

Evaluation of residence time distribution and mineralogical characterization of the biooxidation of sulfide minerals in a continuous stirred tank reactor

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ABSTRACT

The residence time distribution (RTD) of the liquid phase and the mineralogical characterization of the biooxidation of refractory gold mineral was studied in a continuous stirred tank reactor. The latter was achieved using a native mixed culture of acidophilic mesophiles. The RTD was assessed using a mathematical model of stirred tanks in parallel. The oxidation of sulfide and the phases generated through the biooxidation process were evaluated via X-ray diffraction (XRD). The results indicated that the experimental RTD fit to the model. The reactor has a high tendency to behave as a completely mixed reactor. However, the mixed flow inside the reactor has disturbances such as by-pass and dead zones. The estimated mean residence time for the model was approximately 36% greater than the theoretical residence time. It was probably caused by the delay in the outflow of the tracer due to gas hold-up, foaming at the top and the design of the reactor outlet structure. The XRD outcomes showed that the oxidation of arsenopyrite was greater than that of pyrite. Similarly, the formation of jarosite and brushite was observed. It was concluded that the dead zones could increase the probability of jarosite precipitations.

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1. Introduction

The biooxidation of sulfide minerals in continuous stirred tank reactors (CSTR) has become an important method for the pretreatment of refractory gold ore (Chandraprabha et al., 2002; Akcil, 2004; Watling, 2008). Industrial-scale biooxidation was first carried out in South Africa (Fairview) in 1986 as pretreatment for refractory gold concentrates. Currently, CSTR biooxidation is being used successfully in commercial operation in countries such as Brazil, Peru, Australia, Ghana, South Africa, India and China (Gonzalez et al., 2003; Akcil, 2004; Sand and Gehrke, 2006; Watling, 2008). The growth of this technology is promising, and new projects are currently in the research and engineering stages (Van Niekerk, 2009).

Biooxidation is catalyzed by bacteria that oxidize reduced iron and sulfur compounds. In industrial applications of mining and metallurgy, cultures of mesophiles or moderate thermophiles are widely used for the oxidation of sulfide ores (Rossi, 1990; Rawlings et al., 2003; Akcil, 2004; Sand and Gehrke, 2006). Such microorganisms include the iron- and sulfur-oxidizing *Acidithiobacillus ferrooxidans*, the sulfur-oxidizing *Acidithiobacillus thiooxidans* and *Acidithiobacillus caldus*, and the iron-oxidizing *Leptospirillum ferrooxidans* and *Leptospirillum ferriphilum*. (Coram and Rawlings, 2002; Rawlings et al., 2003; Bryan et al., 2011). The operations tend to be

dominated by *L. ferriphilum* and *A. caldus* at 40–45 °C (Rawlings, 2007). The success of bacterial sulfide oxidation in a CSTR requires a mixing that reaches a degree of uniformity that maintains suitable conditions for bacterial growth and solids suspension, thus minimizing the development of concentration, temperature and pH gradients within the reactor (Rossi, 1990; Hayward et al., 1997; González et al., 2003; Deveci, 2004).

Currently, the research and development areas related to biooxidation technology include the development of an improved agitation system, and the reduction of retention time (Van Niekerk, 2009). The scientific literature provides a number of studies on different impellers and their effects on bacterial activity in CSTR biooxidation (d'Hugues et al., 1997; Deveci, 2002, 2004; Liu et al., 2007; Sun et al., 2012). It has been demonstrated that within in the biooxidation process the best alternative is the use of axial flow impellers (Dew et al., 1997; Deveci, 2002, 2004; Gonzalez et al., 2003; Van Aswegen et al., 2007). Indeed, nowadays all commercial stirred-tank biooxidation plants use axial flow hydrofoil impellers (Dew et al., 1997; Arrascue and Van Niekerk, 2006; Batty and Rorke, 2006; Van Aswegen et al., 2007; Van Niekerk, 2009).

Hydrodynamic behavior studies in biooxidation reactors are limited in the literature. The mathematical description of residence time distribution (RTD) is usually expressed either through the dispersion model or through the tanks-in-series model. Romero et al. (1998) determined the RTD for two stirred tank reactors in series at the Rio Tinto plant in Spain. The RTD curve that best fit the experimental data was the two completely stirred tanks-in-series model.

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Nomenclature

C_i	tracer concentration in the effluent at time t_i (mg/L)	τ_1	mean residence time in the large stirred tank (h)
$E(t)$	Residence time distribution function	τ_2	mean residence time in each small stirred tank in series (h)
$E(\theta)$	Dimensionless residence time distribution function	τ_3	mean residence time of the three agitated tanks in series (h)
$E_m(\theta)$	residence time distribution of the model	f_q	flow fraction of the upper path
$E_e(\theta)$	experimental residence time distribution	f_{1-q}	flow fraction of the lower path
$G(s)$	transfer function of the stirred tanks	θ	dimensionless time (t_i/t_m)
t_i	time at which the sample is taken from the reactor effluent during the tracer test		
$(h) \cdot t_m$	experimental mean residence time (h)		
s	complex variable		

Mazuelos et al. (2002) reported the assessment of RTD at various bed heights of a packed bed reactor using the tanks-in-series model. The results indicated that for the maximum bed height of 84 cm, the hydrodynamic behavior fits two completely mixed tanks in series.

A hydrodynamic assessment of a reactor conducted by analyzing the residence time distribution (RTD) is crucial to determine possible flow abnormalities such as by-pass and dead zones which may affect the process operation and performance (Levenspiel, 1981; Andrade and Hodouin, 2005). Additionally, RTD can be used together with biooxidation kinetic models to simulate these processes (Brochot et al., 2004). The previous statement shows that characterizing the hydrodynamic behavior in biooxidation reactors is relevant.

Mixing conditions in stirred tank reactors for the biooxidation of gold concentrates depend on many factors including pulp density, aeration and agitation rate, reactor design features such as tank geometry, impeller type and diameter, and air injection type and location (Gonzalez et al., 2003; Deveci, 2004). It is therefore necessary to determine the RTD for each individual case and thus guarantee a good process performance. In a reactor, hydrodynamic evaluation is performed using a tracer test: a tracer is injected into the reactor's feed and the response to this stimulus is recorded. The response analysis provides information about the mixing properties of the system and the performance of non-ideal chemical reactors (Levenspiel, 1981).

The aim of this paper was to determine the RTD of the liquid phase of a continuous stirred tank reactor used for biooxidizing sulfides at laboratory scale. This was done in order to characterize the mixing conditions established by the operating conditions and reactor design. The RTD was analyzed through a developed mathematical model of completely stirred tanks in parallel. After determining the RTD the bacterial oxidation of sulfides was evaluated and a mineralogical characterization through X-ray diffraction (XRD) was performed to determine the level of sulfide oxidation and the phases generated in the biooxidation process, respectively.

2. Stirred tanks in parallel model

This model consists of three small perfectly mixed reactors in parallel with a combination of a large perfectly mixed reactor in series with two smaller reactors (Fig. 1).

A very useful concept for modeling the RTD of real reactors is the transformation of RTD into the Laplace domain (Pinheiro and Oliveira, 1998; Yianatos et al., 2005), because the Laplace transform is used to solve problems where time is the independent variable.

The Laplace transform of the residence time distribution, or the $E(t)$ -curve, that is, the transfer function of the mixing system, $G(s)$, is given by Eq. (1):

$$G(s) = \int_0^{\infty} E(t) \cdot e^{-st} \cdot dt \quad (1)$$

where s is a complex variable. The response of any array of ideal reactors can be derived from the standard complex algebraic domain rules, and then converted into its time domain expression (Levenspiel, 1981). The solution of Eq. (1) for each ideal continuous stirred tank in Fig. 1 is represented by a transfer function in the Laplace domain in terms of mean residence time of the liquid inside the reactor (τ) by Eq. (2) (Yianatos et al., 2005; Andrade and Hodouin, 2005):

$$G(s) = \frac{1}{\tau s + 1} \quad (2)$$

The overall transfer function for the arrangement of tanks in parallel in Fig. 1 is given by Eq. (3):

$$G(s) = (1 - f_q) \cdot G_1(s) \cdot G_2(s)^2 + f_q \cdot G_3(s)^3 \quad (3)$$

where f_q is the flow fraction circulating through the upper branch, f_{1-q} is the fraction of the flow circulating through the lower branch, $G_1(s)$ is the transfer function of the large stirred tank, $G_2(s)$ is the transfer function of each ideal small stirred tank in series with the large tank, and $G_3(s)$ is the transfer function of each of the three stirred tanks in series in the upper branch of the array in parallel.

By substituting Eq. (2) in each stirred tank in Eq. (3), the transfer function in terms of the parameters of the tanks-in-parallel is obtained as follows (Eq. (4)):

$$G(s) = \frac{(1 - f_q)}{(\tau_1 s + 1) \cdot (\tau_2 s + 1)^2} + \frac{f_q}{(\tau_3 s + 1)^3} \quad (4)$$

If Eq. (4) is decomposed into partial fractions, Eq. (5) is obtained.

$$G(s) = \frac{(1 - f_q) \cdot \tau_1}{(\tau_1 - \tau_2)^2} \cdot \left[\frac{1}{(s + \frac{1}{\tau_1})} - \frac{1}{(s + \frac{1}{\tau_2})} \right] - \frac{(1 - f_q)}{(\tau_1 - \tau_2) \cdot \tau_2} \cdot \left[\frac{1}{(s + \frac{1}{\tau_2})^2} \right] + \frac{f_q}{\tau_3^3 \cdot (s + \frac{1}{\tau_3})^3} \quad (5)$$

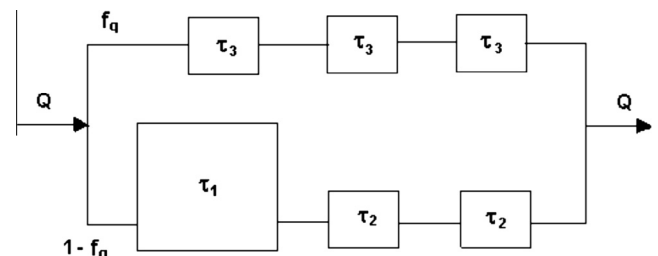


Fig. 1. Graphical representation of the stirred tanks in parallel model.

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