THREE-PHASE, FLUID DISPERSE SYSTEMS FOR GAS/LIQUID COUNTERCURRENT CONTACTING

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Abstract

There are numerous unit operations in the field of chemical engineering (such as absorption, desorption, scrubbing, distillation, etc.) which can only be carried out if gas (vapour)/liquid contacting is applied. The main dimension of the operating equipment depends on the diffusion rate. The application of three-phase, fluid disperse systems yields much better results than the use of traditional fixed bed columns.

Fundamentals

The separation of gas mixtures by application of an absorbent liquid, the stripping of a gas component dissolved in liquid with the help of another gas, purification of gases polluted by solid substances by means of a liquid, and continuous contacting of the vapour and liquid phase at distillation processes, respectively, are often occurring tasks in chemical industry.

It is a well-known fact that solubility of certain gases in liquids may differ very much, even at equal pressure and temperature. This phenomenon enables e.g. separation of components in the course of absorption and desorption operations, respectively. From the point of separation costs the initial costs, as well as the operating costs of the equipment intended for realization of this operation, play a rather significant role. In traditional packed towers and spray columns applied to the above-mentioned operations the established dispersed liquid phase will meet the gas phase in a countercurrent flow, in the form of film or drops. Mass transfer occurs by diffusion at the boundary surface of both phases, the extent of which depends on the diffusion rate.

At the boundary surface of the disperse phases the concentration of the diffusing constituent is in equilibrium. The diffusion rate will be the higher phase, the higher the difference of concentration of the diffusing constituent is in the bulk of the phase and at the boundary surface, i.e. the higher the driving force is. In case of slightly soluble gases (such as CO_2, O_2, air, etc.) in the liquid phase, the driving force is very low. In such a case, diffusion rate between the phases is limited by the low driving force within the liquid film. The larger

1*

phase boundary surface, as well as the higher mass transfer coefficient increase the extent of mass transport.

The height of absorption or desorption columns and herewith their initial costs are determined by the diffusion rate in case of a given task. On the basis of the afore-said considerations this can be increased by means of rising the phase boundary surfaces, and/or by increasing the mass transfer coefficients.

Packed columns (e.g. Raschig rings, Berl saddles, Intalox saddles) which produce the disperse liquid phase in the traditional way, yield large phase boundary surfaces. All these, however, scarcely improve the mass transfer coefficients at the liquid side, since a laminar liquid film is formed at the surfaces of packings and diffusion acts mainly as molecular transport in the liquid phase, which is a rather slow process. In such conventional cases when diffusion rate is controlled by liquid film conditions (resistance), the creation of turbulent diffusion, in order to increase mass transfer, would mean a step forward in this field.

Diffusion is turbulent if instead of the molecules, groups of molecules consisting of a great number of molecules are introduced into the bulk of the liquid phase from the phase boundary surface. This can be done by disintegration of the phase boundary surface and mixing it back into the bulk of the phase, steady renewal of the surface and creation of turbulent diffusion.

All kinds of specially shaped packing elements (e.g. Tellerettes) approach this target [1]. These are usually perforated elements with large surfaces and provided with many edges which serve to divide the liquid into drops. Now, the developed drops collide and hit against the surfaces of packing elements, thus disintegrating the liquid phase repeatedly and renewing the phase boundary surface meanwhile also improving the mass transfer coefficient at the liquid side.

The application of known procedures is rather limited in cases, where the liquid or the gas phase contains any solid substance, e.g. flue-dust in waste-gas, since the packed bed serves here as a filter and will get obstructed, increasing thus resistance, and consequently the operating costs, too.

Turbulent diffusion cannot be increased to a notable extent any further by means of application of ever newer and more particularly shaped packing elements, besides, their manufacture is very expensive.

Reduction of mass transfer at the gas side and especially the increase of the separating effectivity of solid substances in the gas phase are aimed at through known processes and equipment [2], by means of using floating beds which are moving beds with a flow rate lower than that of the liquid phase and are destined to disintegrate the disperse gas phase, to renew its surface and to raise turbulency at the gas side. This can be achieved by the application of a floating bed (lower flow rate than that of the liquid phase) in the continuous liquid phase maintaining the elevation of the bubbling through a disperse gas

phase, renewing the bubbles at the same time. Numerous procedures and equipment of this kind, using floating beds in the liquid phase, have been applied during the past 10–20 years.

Considering the solutions described above it can be stated that gas absorption and desorption, as well as the separating effectivity of dust and simultaneous gas and dust scrubbing, respectively, are insufficient. These installations cannot be loaded within a wide range, except with a rather high increase of operating costs.

Fluid/disperse process and arrangement (FDC)

In order to eliminate the above deficiencies, a more effective process than those known up to now, has been elaborated for application to gas absorption and desorption, dust and simultaneous gas and dust scrubbing, respectively, as well as for continuous contacting distillation processes, namely the three-phase fluid disperse process and arrangement (hereinafter called: FDC-process, where FDC = Fluid Disperse Column).

The FDC-process is based upon the realization that the above aim can be met if for the sake of increasing mass transfer, both turbulent diffusion and surface extension will be increased, furthermore the agitation of the liquid phase will be carried out very intensively, in a way that a fluidized bed will be applied in the continuous gas phase into which the liquid will be introduced in a disperse phase [3].

In the FDC-system the gas and liquid phase related in a countercurrent are introduced into the packed bed area of the column. The packed bed area is filled up continuously by the gas phase, proceding fluidization of the packed bed. Then the liquid phase, as a disperse phase, is led into the packed bed which is kept fluidized by means of the continuous gas phase. The fluidized packing elements carry out continuous disintegration, surface-renewal and agitation of the developing liquid phase. The result is the formation of a large phase boundary surface and intensive turbulent diffusion.

In order to eliminate back-mixing of the fluidized packing elements within the column, it is expedient to apply simultaneously packing layers which consist of elements of diverse flow rate, and/or size, and/or shape. Another practical solution may be the use of separating grids to divide packing layers within the column, in order to hinder the axial back-mixing of the fluidized packing elements. The flow rate of the packing elements which form the bed is higher than that of the flowing phases, and their advantageous shape can be prismatic, cylindrical or spherical.

The schematic operation diagram of an FDC column is shown in *Fig. 1*. Gas and liquid passes in a countercurrent through the column, where gas is the continuous phase and liquid the disperse one all along the process.
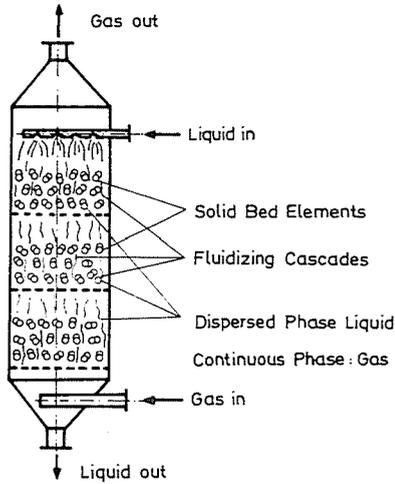
Fig. 1. Scheme of FDC Column

Experiments (pilot plant)

The target of our experiments was to examine the hydrodynamic, mass transfer and scrubbing properties of a three-phase, fluid disperse column.

A pilot plant model has been established for the above purpose which was found convenient to enable a change of the type of packings (material, size, shape), the number of cascades, the gas and liquid loadings. In the course of our experiments we have carried out measurements to examine the pressure drop of the packed bed, the minimum fluidizing velocity, flooding conditions, liquid hold-up in case of air/water flow phases, absorption rate of a very soluble gas (NH_3) introduced into water, absorption rate of a slightly soluble gas (CO_2) introduced into water, as well as the scrubbing of test-powder from air polluted by it, with the aid of water. The diameter of the FDC column (pilot plant) established to carry out our experimental examinations was $\varnothing$ 120 mm, and the packing elements applied to the bed were polystyrene spheres with a diameter of 10 and 14 mm, resp.

Results of Hydrodynamic Examinations

Figure 2 illustrates the characteristic curves of the pressure drop value in the FDC column where the overall operating area was used as a single cascade with the packed bed composed of polystyrene spheres of diam. 14 mm. On the basis of the graph it can be stated that considering the minimum fluidizing velocity of the dry packings, the value of minimum fluidizing velocity decreases
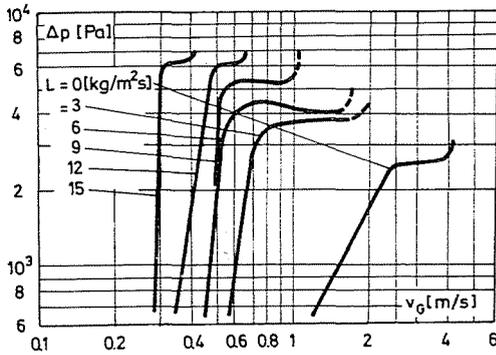
Fig. 2. Effect of Liquid Load on Pressure Drop

remarkably as liquid loading is increased. A quasi-steady range of fluidization develops at each tested loading, thus it can well be seen how pressure drop, measured in fluidized state, rises if liquid loading is increased. This phenomenon can be explained by an increase of the volume of liquid hold-up. The operating range of the column decreases if liquid loading is increased, since flooding then develops earlier in the tower.

The effect of the number of cascades can be seen in *Fig. 3.* If packed beds of identical bulk are divided into different numbers of cascades, the hydro-dynamic characteristic curve changes accordingly. If the number of cascades is increased, the minimum fluidizing velocity increases, the pressure drop occurring in the quasi-steady fluidized state decreases, due to the volume decrease of liquid hold-up in the tower.

The formation of the volume of liquid hold-up is shown in *Figs 4* and *5*. *Figure 4* clearly demonstrates that in case of a fixed bed slow increase in the
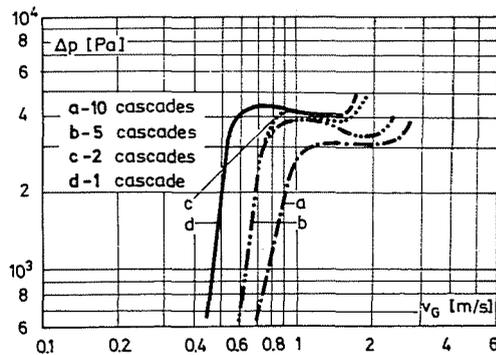
Fig. 3. Effect of Number of Cascades on Press. Drop.
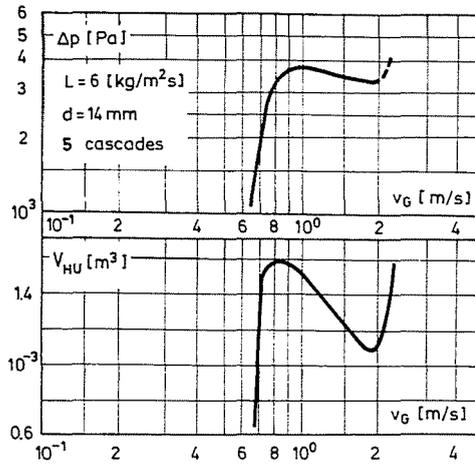
Fig. 4. Eff. of Gas Velocity on Liquid Hold-Up (V_{HU})
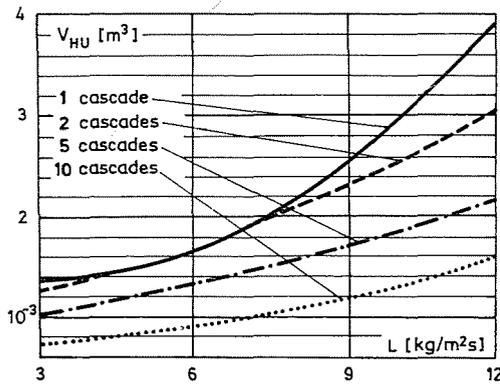
Fig. 5. Eff. of Liquid Rate on Liquid Hold-Up (V_{HU})

volume of liquid hold-up occurs if gas loading is increased, rapid growth of liquid hold-up precedes achievement of the minimum fluidizing velocity. The maximum value of the volume of liquid hold-up coincides with the maximum value of pressure drop of the bed. With a further increase of gas loading, the volume of liquid hold-up will speedily decrease due to the expansion of the bed, then after the starting of flooding it will increase rather rapidly. *Figure 5* illustrates the effect of the number of cascades on liquid hold-up in the range of quasi-steady fluidization. It can be stated that with the increase of the number of cascades the volume of liquid hold-up decreases.

We have compared the result of our experimental examinations with those measured in towers packed with traditional Raschig rings. From the hydrodynamical point of view, the packing spheres of diam. 10 mm were practically equivalent, according to [4], to Raschig rings of $15 \times 15 \times 2$ mm size. *Figure 6* shows the formation of the pressure drop in the FDC column and in the column packed with Raschig rings, without any liquid introduction, and at a given liquid-loading, resp. On the basis of the graph it becomes evident that in the quasi-steady range of fluidization of the FDC column pressure drop is much lower than in the column packed with Raschig rings, which also has a much lower loadability at the gas side, whereas the FDC column bears the increase of loadings very elastically with an overloadability of practically 100%, without neither the danger of flooding, nor a considerable increase of pressure drop.
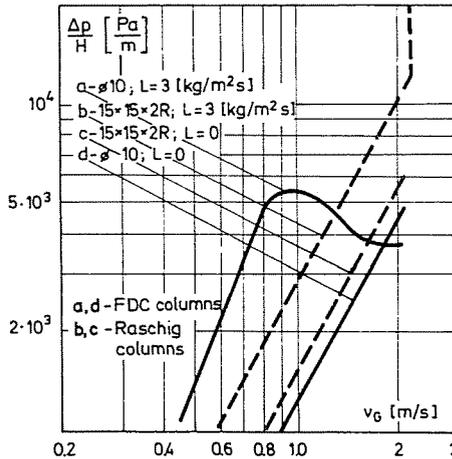
Fig. 6. Comparison of Pressure Drop in FDC and Raschig Columns

Results of Mass Transfer Tests

In the course of mass transfer tests we have determined the values of the so-called height of the transfer unit. It is well known [4, 5] that the height of the packed bed required for the prescribed concentration variation in case of continuous phase contacting can be written as:

$$H = H_{0L} N_{0L} = H_{0G} N_{0G} \tag{1}$$

where

$$H_{0L} = \frac{L_m}{K_L a A_q}, \qquad H_{0G} = \frac{V_m}{K_G a A_q} \tag{2}$$

and

$$N_{OL} = \frac{X_{out} - X_{in}}{\Delta X_{log}^*}, \qquad N_{OG} = \frac{Y_{in} - Y_{out}}{\Delta Y_{log}^*} \qquad (3)$$

The terms ΔX_{log}^* and ΔY_{log}^*, respectively, in Eq. (3) represent the logarithmic mean driving forces.

In case of the absorption of NH_3 (highly soluble gas), the following relationship:

$$H_{OG} \cong H_G = \frac{V_m}{k_G a A_q}, \qquad (4)$$

and for the absorption of CO_2 (slightly soluble gas), the relationship:

$$H_{OL} \cong H_L = \frac{L_m}{k_L a A_q} \qquad (5)$$

can be assumed.

In the course of the tests we have measured the proceeded concentration variations and driving forces for a given bed-height, thus we could determine the values of the height of the transfer unit according to Eq. (1). The results are shown in *Figs* 7 and 8. In the graphs the results obtained by using a fixed bed of Raschig rings have been plotted, too. On the basis of *Fig. 7* it can be stated that in case of absorption of slightly soluble gases, when the velocity of mass transport is controlled by resistance at the liquid side, the results obtained by using the FDC column are much more favourable. It can also be stated that the hold-up of axial back-mixing by dividing the packed bed into cascades, helps to
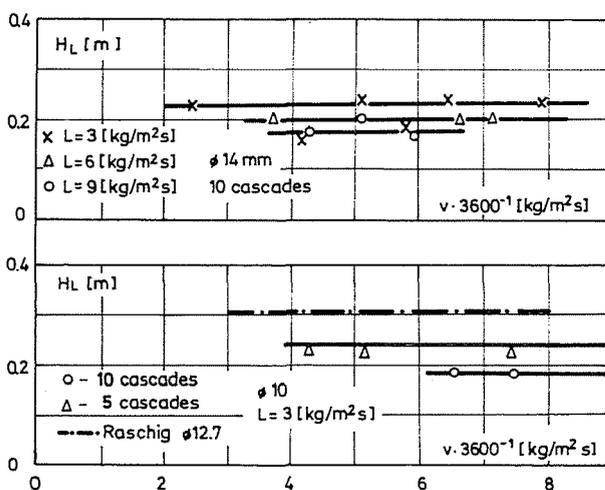
Fig. 7. Comparison of Heights of Transfer Unit (H_L) in FDC and Raschig Towers

increase the rate of mass transfer to a rather high degree. The increase of liquid-loading reduces the value of the height of transfer unit. We have applied the correlation:

$$H_L = \beta \left(\frac{\overline{L}_m}{\mu_L}\right)^n Sc_L^{0.5} \tag{6}$$

suggested by Sherwood and Holloway [5] when calculating the height of the transfer unit, and applying spheres of diam. 14 mm yielded the following results (for the constants):

diam. 14 mm	β	n
1 cascades	0.1096	-0.2
5 cascades	0.0632	-0.2
10 cascades	0.0427	-0.2

Figure 8 illustrates the results obtained by absorption of NH_3. Since the rate of absorption of highly soluble gases is controlled by the gas side, much better results cannot be expected by the application of FDC columns either. The figure clearly shows that due to the hold-up of axial back-mixing of the disperse liquid phase, the results obtained by a column packed with Raschig rings are also attainable, but since the FDC column has a much higher loadability and a lower pressure drop, its application is very economical in such a case, too.

On the basis of the results of tests in the pilot plant, the prototype of a large-scale FDC column has been dimensioned and built for the stripping of CO_2 from a softened water-circuit. This prototype arrangement has been
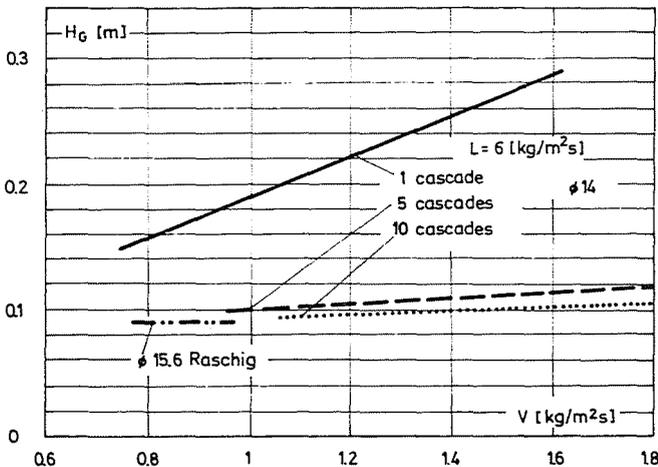
Fig. 8. Comparison of Heights of Transfer Unit (H_G) in FDC and Raschig Towers

established for the stripping of CO_2 from water of 5 m³/h, the prescribed concentration variation being:

$$X_{in} = 250 \text{ mg } CO_2/m^3 \text{ water}$$

$$X_{out} = 20 \text{ mg } CO_2/m^3 \text{ water}$$

The FDC column has been connected in parallel to the technological process which was established in order to accomplish the task by applying the traditional column packed with Raschig rings. In the course of measurements carried out during the operation, the stripping (separation) efficiency of CO_2 has been determined in both towers, as:

$$\eta_{CO_2} = \frac{X_{in} - X_{out}}{X_{in}} 100 \tag{7}$$

Figure 9 illustrates the results. It can be stated that better stripping efficiency can be obtained by means of the FDC column than by using the traditional packed tower with Raschig rings.
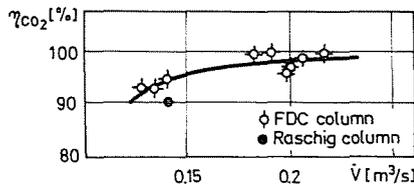
Fig. 9. Comparison of Stripping Efficiency (CO_2) in FDC and Raschig Columns

	FDC column	Raschig tower
Diameter, m	0.5	1.05
Height of the packed bed, m	1.2	2.4
Bulk of the packed bed, kg	41	1000

More data about the results of the tests in the pilot plant can be found in detail in publication [3].

Results of Scrubbing Examinations

Scrubbing experiments were also carried out in the FDC column, with application of test-powders for this purpose. These test-powders were organic pigments, produced by BASF. Their particle-size distribution was highly monodisperse, and their characteristic geometric dimensions were: 0.96 µm

and 2.1 µm, respectively. As far as scrubbing is concerned, axial back-mixing of the liquid phase has no importance what was proved by the experimental results, too. The increase of the number of cascades scarcely increased scrubbing efficiency.

The overall scrubbing efficiency is shown in *Fig. 10*, according to which it can be stated that the FDC column enabled separation of pollution of even a 0.96 µm characteristic particle size with an efficiency higher than 96%.

Comparisons of the separating effectivity of the FDC column were made on the basis of literature cited in (6), considering the separating effectivity and specific energy consumption as demonstrated in *Fig. 11*. The diagram clearly shows that the FDC column requires much less energy to assure separation equivalent to that of e.g. a Venturi washer or approx. equivalent to that of an electrostatic scrubber.
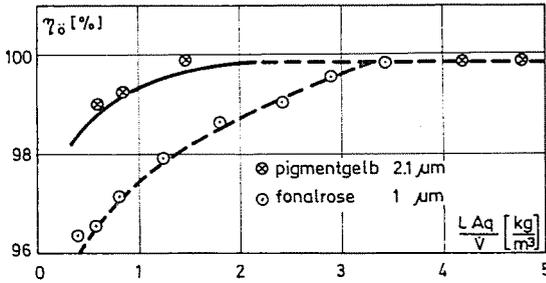
Fig. 10. The overall scrubbing efficiency
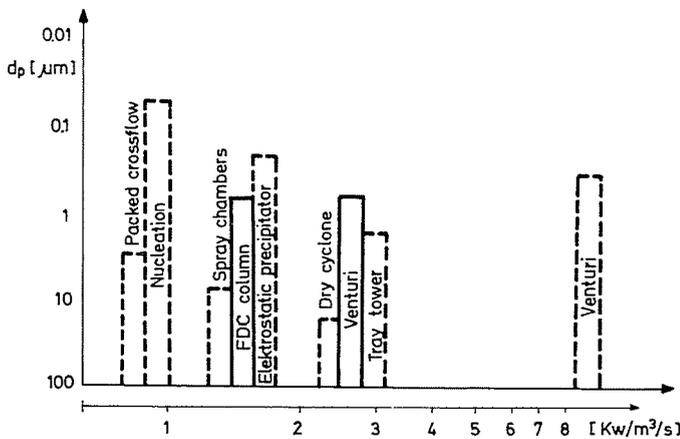
Fig. 11. The separating effectivity and specific energy

Summary, suggestions

According to our experimental observations it has become evident that the application of fluidized beds has several advantages, since due to their operation, as described, besides increase of the rate of mass transfer, the alteration of loadings can be carried out within a wide range, keeping the pressure drop practically constant at lower operation costs than required by the use of traditional fixed bed and floating bed columns.

The field of application of the FDC system is much wider than that of traditional systems, since it is satisfactorily applicable to purification of gases being polluted by solid substances or dust, when technological problems or tasks of environment-protection have to be solved. Moreover, experiments are in course at present, for the application of the FDC system to distillation operations, too.

Scrubbing is carried out in a way that the gas containing solid substances, e.g. carbon deposits, ash or vapours of any kind is introduced from the bottom of the column, according to the described process, thus establishing the fluidizing state of the packed bed. The disperse liquid phase, however, will be led into the moving bed from above. The small liquid-drops then collide with the solid particles contained in the gas. The liquid-drops surround the solid particles and enclose them. The purified gas is led off at the top of the column, while the outlet of the sludges, containing solid substances occurs through the liquid seal at the bottom of the column.

On the basis of our experimental (pilot plant) investigations, as well as of the results of large-scale examinations it can be seen that the FDC process and installation enables separation of materials from gases or liquids, the absorption and desorption of gases, as well as scrubbing in an entirely new and efficient manner. Owing to its high effectivity, the FDC system has the special advantage that smaller equipment can be built for a given task.

Nomenclature

a = specific phase boundary surface, m^2/m^3
A_q = void cross-sectional area of column, m^2
H = height of packed bed, m
H_{0G} = height of transfer unit at the gas side, related to the driving force, m
H_{0L} = height of transfer unit at the liquid side, related to the driving force, m
H_G = height of highly soluble gases, m
H_L = height of transfer unit, in case of slightly soluble gases, m
k = mass transfer coefficient, $kmol/m^2$, s
K = overall mass transfer coefficient, $kmol/m^2$, s

L_m = liquid flow in mols, kmol/s
$\bar{L}_m$ = flow rate of liquid, kmol/m^2, s
$\bar{L}$ = flow rate of liquid bulk, kg/m^2, s
n = constant
N_{0G} = number of transfer units at the gas side, referred to the driving force,
N_{0L} = number of transfer units at the liquid side, referred to the driving force,
Δp = pressure drop, Pa
SC_L = Schmidt number for liquid phase,
v = linear velocity, m/s
V' = volumetric gas flow, m^3/s
V = flow rate of gas bulk, kg/m^2, s
V_m = gas flow in mols, kmol/s
X = mol concentration in the liquid phase, kmol$_{solute}$/kmol$_{solvent}$
Y = mol concentration in the gas phase, kmol$_{solute}$/kmol$_{solvent}$

Greek Letters

β = constant
η = separating efficiency (stripping, scrubbing)
μ_L dynamic viscosity of liquid, Pas

Subscripts

bed = related to bed
in = inlet
G = gas phase
L = liquid phase
m = in mols
out = outlet
0 = resistances of both phases should be considered
q = related to plant
w = water

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