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EFFECTIVENESS FACTOR APPROACH FOR CHEMICAL ABSORPTION PROCESS

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Abstract

Absorption of gases into liquids, with chemical reaction, is an important unit operation useful in many fields, especially synthesis of new products and waste gas treatment. Gas treatment means the separation of pollutants: acid gases, organic sulphur compounds, and other contaminants which may be found in gaseous streams. The absorption into liquid is a process almost exclusively used for removal of contaminants. The removal refers to retention of the majority of acid gases present in high concentration down to a level such as 0.1 % in the treated gas. The gas-liquid processes are carried out in a variety of equipment's including packed towers, bubbling absorbers, spray columns, falling film contactors etc. The absorber selection and design require models describing the interaction between mass transfer and chemical reaction. For design purposes, it may be used the concept of enhancement factor (*E*), related to the positive effect of the reaction rate on the physical mass transfer.

The aim of this paper is to develop a mathematical model that allows the analysis of gas-liquid processes in the intermediate reaction regime. The effectiveness factor approach for gas liquid reactions is developed in this paper by using the concept of fractional conversion in the liquid film (X_F), which precisely indicates the kinetic regime. This new approach for gas-liquid reactions is presented in the current study. When reaction rate does not improve the mass transfer (slow and very slow reaction) the effectiveness factor concept can be used to describe this interaction.

The elaborated mathematical model allows the determination of effectiveness factor values for practical intervals of Hatta (Ha) modulus and modified Sherwood (Sh) number. The development concept is similar with model used in the modelling of the heterogeneous catalytic reactions.

The proposed mathematical model has shown that in the intermediate regime (when $0.01 < X_F < 0.99$), the effectiveness factor approach is more convenient.

Key words: chemical absorption, effectiveness factor, gas-liquid processes, modelling

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1. Introduction

Absorption of gaseous pollutants into liquid phase is widely used in environmental engineering for different contaminants removal (Biard and Couvert 2013; Roustan, 2003; Yildirim et al., 2012). Air pollution is a local, pan-European and hemispheric issue. Sources of air pollution can be natural, anthropogenic, or both. Emissions from industry, transport activities, combustion system of diesel engine, wastewater treatment plants or from agriculture, are only a few examples of sources of anthropogenic pollution (Ge et al., 2016; Su et al., 2016; Wang et al., 2016). Nitrous oxide, methane and carbon dioxide (CO₂) exhibit greenhouse effect and, among these, CO₂ is considered the main greenhouse gas with negative impact over climate change (Bucur and Harja, 2012; Harja and Szep, 2013). CO₂ is

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typically used as a reference for global warming approaching (Jongartklang et al., 2016; Qin et al., 2016; Tan et al., 2012) and is believed to be responsible for about 64% of the greenhouse effect (Ramazani et al., 2016).

Gas treating is the term used to describe the separation of acid gases (CO₂, H₂S, SO₂), organic volatile compounds (VOC), organic sulphur compounds and certain other gases (HCN, NH₃, NO_x etc.) from gaseous wastes (Cai et al., 2017; Favier et al., 2016a, 2016b; Szep and Harja, 2007). There are four main categories of gas treating processes: absorption into a physical or chemical liquid agent; adsorption onto solids (zeolites, activated carbon); cryogenic separation; chemical conversion to other compounds. When the gas compound reacts with the liquid phase, the process is called chemical absorption. Chemical absorption is currently believed to be the most suitable method in the case of post-combustion power plants (Behr et al., 2011; Harja et al., 2008). On the other hand, chemical absorption process between gases and liquids are extensively applied in chemical engineering (Zhao et al., 2017). Significant chemical processes as: wet flue gas desulphurization, carbon capture by absorption, syngas purification, manufacturing of nitric or sulphuric acid, chlorination, hydrogenation, ammonization etc. involve an absorption process for manufacturing new products or for treating waste gases (Gaspar and Fosbøl, 2015; Qin et al., 2016; Yildirim et al., 2012).

This unit operation consists in pollutant transfer from the gas phase to a liquid phase; gas dissolution in liquid phase and chemical reaction at interface or in bulk liquid phase. The absorption into liquids is the process almost exclusively used for bulk removal of acid contaminants. Bulk removal refers to removal of the majority of acid gases present in high concentration down to a level such as 0.1 % in the treated gas.

There are different models that describe the absorption accompanied by chemical reaction process, which is based mainly on three theories: the two-films, the penetration and the surface renewal (Koronaki et al., 2015; Molga and Westerterp, 2013). The two films theory was developed by Whitman (1924), the equations being established by assuming a stagnant film of liquid and one of gas on each of the two sides of the interface area (Astarita, 1967; Morsi and Basha, 2015; Noeres et al., 2003). The resistance to mass transfer is concentrated in these films. Higbie (1935) proposed the penetration theory, where is considered that the gas-liquid interface is not stagnant but liquid packages remain at the interface for a period of time named contact time, then migrate back into the solution. Higbie's model was modified by Danckwerts (1955) resulting in the surface renewal theory that assumes a contact time distribution of the liquid elements.

The chemical absorption processes are carried out in a variety of equipments ranging from a bubble absorber to a packed column, a spray tower, a Venturi scrubber, or a falling film contactor (Kening et al., 2003; Petrescu and Harja, 2006). Chemical absorption processes are intensively used; however, the modelling is still approximate. Other accurate models are necessary in order to improve the absorber design, to upgrade the technical level and to perform a better economic optimization. A balance between modelling accuracy and computational requirements has to be found for simulations and applications of absorber (Gaspar and Fosbøl, 2015). Solving the numerical model is still computationally difficult.

The absorber selection and design require models characterizing the interaction between mass transfer and chemical reaction. Two approaches may be used to describe interaction between mass transfer and chemical reaction.

The first approach, expressing the effect of the reaction on improvement of the physical mass transfer led to the older concept of the enhancement factor (E) (Petrescu et al., 1998; Ramazani et al., 2016). The enhancement factor diminishes the complexity of a mass transfer model. The Hatta number is a parameter which compares reaction rate and diffusion rate in a liquid film. An increased Hatta number indicates a higher reaction rate compared to diffusion. In this context, Hatta number (Ha) and the instantaneous enhancement factor have been used in the modelling. Gaspar and Fosbøl presented a fast and simple enhancement factor model and revealed that the model overlaps the numerical solution of the film model (Gaspar and Fosbøl, 2015).

The second approach, expressing decrease of the reaction rate by the mass transfer resulted in the effectiveness factor concept, which is widely used in the modelling of the heterogeneous catalytic reactions. This is a relatively new approach for gas-liquid reactions.

The design of the absorber requires knowledge of the kinetic regime and the kinetic equation (Petrescu and Harja, 2006). The effectiveness factor approach for gas liquid reactions is developed in this paper by using the concept of fractional conversion in the liquid film (X_F) , which precisely indicates the kinetic regime. The fractional conversion in the liquid film (X_F) is a function of the Ha number. Within this work, the values of the effectiveness factor for practical intervals of Ha modulus and modified Sherwood number are determined, using a derived mathematical model. For the regime of intermediate reaction rates the effectiveness factor approach is more convenient. It is further solved for practical intervals of Hatta modulus and modified Sherwood number

2. Mathematical modelling

2.1. The enhancement factor

Let A be the component of the gas phase reacting with a non-volatile compound B in the liquid phase. The reaction that takes place is confined to the liquid phase are presented in Eq. (1):

$$A + v_B B = \text{products} \tag{1}$$

Considering the Whitman model (Ivaniciuc, 1999; Noeres et al., 2003), the concentration profile in the liquid phase is presented in Fig. 1.



Fig. 1. Concentration profile in the liquid film

Since the two films theory involves a steady state, the mass balance of *A* in a slice of thickness dx and unit cross section in the liquid film may be written as Eq. (2):

$$D_A \frac{d^2 C_A}{dx^2} + k C_A^n C_B^m = 0 \tag{2}$$

For the component *B* the relation is similar (Eq. 3):

$$D_B \frac{d^2 C_B}{dx^2} + k C_A^n C_B^m = 0$$
 (3)

with the boundary conditions given by Eq. (4):

$$x = 0; C_A = C_A^i \quad \frac{dC_B}{dx} = 0 (4)$$
$$x = \delta_L; C_A = C_A^v; C_B = C_B^v$$

By integrating Eqs. (2-4), the concentration profiles of *A* and *B* in the liquid film are obtained. In general, Eqs. (2) and (3) cannot be integrated analytically, excepting the special cases of rate equations. For a pseudo-first-order reaction (n = 1, m= 0 or C_B =const.) the (Eq. 2) becomes Eq. (2'):

$$D_A \frac{d^2 C_A}{dx^2} + k_1 C_A = 0$$
 (2')

with the boundary conditions (Eq. 4'):

$$\begin{array}{l} x = 0; \qquad C_A = C^i{}_A \qquad (4') \\ x = \delta_L; \quad C_A = C^v{}_A \end{array}$$

At the interface (x = 0), Eq. (5) was obtained:

$$\left[\frac{dC_A(x)}{dx}\right]_{x=0} = \sqrt{\frac{k_1}{D_A}} \left[\frac{C_A^v - C_A^i \cosh(\delta_L) \sqrt{\frac{k_1}{D_A}}}{\sinh\left(\delta_L \sqrt{\frac{k_1}{D_A}}\right)}\right]$$
(5)

The modulus
$$\delta_L \sqrt{\frac{k_1}{D_A}}$$
 is Hatta number (*Ha*):

$$\delta_L \sqrt{\frac{k_1}{D_A}} = \frac{D_A}{k_L^0} \sqrt{\frac{k_1}{D_A}} = \frac{\sqrt{k_1 D_A}}{k_L^0} = Ha \tag{6}$$

The flux of *A* transferred through the gas film boundary in the presence of the chemical reaction in the liquid phase is:

$$N_{A}^{'} = D_{A} \left(\frac{d_{A}(x)}{d} \right)_{x=X}^{C} = k_{L}^{0} \frac{H}{t \quad dH} \left(C_{A}^{i} - \frac{a \quad C_{A}^{v}}{n \quad c \quad a \quad H \text{ o}} \right)$$
(7)

In the absence of the reaction, the mass flux physically transferred is given by Eq. (8):

$$N_A = k_L^0 \left(C_A^i - C_A^\nu \right) \tag{8}$$

The enhancement factor (*E*) may be defined as the ratio N_A^N/N_A (Eq. 9):

$$E = \frac{N_{A}}{N_{A}} = \frac{Ha}{tanh(Ha)} \left(\frac{1 - \frac{\gamma}{cosh(Ha)}}{1 - \gamma} \right)$$
(9)
$$\gamma = \frac{C_{A_{l}}^{\nu}}{C_{A_{l}}^{i}}$$
where: (10)

If $C^{\nu}_A \ll C^i_A$, $\gamma \to 0$ then (Eq. 9) becomes:

$$E = \frac{Ha}{tanh(Ha)} \tag{11}$$

There is no agreement in the literature concerning the values of Ha at which the reaction is completed in the film (Ha > 3.0 or Ha > 5.0) or takes place mainly in the bulk liquid.

In order to determine these limits, one can define the fraction of *A* consumed in the liquid film (X_F) (Eq. 12):

$$X_{F} = \frac{\left(N_{A}^{'}\right)_{x=0} - \left(N_{A}^{'}\right)_{x=\delta_{L}}}{\left(N_{A}^{'}\right)_{x=0}} = 1 - \frac{\left(N_{A}^{'}\right)_{x=\delta_{L}}}{\left(N_{A}^{'}\right)_{x=0}}$$
(12)

The mass flux of A which was not consumed in the film is Eq. (13):

$$\left(N_{A}^{'}\right)_{x=\delta_{L}} = k_{L}^{0} \frac{Ha}{\tanh Ha} \left(\frac{C_{A}^{i}}{\cosh Ha} - C_{A}^{v}\right)$$
(13)

Combining Eqs. (7), (12), and (13) leads to Eq. (14):

$$X_F = 1 - \frac{1 - \gamma \cosh Ha}{\cosh Ha - \gamma} \tag{14}$$

Eqs. (9) and (13) include the ratio γ which cannot be directly determined but correlated with

other ready measurable amounts. A material balance of A at the film - bulk boundary gives Eq. (15):

$$-S_{\nu}D_{A}\left(\frac{dC_{A}}{dx}\right)_{x=\delta_{L}} = \left(\varepsilon_{L} - S_{\nu}\delta_{L}\right)k_{1}C_{A}^{\nu}$$
(15)

from which Eq.(16) is found by integration:

$$\gamma = \frac{\frac{1}{\cosh(Ha)}}{1 + Ha(\beta - 1)\tanh(Ha)}$$
(16)

where:
$$\beta = \frac{\varepsilon_L k_L^0}{S_v D_{A_i}}$$
 (17)

This β may be considered a modified Sherwood number. The system Eqs. (14-17) gives X_F as a function of Ha and β .

2.2. Effectiveness factor

The effectiveness factor or *liquid utilization factor* (η_L) may be defined as the ratio (Eq. 18):

$$\eta_L = \frac{(r_A)_{ef}}{(r_A)} \tag{18}$$

where the effective rate of gas-liquid reaction is (Eq. 19):

$$(r_{A})_{ef} = -\frac{dn_{A}}{Vd\tau} = S_{\nu}k_{L}^{0}E(C_{A}^{i} - C_{A}^{\nu})$$
(19)

and the reaction rate in the liquid phase is (Eq. 20):

$$(r_A) = -\frac{d_A n}{V \tau d} \varepsilon_L k_1 C_A^i$$
(20)

By combining Eqs. (18-20) results Eq. (21):

$$\eta_L = \frac{S_v k_L^0 E(1-\gamma)}{\varepsilon_L k_1} \tag{21}$$

Since *E* and β are correlated by Eqs. (9) and (16) respectively, then Eq. (21) could be written as Eq. (22):

$$\eta_L = \frac{1}{Ha\beta} \left[\frac{\tanh Ha + Ha(\beta - 1)}{1 + Ha(\beta - 1)\tanh Ha} \right]$$
(22)

3. Results and discussion

3.1. Experimental and numerical validation of equations

Validation of Eqs. (15) and (22), must be based on experimental values of the kinetic parameters and transfer properties including: k_1 , k_L , \mathcal{E}_L , S_{ν} . The determination of these coefficients by the chemical method, using a model reaction as sulphite oxidation, has been presented in a previous paper (Ivaniciuc and Siminiceanu, 2000). For systems generally found in gas treatment technologies, their values are ranged within the following intervals (Ivaniciuc, 1999):

$$5 \times 10^{-5} < k_L < 5 \times 10^{-4};$$
 10 $< S_v < 10000$
 $0.20 < \varepsilon_L < 0.99;$ 10⁻⁹ $< D_A < 5 \times 10^{-9}$

From here one may assess the limits of the Hatta and modified Sherwood numbers:

$$1.0 < \beta < 10000.0$$

0.1 < Ha < 10.0

The practical values of β are frequently ranged between 10 and 5000, whereas those of *Ha* may exceed 10 and even 100 for very fast reactions.

By solving the Eqs. (14-17), in Mathcad 7, the values of X_F have been obtained. These values are of a great importance for the determining the kinetic regime of a given process and the rational design of the industrial gas-liquid contactor. According to the data presented in Fig. 2, three kinetic regimes can be distinguished, depending on the values of Ha and β : - When Ha < 0.3, for practical values of β (5< β

<5000), more than 99% of *A* is consumed in the bulk liquid ($X_F < 0.01$). This is the *slow reaction regime*. For this regime, from the Eq. (9) results:

$$Ha < 0.3 \ tanh \ Ha = Ha$$
 $E = 1.0$ (23)

A contactor with a large liquid holdup which, according to the Eq. (20), that accelerates the reaction, will be the best choice. This could be a bubble column or a stirred reactor.

Values of $1 \cdot X_F = f(Ha, \beta)$ are presented in Fig. 2, considering: Ha=0.1-10, $\beta = 1.0-10,000$, $\varepsilon_L=0.2-0.99$, $D_A = (1-5) \ 10^{-9}$, $S_v = 10-1000$, $k^0_L = (0.5-5) \ 10^{-4}$. - When Ha > 5.0, the fractional conversion of *A* in the liquid film exceeds 99% for all values of β greater than 5.0. This is *the fast reaction regime*. From Eq. (11) results Eq. (24) (Hagiu and Ivaniciuc, 1997):

$$Ha > 5.0 tanh Ha = 1 \qquad E = Ha \tag{24}$$

The rate of the gas-liquid reaction becomes Eq. (25):

$$\hat{N}_{A}S_{v} = S_{v}\sqrt{k_{1}D_{A}}C_{A}^{i}$$
 [kmol m⁻³ s⁻¹] (25)

A contactor with as great as possible interfacial areas (S_v) will be the best choice. This could be a packed column, a plate column, a Venturi scrubber or a falling film contactor.

- When 0.3 < Ha < 5.0 the values of X_F are ranged between 0.1 and 0.99 and depend on both Ha and β . This is the *intermediate reaction regime*, most frequently found in practice.

3.2. The effectiveness factor approach

For these intermediate reaction rates the use of the enhancement factor is not always in agreement with the standard approach of diffusional limitations in reactor design and may be somewhat confusing. Moreover, there are cases where there simply is no purely physical mass transfer process to refer to, as CO_2 absorption in aqueous alkali solutions. The *effectiveness factor approach* is more convenient in these cases. For pseudo-first-order reactions this approach leads to a simple kinetic equation (Eq. 26):

$$(r_A)_{ef} = \eta_L \varepsilon_L k_1 C_A^i \tag{26}$$

The values of the effectiveness factor (η_L) resulted from Eq. (22) are presented in Fig. 3, for $\beta = 1 - 1000$ și Ha = 0.3 - 10.



Fig. 2. Fraction of A consumed in the liquid film as a function of Ha and β



Fig. 3. Effectiveness factor for gas-liquid absorption with pseudo-first-order chemical reaction as a function of β and Ha



Fig. 4. Concentration profiles for the catalytic process (a) and the gas liquid absorption process (b)

3.3. Analogy between absorption and catalysis processes

In order to study the analogy between the Hatta and Thiele modulus, the model of a grain catalyst plate by length equal to *L* and the two films model for chemical absorption with film thickness equal to δ_L have been considered (Fig. 4). In the case of liquid-gas processes we have demonstrated that the liquid utilization factor can be calculated with the (Eq. 22), which for $\beta = 1$ has the form given by Eq. (27):

$$\eta_L = \frac{thHa}{Ha} \tag{27}$$

For catalytic processes, the mass balance of A in a catalyst plate by length equal to L may be written as:

$$D_{ef} \frac{d^2 C_A}{dx^2} + k_1 C_A = 0$$
 (28)

Thiele modulus has the form of Eq. (29):

$$\Phi = L_{\sqrt{\frac{k_1}{D_A}}}$$
(29)

For catalytic process η_{cat} has the same form with liquid utilization factor for $\beta_{cat} = \beta = 1$ (Eq. 30):

Table 1. Absorption	versus	catalytic	process
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Catalytic	Absorption
$D_{ef} \frac{d_{C_A}^2}{dx^2} + k_1 C_A = 0$	$D_A \frac{d_{C_A}^2}{dx^2} + k_1 C_A = 0$
$\Phi = L_{\sqrt{\frac{k_1}{D_A}}}$	$Ha = \delta_{l} \sqrt{\frac{k_{1}}{D_{A}}}$
$\beta_{cat} = \frac{\varepsilon_p}{S_v L}$	$\beta = \frac{\varepsilon_L k_L^0}{S_v D_{A_1}} = \frac{\varepsilon_L}{S_v \delta_L}$
It can observe similarity: δ_L equival	ent with L ε_L equivalent with ε_p
$\eta_{cat} = \frac{th\Phi}{\Phi}$ for $\beta_{cat} = 1$	$\eta_L = \frac{thHa}{Ha}$ for $\beta = 1$

$$\eta_{cat} = \frac{th\Phi}{\Phi} \tag{30}$$

It must be demonstrated that $\beta_{cat} = 1$.

$$\beta_{cat} = \frac{\varepsilon_p}{S_v L} \tag{31}$$

But:

$$\varepsilon_p = \frac{\sum V_{Si}}{V} = \frac{\sum S_i L}{V} = \frac{L \sum S_i}{V} = LS_v$$
(32)

By replacing in Eq. (31), it was obtained $\beta_{cat} = 1$, which demonstrates the perfect similarity.

Perfect similarity between models is presented in Table 1. From Table 1 it can be seen that even if there are major phenomenological differences between these processes, the mathematical model in simplified form is similar.

Analogy between models leads off a variety of perspectives. This conducted, in collaboration between different scientists, to provide a valuable source of intuitions which have led to a profounder theoretical understanding of processes. Analogy involves the comparison of two different domains: catalysis and absorption. Indeed, the models can be expanding in unknown process.

4. Conclusions

Reactions between gases and liquids are extensively applied in gas purification and synthesis of new products. The rational design of the absorbers requires knowledge of the kinetic regime and the kinetic equation.

The classical mathematical model, based on Ha and the enhancement factor, E, can be used in the case of fast and very fast reactions. The kinetic regime can be determined by means of the fractional conversion in the liquid film (X_F) as a function of the Ha number.

For the intermediate reaction rates (when $0.01 < X_F < 0.99$), it is recommended to use the effectiveness factor approach. Based on the experimentally data, for various practical applications, the variation ranges of the effectiveness factor were calculated. The values of the effectiveness factor for practical intervals of *Ha* modulus and modified Sherwood number are determined, using a derived mathematical model.

The graphical representations presented in the paper allow the first approximation of a new process, the data read on the diagram can be used as a basis for modeling new reactors/absorbers.

This study demonstrated that two different processes can be study on the base of similar model and knowledge can be share between processes. Finally, these results provide additional valuable information about the possibility of using the effectiveness factor approach for the intermediate reaction regime, which can lead to viable application in environmental engineering for mathematical modelling and design of removal processes of the contaminants from gaseous streams.

Notation

- A, component of the gas phase, dissolved in the liquid;
- *B*, component of the liquid phase reacting with *A*;
- C_A , C_B , molar concentration of A, and B (mol L⁻¹);
- C^{i}_{A} , C^{v}_{A} , concentration of A at the interface, and in the liquid bulk, respectively (mol L⁻¹);
- D_A , D_B , molecular diffusivities of A and B in the liquid film (m² s⁻¹);
- *E*, enhancement factor;
- Ha, Hatta number;
- k_L , mass transfer coefficient of *A* from interface to liquid bulk (m s⁻¹);
- *k*₁, rate coefficient for the pseudo-first-order reaction between *A* and *B* (s⁻¹);
- n, m, reaction orders;
- n_A , moles number of component A;
- N_A , molar rate of physical absorption per unit gas-liquid interface area (mol m⁻² s⁻¹);
- N_{A}^{*} , molar rate of chemical absorption per unit gas-liquid interface area (mol m⁻² s⁻¹);
- r_A , reaction rate of component A per unit of liquid volume (mol m⁻³ s⁻¹);
- $(r_A)_{ef}$, effective reaction rate (mol m⁻³ s⁻¹);
- S_{ν} , gas-liquid interfacial area per unit of liquid volume (m² m⁻³);
- *V*, volume of liquid phase (m³);

 X_F , fractional conversion of A in the liquid film;

- *x*, coordinate perpendicular to gas-liquid interface (m);
- β , modified Sherwood number for liquid film;
- γ , the ratio C^{ν}_{A}/C^{i}_{A} ;
- δ_L , liquid film thickness for mass transfer (m);
- EL, liquid holdup;
- η_L , effectiveness factor/ liquid utilization factor;
- v_B , stoichiometric coefficient of component *B*; τ , time (s).

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