from surface which is

more than capillary wave length but much less than ∂S_0 sizes.

Due to mass conservation law, input and output flows equal to flow caused by concentration interaction between phases on undulating surface ∂S_{us} . The flow equals to $K_s (C_s - C) \partial S_{us}$. However $\partial S_{us} = \alpha \partial S_0$. It causes the boundary condition (5).

For determination modes providing maximum increasing of absorption productivity (2) the analysis of the model (3-10) consists two stages:

- 1) determination of the diffusion coefficient of target component into liquid;
- evaluation of concentration of target component in liquid on fixed distance (x=L) from begin of the system (x=0).

Further the stages are described in details.

Determination of the diffusion coefficient into liquid.

The diffusion coefficient in liquid at ultrasonic influence can be determined by averaging of diffusion equation over small volume of acoustic micro flow:

$$\langle (\mathbf{u} + \mathbf{w}, \nabla) C \rangle = D \Delta \langle C \rangle;$$

where *D* is real diffusion coefficient of target component in liquid, m^2/s ; **u** is velocity of stationary liquid flow, m/s; **w** is oscillating velocity caused by ultrasonic influence.

The oscillating velocity includes velocity of first order \mathbf{w}_1 (primary ultrasonic wave) and second order \mathbf{w}_2 (acoustic micro flows caused by non-linear effects and vibrations weakening):

$$\mathbf{w} = \mathbf{w}_1 + \mathbf{w}_2$$

The \mathbf{w}_{1} , \mathbf{w}_{2} and \mathbf{w} can be found from non-linear acoustic equations given in Rosenberg's book [12]. After volume averaging of diffusion equation in liquid we derives following equation

$$(\mathbf{u}, \nabla) \langle C \rangle = D \Delta \langle C \rangle - \langle (\mathbf{w}, \nabla) C \rangle.$$
(11)

At this stage, our task is derivation from equation (11) to the non-isotropic diffusion equation in general form (3). It is achieved by decomposition of term $\langle (\mathbf{w}, \nabla)_C \rangle$ in Taylor's series with taking into account assumption

$$\left\langle \sum_{i=1}^{n} w_{i} \right\rangle (\mathbf{r}_{0}) = 0 \text{ due to conservation of liquid mass.}$$

$$\left\langle (\mathbf{w}, \nabla) C \right\rangle (\mathbf{r}_{0}) = \left\langle (\mathbf{w}, \nabla_{\mathbf{r}}) (C(\mathbf{r}_{0}) + (\mathbf{r}, \nabla_{\mathbf{r}}) C + ...) \right\rangle (\mathbf{r}_{0}) = \left\langle \sum_{i,j=1}^{n} w_{i} \frac{\partial}{\partial x_{i}} \left[r_{j} \frac{\partial}{\partial x_{j}} C(\mathbf{r}_{0}) \right] \right\rangle (\mathbf{r}_{0})$$

$$= \left\langle \sum_{i,j=1}^{n} w_{i} \left[\delta_{ij} \frac{\partial}{\partial x_{j}} C(\mathbf{r}_{0}) + r_{j} \frac{\partial^{2}}{\partial x_{j} \partial x_{i}} C(\mathbf{r}_{0}) \right] \right\rangle (\mathbf{r}_{0}) = \left\langle \sum_{i=1}^{n} w_{i} \right\rangle (\mathbf{r}_{0}) \frac{\partial}{\partial x_{i}} C(\mathbf{r}_{0}) + \left\langle \sum_{i,j=1}^{n} w_{i} r_{j} \right\rangle (\mathbf{r}_{0}) \frac{\partial^{2}}{\partial x_{j} \partial x_{i}} C(\mathbf{r}_{0}) + \left\langle \sum_{i,j=1}^{n} w_{i} r_{j} \right\rangle (\mathbf{r}_{0}) \frac{\partial^{2}}{\partial x_{j} \partial x_{i}} C(\mathbf{r}_{0})$$

The decomposition allows determining effective diffusion coefficients included in equation (3):

$$D_{x} = D - \langle w_{x}x \rangle (\mathbf{r}_{0}); D_{y} = D - \langle w_{y}y \rangle (\mathbf{r}_{0}); D_{xy} = -\frac{\langle w_{x}y \rangle (\mathbf{r}_{0}) + \langle w_{y}x \rangle (\mathbf{r}_{0})}{2}$$

At the cavitation in liquid layer and big sizes of acoustic vortices coefficient D_{xy} can be assumed to be zero, because Eccart's flows prevail over rotationary acoustic flows due to high coefficient of acoustic waves weakening. Moreover, at small layers thicknesses h and h_g and at uniform acoustic influence all diffusion coefficients can be assumed to be constants.

Thus, the diffusion equation in liquid (3) can be modified as follows:

$$u \frac{\partial C}{\partial x} = D_x \frac{\partial^2 C}{\partial x^2} + D_y \frac{\partial^2 C}{\partial y^2}$$

The equation is used to following determine of concentrations of target component.

Evaluation of concentration of target component.

To determine concentration of target component, author has proposed method based on decomposition target

component concentrations into Taylor's series relatively from $\left(\frac{y}{h}\right)^2$ for C and $\left(\frac{h+h_g-y}{h_g}\right)^2$ for C_g .

$$C = \sum_{k=0}^{\infty} C_k \left(\frac{y}{h}\right)^{2k};$$

$$C_g = \sum_{k=0}^{\infty} C_{gk} \left(\frac{h+h_g-y}{h_g}\right)^2$$

where C_k and C_{gk} is functions depending on x only, mol/m³.

;

The concentration representations automatically satisfy to boundary conditions (6) and (7). Because $\left\{ \left(\frac{y}{h}\right)^{2k} \right\}$ and $\left\{ \left(\frac{h+h_g-y}{h_g}\right)^{2k} \right\}$ are families of linearly independent functions, the diffusion equations are

decomposed into

$$u \frac{\partial C_k}{\partial x} = D_x \frac{\partial^2 C_k}{\partial x^2} + D_y (2k+2)(2k+1) \frac{C_{k+1}}{h^2};$$
(12)

$$u_{g} \frac{\partial C_{gk}}{\partial x} = D_{g} \left(\frac{\partial^{2} C_{gk}}{\partial x^{2}} + (2k+2)(2k+1) \frac{C_{g(k+1)}}{h_{g}^{2}} \right)$$
(13)

Equations (12) and (13) give expressions for C_{k+1} and $C_{g(k+1)}$ depending on C_0 and C_{g0} in operator view:

$$C_{k} = \frac{1}{(2k)!} L^{k} [C_{0}];$$
(14)

$$C_{gk} = \frac{1}{(2k)!} L_{g}^{k} [C_{g0}];$$
(15)

where *L* and *L_g* are linear operators $L = \frac{h^2}{2D_y} \left(u \frac{\partial}{\partial x} - D_x \frac{\partial^2}{\partial x^2} \right); L_g = \frac{h_g^2}{2D_g} \left(u_g \frac{\partial}{\partial x} - D_g \frac{\partial^2}{\partial x^2} \right).$

The representations (14, 15) allow to reduce the concentrations C and C_g determining problem to determining C_0 and C_{g0} depending on x only.

Using the operator view, the boundary condition on interphase surface can be as follows:

$$\sum_{k=1}^{\infty} \frac{D_{y}}{(2k-1)!K_{s}h} L^{k} [C_{0}] = -\sum_{k=1}^{\infty} \frac{D_{g}}{(2k-1)!K_{s}h_{g}} L_{g}^{k} [C_{g0}] = \sum_{k=0}^{\infty} \frac{1}{(2k)!} (L_{g}^{k} [C_{g0}] - L^{k} [C_{0}])$$

The conditions on input and infinity of considered channel is represented through C_0 and C_{g0} as follows:

$$\sum_{k=0}^{\infty} \frac{1}{(2k)!} L_{g}^{k} \left[C_{g0} \left[\left(\frac{h+h_{g}-y}{h_{g}} \right)^{2k} \right]_{x=0} = C_{gin};$$

$$\sum_{k=0}^{\infty} \frac{1}{(2k)!} L^{k} \left[C_{0} \right] \left(\frac{y}{h} \right)^{2k} \bigg|_{x=0} = 0$$

almost everywhere.

For the calculation of concentrations from the infinite series we take first N members. This gives approximate linear differential equations for functions C_0 and C_{g0} depending on x only.

$$\sum_{k=1}^{N} \frac{D_{y}}{(2k-1)!K_{s}h} L^{k}[C_{0}] = -\sum_{k=1}^{N} \frac{D_{g}}{(2k-1)!K_{s}h_{g}} L_{g}^{k}[C_{g0}] = \sum_{k=0}^{N-1} \frac{1}{(2k)!} \left(L_{g}^{k}[C_{g0}] - L^{k}[C_{0}] \right); \quad (16)$$
with boundary conditions (when $N > 0$)

with boundary conditions (when N > 0)

$$\frac{1}{(2N-2)!} L^{(N-1)} [C_0] \Big|_{x=0} = \frac{C_{gin}}{\frac{(2N-2)D_y}{\alpha K_c h} - \frac{1}{2N-1} - 1 + \frac{2N-2}{2N-1} \frac{h_g D_y}{hD}}$$
(17)

$$C_{0}(0) = -\frac{1}{(2N-1)!} L^{N-1} [C_{0}](0); \qquad (18)$$

$$C_{g0}(0) = C_{gin} - \frac{1}{(2N-1)!} L_{g}^{N-1} [C_{g0}](0);$$
⁽¹⁹⁾

$$\lim_{x \to \infty} C_0 = \lim_{x \to \infty} C_{g0} = \frac{u_g h_g C_{gin}}{u h + u_g h_g};$$
(20)

$$\lim_{x \to \infty} \frac{\partial^k C}{\partial x^k} = \lim_{x \to \infty} \frac{\partial^k C_g}{\partial x^k} = 0 \text{ at } k = 1 \dots 2N - 2.$$
(21)

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The equations allows to determine relative increasing of absorption productivity (see expression (2)) with ultrasonic $(D_x = D - \langle w_x x \rangle (\mathbf{r}_0); D_y = D - \langle w_y y \rangle (\mathbf{r}_0); \alpha > 1)$ and without ultrasonic influence $(D_x = D_y = D; \alpha = 1)$.

Obtained dependences of relative absorption productivity increasing under ultrasonic influence are presented in the next section.

IV. OBTAINED RESULTS

Obtained dependences of relative increase of the absorption productivity (see expression (23)) in the first approximation on the modes of ultrasonic action are shown in Fig. 4. The breakage of graphs means a loss of capillary waves stability and its breakage into drops [14].



Fig.4. Dependences of specific area of the interface on the modes of ultrasonic action: (a) on amplitude at different frequencies; (b) on frequency at maximum amplitude.

From presented dependences it is evident, that with the increase of amplitude absorption productivity grows up to more than 2 times. Starting with the frequency of 60 kHz the growth of the maximum productivity essentially becomes slower, and energy loss of the ultrasonic radiator increases quadratically. That is why; the application of frequencies of more than 60 kHz is unpractical.

Fig. 5 shows the dependences of relative increasing of absorption productivity at the change of physical properties of liquid – viscosity (a) and surface tension (b), which influence on the contact surface area and the diffusion together with the modes of ultrasonic action.



(b)

Fig.5. Dependences of increase of absorption productivity on amplitude at different properties of liquid (frequency of 60 kHz): viscosity (a) and surface tension (b).

Presented dependences on amplitude at different liquid properties (Fig. 5) can be used for the determination of the absorption productivity under ultrasonic influence caused by change of the liquid type and change of its properties. In particular it is stated, that growth of viscosity leads to the decreasing productivity or the increasing amplitudes which are necessary to achieve defined productivity. At that decrease of surface tension leads to the decreasing amplitudes, which are necessary to achieve defined productivity, and the decreasing maximum amplitude providing capillary waves stability (no breakup into drops).

V. CONCLUSION

Thus, the model of acoustic cavitation absorption intensification was developed. Analysis of the model has shown, that acoustic cavitation influence leads to increase absorption productivity up to 2 times. For maximum absorption productivity increasing, the optimum modes of ultrasonic action are evaluated. It is shown, that the most appropriate frequency of ultrasonic vibrations is 60 kHz at amplitudes no more than 1.2 μ m, at which more than 2 times increase of absorption productivity. Herewith, the liquid phase viscosity growing leads to increasing amplitudes which is necessary for achievement of defined productivity. And decreasing of surface tension leads to the decreasing amplitudes, which are necessary to defined productivity achievement, and the decreasing maximum amplitude providing capillary waves stability (no breakup into drops). Obtained new scientific results have fundamental interest for the understanding of physical mechanism of the absorption ultrasonic intensification and they can be used for development of high-efficiency absorption apparatus that is supplemented by ultrasonic influence sources.

ACKNOWLEDGEMENTS

The reported study was supported by Grant of President of the Russian Federation No.MK-4515.2016.8.

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