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Proposed prediction method for the frictional pressure drop of bubble-filled electrolytes

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A relationship is derived to predict the pressure drop in a two-phase flow system between gas evolving electrodes and in the pipes between the cells. The design equation $(\mathrm{d}p/\mathrm{d}x) = [(1+\phi)^n/(1-\phi)](\mathrm{d}p_\mathrm{L}/\mathrm{d}x)$ only requires the flow rates of the gas and liquid and the single-phase (liquid) pressure drop to be known. The equation is compared with other theoretical and empirical prediction methods, and with experimental data.

Nomenclature

C	geometry factor
d_B	diameter of the departing bubbles (m)
d_h	hydraulic diameter (m)
k_s	wall roughness (m)
K_L	multiplier
L	length of electrode in flow direction (m)
n	exponent in Equation 16
p	pressure ($\mathrm{kg\,m^{-1}\,s^{-2}}$)
Re	Reynolds number
s	interelectrode distance (m)
S	cross-sectional flow area ($\mathrm{m^2}$)
V_G, V_L	volumes of gas and liquid, respectively ($\mathrm{m^3}$)
$\dot{V}_\mathrm{G}$, $\dot{V}_\mathrm{L}$	volumetric flow rate of gas and liquid, respectively ($\mathrm{m^3\,s^{-1}}$)
x	coordinate in flow direction (m)
X	parameter due to Equation 19
η	viscosity ($\mathrm{kg\,m^{-1}\,s^{-1}}$)
θ	fractional surface coverage
ξ	friction coefficient
ρ	density ($\mathrm{kg\,m^{-3}}$)
ϕ	volumetric gas fraction
ψ	Thorpe's multiplier, Equation 25

Indices:

A	anode
C	cathode
G	gas
L	liquid
T	cell exit

1. Introduction

In chemical engineering, two-phase flow of a gaseous phase and liquid is frequently encountered in boiling and in cases where gas is dispersed into liquids by means of nozzles. Furthermore, two-phase flow occurs in cells with gas evolving electrodes and in connecting pipes. This is a different case from the others essentially because: 1. The volume fraction of gas in the electrolyte is always small in the interelectrode space to avoid an unreasonable increase in ohmic voltage drop. The average volume fraction of gas does not usually exceed 40% and is very often much lower. The flow pattern is usually bubble flow. The electrolyte always forms the continuous phase. 2. Gas bubbles departing from the electrodes are extraordinarily small. Whereas steam bubbles or gas bubbles from nozzles have diameters of some millimeters, the size of gas bubbles evolved at gas evolving electrodes is of the order of 50 μm [1]. As a result, the relative velocity of gas bubbles in electrolytes is very small and, hence, momentum transport between the two phases is negligible.

Although many papers are available dealing with the pressure drop in two-phase flow, particularly of vapour and boiling liquid in the full range of fractions of the gaseous or vapour phase (from mist flow to bubble flow), except for an empirical correlation by Thorpe *et al.* [2] as far

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as the author is aware no attempt has been made to develop a pressure drop relationship specifically designed for bubble filled electrolytes, taking into account the features mentioned above.

2. Proposed method

A single gas bubble having a diameter of 50 μm rises with a steady-state velocity of about 2 mm s^{-1} relative to the electrolyte. Bubbles in a swarm rise even slower [1]. On the other hand, the absolute electrolyte velocity is larger by some orders of magnitude in industrial cells where an instantaneous gas release is required. Under these conditions, the slip between gas and liquid may be neglected in agreement with experimental findings [3]. This situation is typical of vertical inter-electrode gaps and pipes at even moderate liquid velocities. It also applies approximately to horizontal gaps with large electrolyte velocities. Therefore, it is reasonable to consider the dispersion of bubbles in the electrolyte hydrodynamically as a quasi-homogeneous system, the pressure drop of which is decisively affected by the mean properties of the dispersion. Thus the pressure drop may be written as

$$\frac{\mathrm{d}p}{\mathrm{d}x} = \frac{\xi}{d_\mathrm{h}}\frac{\rho}{2}\left(\frac{\dot{V}_\mathrm{G} + \dot{V}_\mathrm{L}}{S}\right)^2 \tag{1}$$

where the symbols without an index refer to the mixture.

Since in electrolysis, in contrast to boiling, the density of the liquid is always much greater than the density of the gas, the density of the dispersion

$$\rho = (1-\phi)\rho_\mathrm{L} + \phi\rho_\mathrm{G} \tag{2}$$

approximates to

$$\rho = (1-\phi)\rho_\mathrm{L}. \tag{2a}$$

With the gas fraction, written for zero slip

$$\phi \equiv \frac{V_\mathrm{G}}{V_\mathrm{G} + V_\mathrm{L}} = \frac{\dot{V}_\mathrm{G}}{\dot{V}_\mathrm{G} + \dot{V}_\mathrm{L}} \tag{3}$$

Equation 1 becomes

$$\frac{\mathrm{d}p}{\mathrm{d}x} = \left(\frac{1}{1-\phi}\right)\frac{\xi}{d_\mathrm{h}}\frac{\rho_\mathrm{L}}{2}\left(\frac{\dot{V}_\mathrm{L}}{S}\right)^2 \tag{4}$$

or

$$\frac{\mathrm{d}p}{\mathrm{d}x} = K_\mathrm{L}\frac{\mathrm{d}p_\mathrm{L}}{\mathrm{d}x}. \tag{5}$$

Prediction of the two-phase pressure drop is dependent only on the pressure drop of the bubble-free electrolyte

$$\frac{\mathrm{d}p_\mathrm{L}}{\mathrm{d}x} = \frac{\xi_\mathrm{L}}{d_\mathrm{h}}\frac{\rho_\mathrm{L}}{2}\left(\frac{\dot{V}_\mathrm{L}}{S}\right)^2 \tag{6}$$

and a multiplier

$$K_\mathrm{L} = \left(\frac{1}{1-\phi}\right)\left(\frac{\xi}{\xi_\mathrm{L}}\right) \tag{7}$$

which contains all the unknowns in the method. It will be shown, however, that apart from the gas fraction it is only dependent on the viscosity ratio of the dispersion and the pure electrolyte.

2.1. *Multiplier*

The ratio of the friction coefficients in Equation 7 depends on the flow conditions. In laminar flow, the friction coefficient is

$$\xi = \frac{\text{const.}}{Re} = \frac{\text{const.}\, S\,\eta}{\dot{V} d_\mathrm{h}\,\rho}. \tag{8}$$

For the limiting case where both the electrolyte, calculated to flow under the assumption of the absence of gas, $(Re_\mathrm{L} = (\dot{V}_\mathrm{L} d_\mathrm{h}\rho_\mathrm{L})/(S\eta_\mathrm{L}) < 2300)$, and the dispersion are in laminar flow one obtains

$$\frac{\xi}{\xi_\mathrm{L}} = \frac{\eta}{\eta_\mathrm{L}}\frac{\rho_\mathrm{L}}{\rho}(1-\phi) \tag{9}$$

and a multiplier (together with Equation 2a):

$$K_\mathrm{L} = \left(\frac{1}{1-\phi}\right)\left(\frac{\eta}{\eta_\mathrm{L}}\right). \tag{10}$$

With the friction coefficient for turbulent flow at moderate Reynolds numbers ($Re < 10^5$) [4]

$$\xi = \frac{\text{const.}}{Re^{0.25}} \tag{11}$$

follows

$$\frac{\xi}{\xi_\mathrm{L}} = \left(\frac{\eta}{\eta_\mathrm{L}}\right)^{0.25} \tag{12}$$

and

$$K_\mathrm{L} = \left(\frac{1}{1-\phi}\right)\left(\frac{\eta}{\eta_\mathrm{L}}\right)^{0.25} \tag{13}$$

while for strongly turbulent flow at very large Reynolds numbers, where the friction coefficient is independent of the Reynolds numbers,

$$\frac{\xi}{\xi_\mathrm{L}} = 1 \tag{14}$$

and

$$K_L = \left(\frac{1}{1-\phi}\right). \tag{15}$$

The various expressions of K_L may uniformly be written for bubble-filled electrolytes as

$$K_L = \left(\frac{1}{1-\phi}\right)\left(\frac{\eta}{\eta_L}\right)^n \tag{16}$$

where $n = 1$ for laminar flow and $n = 0.25 \ldots 0$ for turbulent flow.

The effect of the viscosity ratio is smaller the larger the Reynolds number. Note that Equation 16 replaces Equation 7 provided there is equal wall roughness both in the one-phase and two-phase flow systems.

The viscosity ratio in Equation 16 may be estimated by introducing the equation of Taylor [5] who extended the Einstein equation [6, 7] for the viscosity of a suspension of solid spheres in liquids to fluid drops dispersed in liquids. The essential assumptions of the treatment (the shape of the dispersed phase being nearly spherical, and slipping at the interface being zero) are fairly realistic for electrolytically evolved bubbles.

$$\frac{\eta}{\eta_L} = 1 + 2.5\phi\left(\frac{\eta_G + \frac{2}{5}\eta_L}{\eta_G + \eta_L}\right). \tag{17}$$

As pointed out by Taylor [5], the factor in brackets takes account of the flow set up inside a drop or a bubble and may be compared with a similar factor proposed by Hadamard [8] and Rybczynski by which Stokes' expression of the relative bubble velocity must be multiplied to take account of the internal flow.

Viscosity values for common gases evolved at gas evolving electrodes (H_2, O_2, Cl_2) and for common electrolytes (aqueous solutions and fused salts) in the industrial temperature range show that the viscosity ratio is always $\eta_G/\eta_L < 0.1$. Values of the multiplier K_L from Equation 16 with two ratios η_G/η_L from Equation 17 are plotted in Fig. 1 for various flow conditions. It is seen that the effect of viscosity ratio is small. Equation 17 may, therefore, be approximated by

$$\frac{\eta}{\eta_L} = 1 + \phi \tag{17a}$$

for bubble filled electrolytes. The multiplier simplifies to

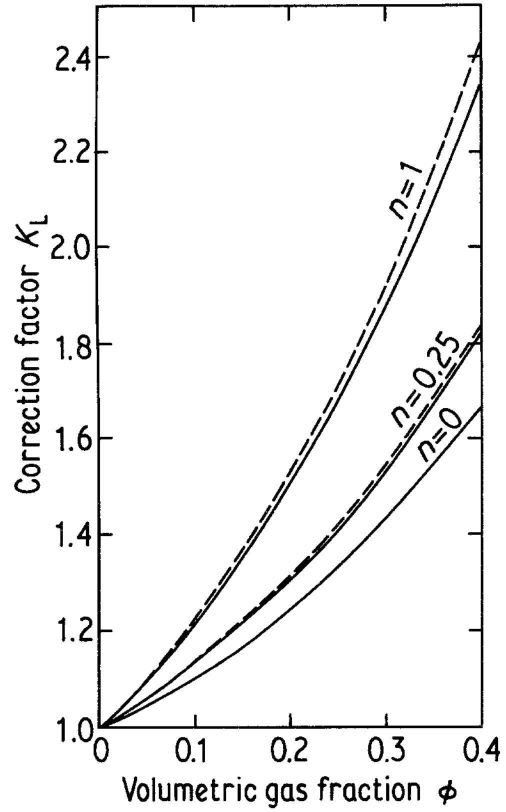

Fig. 1. The correction factor K_L versus volumetric gas fraction ϕ for laminar ($n = 1$) and turbulent ($n = 0$; 0.25) flow based on Equations 16 and 17. —— $\eta_G/\eta_L = 0$; ---- $\eta_G/\eta_L = 0.1$.

$$K_L = \frac{(1+\phi)^n}{(1-\phi)}. \tag{18}$$

2.2. *Pressure drop of bubble-free electrolytes*

In computing the pressure drop of the pure liquid due to Equation 6 the following features have to be taken into account. In *laminar* flow the friction coefficient $\xi_L = 64\,C/Re_L$ depends on the shape of the duct: $C = 1$ for circular ducts (tubular pipes) and $C = 1.5$ for narrow gaps (interelectrode gaps). In cases where the sectional flow area is diminished by bubbles adhering to the electrodes it is recommended, based on the experiments of Nunner [9], that the hydraulic diameter is calculated from $s - (d_{B,A} + d_{B,C})$ instead of the interelectrode distance s. d_B is the average diameter of the bubbles departing from the anode and cathode, respectively, where $d_B \approx 0.05 \ldots 0.1$ mm in aqueous electrolytes [1].

In *turbulent* flow the hydraulic diameter remains rather unaffected by adhering bubbles. However, the friction coefficient ξ_L is strongly affected by the wall roughness which need not necessarily be the roughness of the wall material. If the wall is partly covered by additional obstacles (such as devices in the form of rings attached to the wall) the effective wall roughness may greatly exceed the roughness of the wall material, as shown by Nunner [9]. The same effect must be attributed to adhering bubbles in all cases where the walls of a duct are gas evolving electrodes. Since Equation 16 applies to equal wall roughness in one-phase and two-phase flow, calculation of the friction coefficient ξ_L in the single-phase pressure drop of Equation 6 must be based on the wall roughness effective in two-phase flow.

In gas evolving electrodes, the additional wall roughness may tentatively be characterized by the well-known fractional coverage of gas, θ, i.e. the fraction of the electrode surface shadowed by adhering bubbles in orthogonal projection [10]. For that purpose, θ is equated with the portion of the wall area covered by the additional devices installed to increase the effective wall roughness in the experiment. Fig. 2 expresses the effective wall roughness due to devices of various shapes covering the wall versus the fractional coverage based on experimental data [9]. The diagram relates the artificial wall roughness, expressed as the bubble departure diameter d_B, to the equivalent wall roughness k_s which is needed to determine the friction coefficient $\xi_L = f(Re_L, k_s/d_h)$ from the customary diagrams. Values of θ depend on the properties of the electrodes, the electrolyte and, mainly, on the current density. Reference values were compiled recently [11]. For computing the pressure drop in the connecting pipes, where gas evolution does not occur, the effective wall roughness is the roughness of the pipe material.

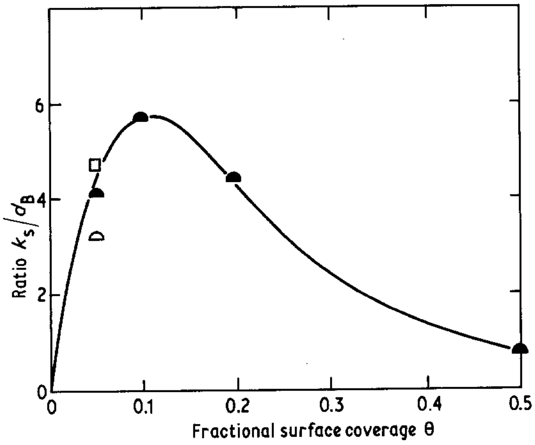

Fig. 2. The ratio of the effective wall roughness k_s and the bubble departure diameter d_B versus the fractional surface coverage, θ, based on experimental data of Nunner [9] with artificial wall roughness. □ ring with rectangular cross-section 2 mm × 2 mm; ○● ring with a semicircular cross-section of 2 and 4 mm radius, respectively.

3. Comparison of the proposed method with other design methods and with experimental data

The classic design method is the empirical one of Lockhart and Martinelli [12], based on Equation 5. The multiplier, however, depends on the parameter

$$X = \left(\frac{dp_L}{dp_G}\right)^{0.5} = \left(\frac{\xi_L}{\xi_G}\frac{\rho_L}{\rho_G}\right)^{0.5}\frac{\dot{V}_L}{\dot{V}_G} \tag{19}$$

where

$$\frac{dp_G}{dx} = \frac{\xi_G}{d_h}\frac{\rho_G}{2}\left(\frac{\dot{V}_G}{S}\right)^2. \tag{20}$$

The boundary lines of laminar and turbulent flow for both phases which are each assumed to flow in the absence of the other one, are shown in Fig. 3 as lines 3 and 3′. Line 4 represents the lower bound of the multiplier based on work of Kriegel [13]:

$$K_L = 1 + X^{-2}. \tag{21}$$

Fig. 3 also shows the experimental data points of Arnold and Stechemesser, published by Brauer [14]; a very small selection out of an abundance of data collected in data banks [15], but representative in their scatter.

For comparison with the method proposed here the parameter X was transformed with Equation 3 for zero slip conditions to

$$X = \left(\frac{\rho_L}{\rho_G}\right)^{(1-n)/2}\left(\frac{\eta_L}{\eta_G}\right)^{n/2}\left(\frac{1-\phi}{\phi}\right)^{1-(n/2)}. \tag{22}$$

This was used to draw lines 1 and 2 in Fig. 3, representing Equation 18 for hydrogen and oxygen evolved in an aqueous electrolyte and for chlorine in a melt, based on the properties given in Table 1.

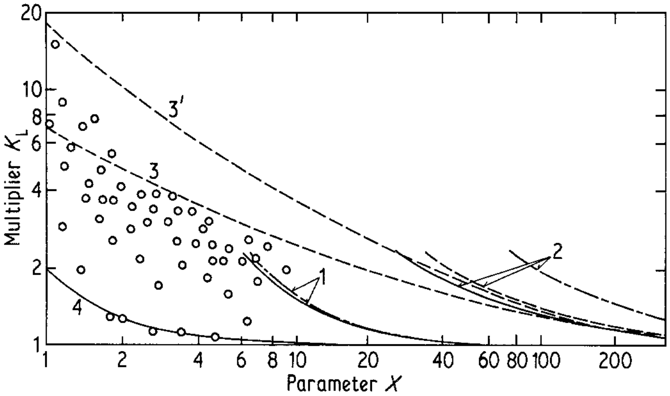

Fig. 3. Comparison of Equation 18 with various theoretical and experimental results. Equation 18: Line 1, laminar flow ($n = 1$); Line 2, turbulent flow ($n = 0.25$); properties of Table 1. Lines 3 and 3' are based on Lockhart and Martinelli [12] for laminar/laminar and turbulent/turbulent flow, respectively. Line 4 is based on Kriegel [13]. Data points of Arnold and Stechemesser [14].

Pressure drop data calculated from the proposed method are mostly somewhat lower than the Lockhart–Martinelli lines and larger than the lower bound presented by Kriegel [13]. Calculated data agree well with the experimental data.

Another method, developed by Bankoff [16], is restricted to low momentum transport between the phases, as occurs in gas bubble filled electrolytes. The basic equation is again Equation 5 but the meaning of the multiplier is now [16]

$$K_L = \left[1 - \phi\left(1 - \frac{\rho_G}{\rho_L}\right)\right]^{3/4} \left[\frac{1 + \left(\frac{\dot{V}_G}{\dot{V}_L}\right)}{1 + \left(\frac{\dot{V}_G}{\dot{V}_L}\frac{\rho_G}{\rho_L}\right)}\right]^{7/4}. \tag{23}$$

Realizing that for electrolytes containing gas bubbles, $\dot{V}_G/\dot{V}_L < 1$ and $\rho_G/\rho_L \ll 1$, Equation 23 reduces with Equation 3 to

$$K_L = \frac{1}{1-\phi}. \tag{24}$$

This result coincides with Equation (18) for the limiting case of strong turbulence in both phases, $n = 0$.

Finally the multiplier ψ is derived by Thorpe *et al.* [2] from experiments in a gas evolving cell,

$$\psi \equiv \frac{\Delta p}{\Delta p_L} = 1 + \text{const.}\, \phi_T \frac{\dot{V}_{G,T}}{\dot{V}_L} \frac{s}{L} \left(\frac{\rho_G}{\rho_L}\right)^{1/3}, \tag{25}$$

where Δp and Δp_L are the integral pressure drops of two-phase and single-phase flow, respectively, for the length L:

$$\Delta p = \int_0^L \frac{dp}{dx}\, dx. \tag{26}$$

The index T refers to the cell exit condition and $\dot{V}_{G,T}$ is the total gas flow rate evolved in the cell. Inserting Equation 3 for zero slip, as stated by Funk and Thorpe [3], Equation 25 results in

Table 1. Properties of electrolytes

Parameter		H_2 *evolution in* 1.75 N NaOH *at* 70° C	O_2 *evolution in* 1.75 N NaOH *at* 70° C	Cl_2 *evolution in* NaCl/LiCl/AlCl$_3$ *at* 700° C	*Reference*
η_L	($\times 10^{-6}$ kg m^{-1} s^{-1})	590	590	1200	[17, 19]
η_G	($\times 10^{-6}$ kg m^{-1} s^{-1})	19.7	23	39	[18]
$\frac{\eta_L}{\eta_G}$		30	26	31	—
ρ_L	(kg m^{-3})	1 046	1046	1500	[17, 19]
ρ_G*	(kg m^{-3})	0.084	1.34	1.05	—
$\frac{\rho_L}{\rho_G}$		12500	780	1430	—
Line in Fig. 3		—·—·—	———	--------	

* At 1.2 bar.

$$\psi = \frac{1 - \phi_T + \text{const.}\ \phi_T^2 \frac{s}{L}\left(\frac{\rho_G}{\rho_L}\right)^{1/3}}{1 - \phi_T}. \qquad (27)$$

Comparison of Equation 26 with Equation 5 gives

$$\psi = \int_0^L K_L \frac{dx}{L}. \qquad (27)$$

Inserting K_L from Equation 18 together with Equation 3 allows us to compare the Thorpe equation with the equation proposed in the present paper. Assuming, for the sake of simplicity, a linear increase of gas flow rate in the flow direction, $\dot{V}_G = \dot{V}_{G,T} \cdot x/L$, it follows for the case of fully developed turbulent flow ($n = 0$) that

$$\psi = \frac{1 - \frac{\phi_T}{2}}{1 - \phi_T} \qquad (29)$$

which shows a striking similarity to the empirical Equation 27.

4. Conclusion

Two-phase pressure drop in bubble-filled electrolytes may easily be computed from Equation 5 together with Equations 6 and 18 with sufficient accuracy for most industrial cases. The range of application is restricted to moderate volume fractions of gas, which usually occur in industrial cells with gas evolving electrodes.

The method is proposed to serve as an engineering tool for the design of two-phase flow in ducts inside and outside gas evolving cells. It is applicable to turbulent flow as well as to laminar flow, i.e. a condition which is of major importance in electrochemistry compared with other fields of chemical engineering. Since the flow pattern is of minor influence on the pressure drop, as shown by many investigations, the method does not apply only to small bubbles in vertical electrolyte flow but may approximately be used for horizontal interelectrode gaps and also for the pressure drop of dispersions of gas in fused salts.

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