



Kinetic evidence for the effect of salts on the oxygen solubility using laboratory prototype aeration system



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ARTICLE INFO

Article history:

Received 6 March 2015

Received in revised form 15 July 2015

Accepted 25 July 2015

Available online 11 August 2015

Keywords:

Aeration
Oxygen mass transfer
Salt aqueous solutions
Hydrogen bond
Hydration

ABSTRACT

This study is dedicated to an accurate evaluation of thermodynamic and kinetic aspects in the oxygenation process of water/salt solutions. Oxygenation has been analyzed by means of kinetic study of oxygen dissolution from the oxygen mass transfer coefficient (K_{La}) and oxygen transfer rate ($SOTR$). A stirred, submerged aerated 4-liter system has been designed and the operational conditions have been optimized by studying the influence of hydraulic head, air flow and salinity of water using an optical oxygen sensor. Concerning the thermodynamic phase equilibria, experimental and modeling results are obtained from different binary systems (water/air) and ternary systems (water/air/salts). This information is necessary to predict the composition of the gas phase during the process and is also important for an implementation in a process simulation. The oxygen mass transfer coefficients were firstly measured, by monitoring in the time the oxygen concentration in various synthetic liquid phases containing different salts (NaCl, KCl, LiCl). When compared to clean water, noticeable increase of K_{La} was observed; the variation of K_{La} and $SOTR$ with the solution salinity was modeled and found dependent on the nature of cation in the salt added. For all cases, an increase of K_{La} with increasing salinity was observed but, when the results are correlated with salt molar concentration, very important relation attributed to different salts has been obtained. The present study clearly confirmed the importance to define the experimental conditions before to describe and to model appropriately the gas–liquid mass transfer phenomena.

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1. Introduction

Gas transfer in water and wastewater treatment is a process in which water is brought into contact with air or gas and, with the presence of a concentration gradient, the transfer of gases to and/or from the water occurs [1].

In the plant operation, aeration process provides to use air compressors or mechanical aerators to induce a higher concentration gradient in the wastewater and thereby to accelerate oxygen transfer.

Gas transfer is a transport phenomenon in which molecular diffusion plays an important role. Molecular diffusion effects can be described as the product of molecular diffusivity and the concentration gradient, as defined by Fick's law.

The various theories generally developed on the assumption that exist at the interface both gas and liquid films that can be classified into: a) the film model, b) surface renewal models, and c) other models representing various fields of interest [2].

Aeration theory is based on Henry's law: at equilibrium the partial pressure of a component in the gas phase is proportional to the concentration of this component dissolved in the liquid phase.

When the liquid phase is the mixed sewage and the gas phase is air, in particular molecular oxygen present in the air, the Henry's law can be written as:

$$DO_S = k_H \cdot P_{O_2} \quad (1)$$

where DO_S is the saturation concentration of dissolved oxygen in the sewage/water (mol l^{-1} or mg l^{-1}), k_H is the Henry's constant and P_{O_2} is the partial pressure of oxygen in air that is 0.21 atm at atmospheric pressure. k_H is dependent on temperature, pressure and salinity: with high values of temperature, saturation concentration of oxygen decreases, on contrary with high pressure values it observed an increase of concentration saturation value for oxygen.

Also, the presence of salt in water solution can affect the solubility of oxygen, in fact the saturation concentration for dissolved oxygen in water decreases as function of salt concentration [3–5].

Obviously in biological treatment system the dissolved oxygen concentration in the sewage will be less with respect to the saturation value in clean water, because oxygen is being consumed by the microorganisms. For clean water, the oxygen consumption rate depends on oxygen mass transfer.

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The specific mass transport rate of diffusion without convection is given by Fick's law [6]:

$$J_D = -D(dC/dx) \text{ mol m}^{-2} \text{ h}^{-1} \quad (2)$$

where D is the diffusion coefficient ($\text{m}^2 \text{h}^{-1}$) of oxygen in water, and C (mol m^{-3}) is the concentration of dissolved oxygen in water solution.

At the gas/liquid boundary, the calculated specific oxygen transfer rate in water by using Eq. (2) is low if only diffusion is involved. This follows not only from the low diffusion coefficient, but also from the low solubility of oxygen in water.

For 20 °C and deionized water k_H is equal to 32.6, for a concentration of O_2 in air equal to 298.7 mg l^{-1} , only 9.17 $\text{mg O}_2 \text{l}^{-1}$ are dissolved in water [6].

Therefore, oxygen transfer into water by diffusion is a slow process, due to the low diffusion coefficient and the low solubility.

Small bubbles, having a high surface area per unit volume, provide good oxygen–water contact, leading to relatively high values of the oxygen-transfer efficiency.

Lewis and Whitman in 1924 [7] postulated a theory, called Two-Film Theory, that is a transposition of Fick's law for the diffusion of a gas in turbulent flow as in the treatment water, in which laminar films exist at the gas–liquid interface.

The gas is dispersed in a liquid phase through a diffusion process due to a partial pressure gradient in the gas phase and by a concentration gradient in the liquid phase (water).

The control factor of this process, for a perfect mixed reactor, can be represented by the rate of the gas passage through the interface between the two phases according to Lewis and Whitman and it is given by [8]:

$$dC/dt = K_{La} (C_s - C_t) \quad (3)$$

where $K_{La} = K_L (S/V)$ (t^{-1}), C_s is the saturation concentration of oxygen in water at the operating condition (pressure, temperature, and salt concentration) and C_t is the dissolved oxygen concentration at time t during the oxygenation process.

K_{La} is the kinetic constant and it is called *gas liquid mass transfer coefficient* (with dimensions of t^{-1}) and derived from the product of the *liquid film coefficient* (K_L) in water with the ratio between the *total interface area* (S) and the *volume* (V) of the liquid mass that will be aerated [9].

In evaluation of an aerator the oxygen transferred is estimated under standard conditions, corresponding to a temperature of 20 °C and standard atmospheric pressure (1013.25 mbar, 1 atm).

The rate of oxygen transferred by an aerator is reported as its oxygenation capacity, which is defined as the rate of oxygen transfer dC/dt at start concentration of $C_0 = 0$ ($t = 0$), in standard conditions, *standard oxygen transfer rate* (SOTR) [9] represented by the Eq. (4):

$$SOTR(\text{kg O}_2 \text{ h}^{-1}) = K_{La(20^\circ\text{C})} \cdot C_{s(20^\circ\text{C}, 1 \text{ atm})} \cdot \text{volume}. \quad (4)$$

To determine the gas–liquid mass transfer in standard conditions, it is possible to use Eq. (5).

$$K_{La(20^\circ\text{C})} = K_{La(T^\circ\text{C})} / \theta^{(T^\circ\text{C} - 20)} \quad (5)$$

where θ is 1.024.

The interactions between water and solute are very important in high number of physical phenomena as partitioning of ions near interfaces [10–13], molecular structures [14–18] and solubility of dissolved gas [19].

Dissolved salts and organics tend to decrease the saturation concentration of oxygen in wastewater as compared with distilled water. When ionic salts are added to water, the introduced ions from the salt attract the water molecules in an effort to “solvate” the ions. This has the tendency to decrease the weak affinity of non-polar oxygen molecules to water and drive the dissolved oxygen out of the polar water giving salting out effect [9,20–22].

Although the activity of an oxygen-saturated solution of water is by definition independent of the dissolved contaminants, the concentration of oxygen changes as the activity coefficient is altered by the salting-out effect. Experimental studies conducted with distilled water and mixtures of distilled water and tap water with a total dissolved solid concentration of about 600 mg/L showed that salts also impact K_{La} that increases with increased specific conductivity [23]. Similar trends were obtained with the addition of sodium chloride to water demonstrating that high salt concentrations canceled the effects of surfactants added to the mixture [24].

Several study described that, in the presence of salts, the enhanced of mass transfer coefficient occurs because higher salt concentration increases surface tension with concomitant finer bubbles [6,21,25]. The salt does not apparently affect surface renewal nor does it block transport at the air–liquid interface thus, K_{La} will increase as the surface area to volume ratio increases.

Because only few information are reported about the effect of salts on oxygen mass transfer, this study is dedicated to an accurate evaluation of thermodynamic and kinetic aspects in the water oxygenation process in various synthetic liquid phases containing either salt (NaCl, KCl, LiCl). For this, the oxygenation has been analyzed by means of kinetic study of oxygen dissolution in order to obtain information about the oxygen mass transfer coefficient (K_{La}) and oxygen transfer rate SOTR, using a stirred, submerged aerated 4-liter system. The results obtained are correlated with the operational conditions of hydraulic head, air-flow and salinity of water using an optical oxygen sensor.

2. Experimental

The determination of oxygen diffusion coefficients was based on measurements of gas–liquid mass transfer coefficients (K_{La}) occurring at a free gas–liquid interface under controlled hydrodynamics conditions. For all the experiments, submerged aerated 4-liter system, equipped with Luminescent Dissolved Oxygen Probe (HACH, IntelliCAL™ LDO101) for the measure of water-dissolved oxygen, is used.

The designed-laboratory prototype described in Fig. 1 is composed by a vessel with a circular base of 70.84 cm^2 and 75 cm in height. The prototype aerator is equipped by a thermostated jacket for controlled temperature analysis, magnetic stirrer, compressed air and nitrogen from laboratory lines and with an air flowmeter to set different air-flows of 0.2, 0.3 and 0.4 (l/min). The temperature was monitored and maintained constant by using Haake F3 Thermostat.

The size of air bubbles was monitored photographically and, in order to obtain regular air bubbles, a particular porous septum with specific porosity of P3 (16–40 μm) (ISO 4793) was used.

To investigate the influence of salt concentration on the oxygen solubility, NaCl solutions at different concentrations were prepared utilizing NaCl 99% by Sigma-Aldrich and the salinity monitored by CyberScan PC 300 conductometer. For these measurements, a blank of deionized water, a clean tap water sample with salinity as 0.164 ppt (g/l), and NaCl water solutions with salinity of 4.1 and 8.26 ppt were used. A Metrohm655 pH meter with a combined electrode (Inlab 413) was used for pH determination.

For each sample, the influence of salt concentration on the oxygenation process has been investigated as function of height of the water column inside the reactor (hydraulic head: 62.2, 51.8, 41.5, 31.2 and 20.8 cm), at the lower air-flux of 0.2 l/min with a constant temperature of 23.5 °C. To obtain information related to the effects of different salts, LiCl, NaCl and KCl (from Sigma-Aldrich) solutions were prepared by dissolving the same amount of salts: 4.091, 8.151, 16.244 and 32.289 g/l , and the salinity of each sample was checked by CyberScan PC 300 conductometer; the temperature was maintained constant at 23.5 °C and the air-flux set to 0.4 l/min at constant hydraulic head of 41.5 cm.

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