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## Determination of mass transfer between gas and liquid in atmospheric leaching of sulphidic zinc concentrates

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#### Abstract

In atmospheric direct leaching of sulphidic zinc concentrates, oxygen acts as an oxidant in the dissolution. The amount of oxygen in the solution has an important effect on the kinetics of the whole process. In this paper, four laboratory scale equipment are used for studying the gas mass transfer in the leaching process. With each experimental set-up it is possible to analyse some of the factors affecting the  $k_L a$ ,  $k_L$  and a coefficients. The gas flow rate and mixing intensity increased the  $k_L a$  as expected. Also the  $k_L$  value was increased with mixing intensity, but increasing salt concentration had an opposite effect. Lowering the surface tension and increasing the density of liquid decreased the bubble Sauter mean diameter and therefore the total gas–liquid interfacial area a was increased. Oxygen consumption into solution was slightly increased with temperature. On the other hand the effect of pressure and mixing was notable. By combining the results of different equipment enables us to study the mass transfer as a function of parameters such as: temperature, pressure, liquid properties and mixing conditions. Knowing the mass transfer coefficients is essential for process development and design.

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#### 1. Introduction

In atmospheric leaching, the zinc concentrates are oxidised in sulphuric acid media, where iron couple  $Fe^{3+}/Fe^{2+}$  act as a intermediate between the dissolved oxygen and the mineral according to reactions Fugleberg (1998) and Takala (1999)

$$ZnS + Fe_2(SO_4)_3 = ZnSO_4 + 2FeSO_4 + S^0$$
<sup>(1)</sup>

and

$$2FeSO_4 + H_2SO_4 + 0.5O_2 = Fe_2(SO_4)_3 + H_2O$$
(2)

Because of relatively fast chemical reactions the dissolution of oxygen gas into the slurry has an important role on the kinetics of the whole process Salminen and Kaskiala (2003), which reaction formula is

$$ZnS + H_2SO_4 + 0.5O_2 = ZnSO_4 + H_2O + S^0$$
 (3)

Oxygen injection and dispersion into the process requires significant amount of effort and is one of the possible bottlenecks especially at the early stages of the processing. Therefore, knowing the factors affecting the gas-liquid mass transfer is essential for designing and optimising the process.

Gas-liquid mass transfer has been widely studied and a great deal of published information can be found. Volumetric mass transfer coefficient  $k_{L}a$  [1/s] correlations have been determined by several authors as a function of impeller rotational speed, gassing rate, tank and impeller geometry, fluid properties and rheology Tatterson (1991). Volumetric mass transfer coefficient  $k_{L}a$ 

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combines parameters  $k_{\rm L}$  and a, which eases the problems concerning total gas-liquid contact area and the interfacial phenomena are partly bypassed. However, the volumetric mass transfer coefficient is valid only for the equipment and the conditions examined. Analysing separately the coefficients  $k_{\rm L}$  and a enables the profound understanding of the physical properties of the fluids, chemical interactions and reactions effect on the gas-liquid interface.

In this paper, four laboratory scale equipment are used to determine the gas-liquid mass transfer coefficients  $k_L a$ ,  $k_L$  and a. With each experimental set-up it is possible to analyse some of the factors affecting them. Combining results by different equipment enables us to describe the gas-liquid mass transfer rate as a function of parameters such as: temperature, pressure, liquid properties and mixing conditions.

### 2. Theory

Chemical engineering operations can be divided into three different transfer phenomena: momentum, heat and mass transfer, which are presented with the same basic formula (4):

$$Flux = \frac{driving \ force}{resistance}$$
(4)

In most gas–liquid mass transfer cases the greater resistance for mass transfer is on the liquid side. This implies that  $k_{\rm L}$  [m/s] presents the total mass transfer resistance and formula of Fick's law for diffusional gas and liquid mass transfer flux can be written as:

$$J = k_{\rm L}(\Delta C) \tag{5}$$

where the driving force is the concentration difference  $(C^* - C)$  of saturated concentration of the gas in the liquid  $C^*$  and concentration C of the gas at the interface.

Mass transfer rate of gas A penetrating into liquid volume can be described as follows:

$$N_{\rm A} = k_{\rm L} a \Delta C \left[ \frac{\rm mol}{\rm m^3 s} \right] \tag{6}$$

Oxygen transport is dependent of the total gas-liquid interfacial area A [m<sup>2</sup>], which can be presented also as gas-liquid interfacial area per unit volume of fluid a[m<sup>2</sup>/m<sup>3</sup>]. Typically, the parameters  $k_L$  and a are combined into one parameter: the volumetric mass transfer coefficient  $k_L a$  [1/s]. The gas-liquid equilibrium of oxygen is described with Henry's law, which linear relationship is valid for dilute solutions of non-reacting systems and for gases which are weakly soluble in liquid. The temperature dependence of Henry's constant for oxygen in water is given by Fogg and Gerrard (1991) as follows:

$$\ln\left(\frac{P}{H_0}\right) = A + \frac{B}{T} + C\ln T \tag{7}$$

where  $H_0$  denotes the dimensionless representation of Henry's law constant in partial oxygen pressure P at 1 atm. The coefficients A, B and C are listed in Table 1.

Increasing salt concentration, the gas solubility is nearly always found to decrease due to "salting-out effect" of the ions. At moderate high salt concentrations, the effect of salt concentration,  $C_s$ , on the solubility,  $C_G$ , of a sparingly soluble gas as compared to that in pure water,  $C_{G,0}$  was described by Setschenow (1889) in the following from:

$$\log\left(\frac{C_{\rm G,0}}{C_{\rm G}}\right) = KC_{\rm s} \tag{8}$$

Parameter K, known as Setschenow constant, is specific to the gas as well as to the salt and shows a moderate temperature dependency Rönnholm et al. (1999). The equation can also be applied for mixed electrolyte solutions according to Schumpe (1993):

$$\log\left(\frac{C_{\rm G,0}}{C_{\rm G}}\right) = \log\left(\frac{H}{H_0}\right) = \sum_i (h_i + h_{\rm G})C_i \tag{9}$$

where  $C_i$  denotes the concentration of ion *i* in the solution and  $h_i$  is a ion-specific parameter. Weisenberger and Schumpe (1996) extended the model of Schumpe (1993) to the temperature range 273–363 K by assuming  $h_{\rm G}$ , (gas-specific constant) as a linear function of the temperature:

$$h_{\rm G} = h_{\rm G,0} + h_{\rm T}(T - 298.15) \tag{10}$$

where  $h_{G,0} = 0$  [m<sup>3</sup>/kmol],  $h_T = -0.334 \times 10^{-3}$  [m<sup>3</sup>/kmol K] and parameters  $h_i$  are listed in Table 2.

Solubility of oxygen in water and in salt solutions containing 1 M  $H_2SO_4$ , 1.3 M  $ZnSO_4$  and two complex process solution containing different composition of sulphuric acid, zinc sulphate and iron sulphates was calculated, see Fig. 1.

Table 1 Coefficients A, B and C used for calculating Henry's constant Fogg and Gerrard (1991)

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A	В	С	Temperature interval (K)
-171.2542 -139.485	8391.24 6889.6	23.24323 18.554	273–333 273–617

Table 2

Gas solubility parameters Weisenberger and Schumpe (1996)

$h_i$	$h_j$	$h_j$	
Cation m <sup>3</sup> /k	amol Anion	m <sup>3</sup> /kmol	
$H^+ = 0$	$SO_{3}^{2-}$	0.1270	
$Mg^{2+}$ 0.169	94 $SO_4^{2-}$	0.1117	
Mn <sup>2+</sup> 0.210	68		
$Fe^{2+}$ 0.140	63		
Fe <sup>3+</sup> 0.110	61		
$Cu^{2+}$ 0.165	54		
$Zn^{2+}$ 0.16'	75		

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