HYDRODYNAMIC STUDY OF COLUMN BIOLEACHING PROCESSES

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Abstract: The modelling of flow leaching solution through the porous media has been considered. The heap bioleaching process can be tested using the column experimental equipment. This equipment was employed to the hydrodynamic studies of copper ore bioleaching. The copper ore (black shale ore) with the support, inertial materials (glass small balls and polyethylene beads) was used to the bioleaching tests. The packed beds were various composition, the ore/support ratio was changed. The correlation between the bed porosity and bioleaching kinetics, and copper recovery was investigated.

Key words: heap bioleaching, packed bed porous media, bioleaching, *Acidithiobacillus ferrooxidans*, Ergun equation, support material

1. Introduction

The hydraulic properties of porous media are important in heap or dump bioleaching processes. There is necessary to improve understanding of heap bioleaching process to achieve more effective metal recovery. The leaching solution flow through a porous medium (heap) can be described using soil liquid hydrodynamic (CLEMENT *et al*., 1998) or chemical engineering theory (LI *et al*., 2001; HENDERSON *et al*., 2010; NEMEC and LEVER, 2005).

The two-dimensional model for a heap bioleaching of copper ore has been developed (CASAS *et al*., 1998). The main attention was focused on the gas transport through the heap. When the bed permeability was higher, air convection was the predominant mechanism. The Darcy’s law has been used to a fluid transport description in a porous medium. The leaching solution flow through a porous medium is the essence of leaching process. The leaching solution in the porous bed can move to the button of heap and stagnant attached to the pore walls. The bioleaching ore dissolution is occurred inside the stagnant part of solution (SHEIKHZADEH *et al*., 2005).

Understanding of hydrodynamic and transport phenomena in a porous packed bed is essential for the heap bioleaching. The liquid flow is considered to be governed by both the gravitation and capillary forces. A part of liquid is holdup between the adjacent void cells. The transport of microbial cell is mainly connected with the leaching solution migration through the porous media. Mathematical model of microbial cells transport in unsaturated porous media is described by a simplified form of advection-dispersion equation (TUFENKJI, 2007).

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$$\frac{\partial c}{\partial t} = D\frac{\partial^2 c}{\partial x^2} - v\frac{\partial c}{\partial x} - R \tag{1}$$

where c is the microbial cells concentration in the leaching solution at a distance x and time t. D is the hydrodynamic dispersion coefficient, and v is the interstitial microbe velocity. R is the retardation factor. The microbial cells retardation factor is controlled by the cell attach to the surface of particles and the release from this surface. This equation can be derived from basic mass balance principles. In Eq. 1, only hydrodynamic dispersion of microbial cells is considered. However, the overall transport process should take into consideration attachment and detachment of cells to the solid surface, and death of microorganisms.

The attachment and detachment processes can be described by using the classical colloid filtration theory (TUFENKJI and ELIMELECH, 2004). The bacteria attachment to the mineral surface depends on the potential energies of London-van der Waals attraction and electrical double layer repulsion. The extended Drejaguin-Landau-Verwey-Overbeek (DLVO) colloid stability theory can be used to quantitative predicting the condition to detach bacteria cell to the pore surface. The bacteria cells adhering to the pore surface of porous bed can be detached by the hydrodynamic forces. The immobilization of microbial cells is correlated with the biofilm creation on the pore surface. The adsorption of biopolymer on the surface gives the foundations for the biofilm creation. The data from column bioleaching experiments are presented that the effect of biofilm growth inside the porous media is important for leaching process (BERGENDAHL *et al*., 2000). A number of experimental studies have been conducted to prove the biofilm and microcolony model and they influence on bioleaching processes (ROCKHOLD *et al*., 2002; HERNANDEZ-LOPEZ *et al*., 2011).

Darcy's law is commonly used to describe one-dimensional liquid flow through uniform incompressible porous bed. Generally, Darcy's law is presented as follows:

$$\frac{1}{A}\frac{dv}{dt} = \varphi\frac{\varepsilon\Delta p}{\mu l} \tag{2}$$

where: v is the liquid velocity, A is the cross sectional area, φ is the permeability of porous bed, Δp is the pressure drop, μ is the liquid viscosity, l is the thickness of porous bed. For non-overlapped spherical particles, the permeability (ε) is related to the drag coefficient (C_d).

$$\varepsilon = \frac{v}{6\pi r C_d} \tag{3}$$

where: r is the sphere radius.

The permeability of the porous media (φ) is estimated using Carman-Kozeny's equation (HENDERSON *et al*., 2010). The Carman-Kozeny's equation can be written as:

$$\varphi = C_0\frac{\varepsilon^3}{M_b} \tag{4}$$

where: C_0 is a Kozeny's coefficient and M_b is the specific surface of the porous bed.

Leaching liquid flowing through porous abundance is wasting the energy. The viscous energy loss is connected with the liquid velocity and the inertial energy loss is proportional to the velocity squared.

$$\frac{\Delta p}{l} = a\nu + b\nu^2 \tag{5}$$

where: Δp is the pressure loss, l is the bed height, v is the liquid velocity (averaged over the flow cross-section), and a and b are tow empirical parameters (a = 150; b = 1.75). For the laminar flow Eq. 5 has a form:

$$\frac{\Delta P}{l} = \frac{150\mu}{\varphi^2 d_p^2}\nu + \frac{1.75\rho(1-\varepsilon)}{\varphi d_p \varepsilon^3}\nu^2 \tag{6}$$

where φd_p gives the diameter of the equivalent-volume sphere. The equation (6) is called "Ergun equation".

The objective of this study was to investigate the effect of hydrodynamic parameters of porous bed in a packed column bioreactor on the bioleaching process of black shale copper ore.

2. Materials and methods

2.1 Bacteria strain and growth condition

Column bioleaching experiments were conducted with *Acidithiobacillus ferrooxidans* strain. Bacteria strain was isolated from the acid mine drainage located at the old pyrite mine (Lower Silesia, Poland). Strain was grown in 250 ml Erlenmeyer flasks containing 150 ml of Silvermann-Lundgren medium (9K).

2.2 Ore and support materials

The black shale ore sample used throughout this study was provided by Lubin Concentrator (KGHM Polish Copper) as middling. Black shale is composed of clay-dolomite matter saturated with organic matter and with finely dispersed sulphide mineral particles. This material was a multi-mineral resource in which the copper sulphides were mainly chalcocite, covellite, bornite and chalcopyrite. In the flotation circuit of Lubin Concentrator, the black shale is partially concentrated at middlings (tailing from 1st cleaning flotation step). The average size of middling was 42.15 μm. According to XRD analysis (Table 1), the bioleaching material contains 2.5 % of copper in the form of sulphide.

Two different types of support particles have been employed. The support materials used in this study were glass small bolls and polyethylene beads. These materials have different surface properties. The surface of glass bolls is hydrophilic but the surface of polyethylene beads is hydrophobic. The pictures of support materials are presented at Fig. 1a and 1b.

Table 1. Chemical composition of shale sample

Elements	Unites	Concentrations
Cu	%	2.5
Fe	%	1.75
Ni	[g/t]	374
As	%	0.08
Ag	[g/t]	175
S_{total}	%	2.93
C_{total}	%	14.46

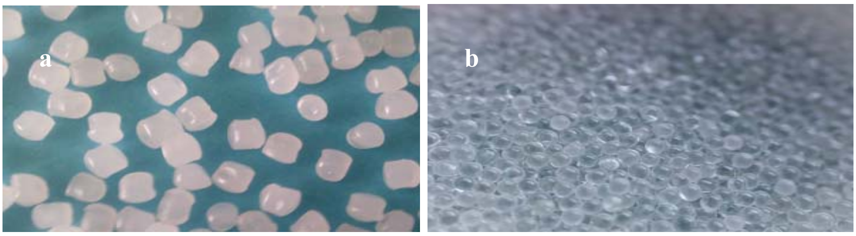

Fig. 1. Glass small bolls (a) and polyethylene beads (b).

Glass bolls have equal diameter 1.5 mm. The size of polyethylene beads was the range of 2 to 5 mm. The calculated surface area of this beads was 0, 08 m^2/g.

2.3 Column bioleaching experiments

Column leaching experiments were conducted in small column (37 cm/5.0 cm). The leaching solution was passed through the column by gravity. It was collected in the thermostatic bath and recycled to the top the column by peristaltic pump. The liquid layer was kept on the top of column. The bypass construction provides both a constant liquid layer and constant hydrostatic pressure. This liquid layer was aerated. The column was charged with 80 g of black shale material. At the next column experiments, the ratio black shale ore to support material was changed. The leaching solution (200 ml) was pumped from the feed container to the top of the column. The gas (air) was also introduced to the liquid layer at the top of column, where a layer of leaching solution was created during the leaching tests.

The particle size distribution has been obtained by powder analysis using a laser dyfractiometer Mastersizer 2000 (Malvern Instruments G.B).

During the experiments Fe^{2+}, Fe^{3+} ions and protein concentrations, as well as pH and Eh were measured. Ferrous iron concentration in solution was determined by spectrophometric methods. The concentration of copper was determined using atomic spectroscopy method. The bacterial population was controlled by protein analysis.

3. Results and discussion

The considerations regarding the optimum construction of heap and column packed bed are complex; therefore, an intensive study is needed. It is known, that the bed porosity increase the more if the shape of particles deviates from the spherical shape. In addition the particle size and packing procedure have a strong effect on the hydroprocessing conditions (NEMEC and LEVEC, 2005). When the ore particles were used, the porosity of packed bed was relatively narrow. In such case, the flow through packed bed can be regulated by the support addition.

Bioleaching in column with recirculation of the leaching liquid is a lab-scale simulation of heap bioleaching processes (LONG *et al*., 2004; LIZAMA *et al*., 2005). The bed permeability is an essential process parameter, in the case of column bioleaching. The particle size distribution of shale sample (Fig. 2) shows that this material contains a lot of fine particles. The amount of fine particles is a key parameter for the modelling of flow through packed bed. It was recommended that the fine fraction should be less than 10 % (LIN *et al*., 2004; 2005).

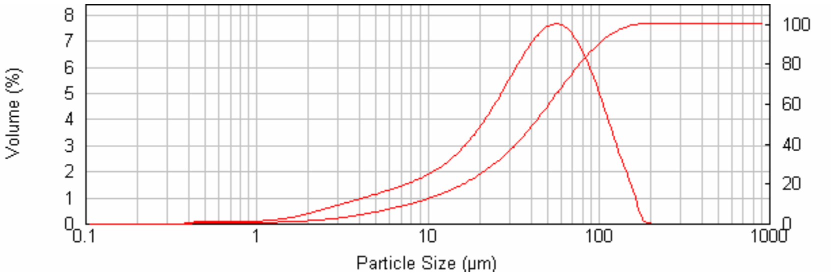

Fig. 2. Particle size distribution of black shale sample.

The present work shows the influence of two supports (glass small bolls and polyethylene beads) on the hydrodynamic parameter. To obtain a wide variety of bed porosity different quantity of support materials were employed. The leaching solution flow inside the heap or column can be divided into saturated and unsaturated.

As mentioned before, understanding of the liquid flow phenomena inside the packed bed is important to enhance the bioleaching performance. The milli-Ct scanner has been used to determine the pore structure of the packed column before and after bioleaching. The application of this sophisticated technique gives an opportunity to establish a fundamental relationship between pore microstructure and effective transport coefficient (LIN *et al*., 2004; 2005).

The velocity of liquid flowing through the porous bed (v) can be described by the following equation:

$$v = \varphi \varepsilon_g \left(h_1 - h_2 \right) \tag{7}$$

where: φ is a permeability of the porous bed, ε_g is the gas (air) holdup in the pores, h_1 is the upper level of liquid, and h_2 is the bottom of bed.

The porosity is an important parameter of porous bed at the macroscopic scale. It is defined as the pore volume of a representative sample divided by its bulk volume. The specific surface of a porous bed, here denoted as the bed surface, is defined as the interstitial surface area of the porous bed.

The porous bed can be treated as a bundle of capillary tubes of equal length. Hydraulic radius is defined for straight pipes as:

$$R_H = \frac{A_i}{P_i} \tag{8}$$

where: A_i is the cross-sectional flow area and P_i the wetted parameter (NIVEN, 2002).

A local Reynolds number is defined using as a scale length the square root of across-sectional area of the pore as follows:

$$R_e = \frac{\varphi A^{\frac{1}{2}} \nu}{\mu} \tag{9}$$

At lower local Reynolds number the flow of leaching solution is laminar. The local Reynolds number is usually less than one (SEDERMAN *et al*., 1998).

The experimental tests were conducted using a column reactor with a different composition of bed. The liquid flows were measured by such porous beds. The flow measurements were monitored at the time when the liquid flow did not change. This means that the deposit was porous in a natural way, unified, and sections of transport channels are not changed during the measurements. On the basis of data on the composition of the deposits, and the flow of liquid parameters calculated deposits such as porous: the degree of dispersibility deposits, deposits of hydraulic diameter and the number of Reynolds with a kind of liquid flow by deposit. The calculated data are shown in Tables 2 and 3.

Table 2. The flow of liquids through porous shale ore deposit (fixed) with a different amount of polyethylene beads fittings.

No.	**Polyethylene beads weight [g]**	**Black shale weight [g]**	**Liquid flow [ml/s]**	**Porous bed height [cm]**	**Liquid layer height [cm]**
1	0	80	0.050	5.6	15.0
2	15	80	0.034	6.4	13.2
3	30	80	0.036	8.0	13.0
4	50	80	0.020	9.0	10.0

Table 3. Porous bed characteristics.

No.	**Porous bed porosity**	**Calculated flow 10^{-3}**	**Surface area [m^2/g]**	**Particles medium size [μm]**	**Bed dispersion degree**	**Hydraulic diameter 10^{-3}**	**Reynolds number**
1	0.5542	3.98	4.128	34.143	3.414	64.9	0.3048
2	0.4298	2.71	3.4888	387.18	26.328	4.40	1.8401
3	0.4195	2.86	3.0240	643.86	46.358	2.606	3.1721
4	0.3387	1.59	2.5711	873.04	34.921	1.552	2.0991

At the next series of experience, the weight of porous deposits was chanded using a different quantity of shale ore and polyethylene beads. The data collected by such flows of deposits are shown in Table 4 and 5.

Table 4. The flow of liquids through porous deposit with a fixed mass of shale ore and polyethylene beads.

No.	Polyethylene beads weight [g]	Black shale weight [g]	Liquid flow [ml/s]	Porous bed height [cm]	Liquid layer height [cm]
1	0	80	0.050	5.6	15.0
2	15	65	0.049	6.0	13.6
3	30	50	0.307	6.5	14.0
4	50	30	0.857	8.5	8.8

Table 5. Porous bed characteristic.

No.	Porous bed porosity	Calculated flow 10^{-3}	Surface area $[m^2/g]$	Particles medium size [µm]	Bed dispersion	Hydraulic diameter 10^{-3}	Reynolds number
1	0.5542	3.98	4.128	34.143	3.41	64.9	0.3048
2	0.4748	3.90	3.369	453.36	37.94	4.905	3.3658
3	0.4386	24.44	2.61	872.59	425.8	2.725	39.987
4	0.3928	68.23	1.598	1 431.55	153.24	1.757	160.86

Similar experiences were carried out by replacing the polyethylene fittings by glass balls. The results obtained are presented in Table 6 and 7.

Table 6. The flow of liquids through porous deposit with a fixed mass (80 g) and various quantities of ingredients.

No.	Glass bolls weight [g]	Black shale Weight [g]	Liquid flow [ml/s]	Porous bed height [cm]	Liquid layer height [cm]
1	0	80	0.050	5.6	15.0
2	15	65	0.051	4.5	14.3
3	30	50	0.054	4.6	14.5
4	50	30	0.117	4.0	15.5

Table 7. Porous bed characteristic.

No.	Porous bed porosity	Calculated flow 10^{-3}	Surface area $[m^2/g]$	Particles medium size [µm]	Bed dispersion	Hydraulic diameter 10^{-3}	Reynolds number
1	0.5542	3.98	4.128	34.143	3.41	64.9	0.3048
2	0.4126	4.06	3.376	74.61	7.61	2212	0.5157
3	0.4276	4.30	2.625	115.09	12.43	14.86	0.8646
4	0.3451	9.31	1.623	169.05	39.56	8.165	2.405

The following conclusions canbe inferred from the data presented in tables: adding to the finely ground black shale ore polyethylene beads or small glass beads increases the degree of deposit dispersion. The systematic reduction of the hydraulic diameter of poroud bed isevident. Generelly, Reynolds number has a value not exceeding the value 10. It is supported the idea of a laminar flow inside the porous deposit. The exception is the flow of liquid on a large number of polyethylene beads in a porous bed (Table 5). For this case, the value of Reynolds are larger than 10 (40 and 160), which may suggest picking off like strug liquid from the surface of the beads. This phenomenon should be explained by the varying degrees of surface wettability of polyethylene surface and glass surface. Polyethylene surface is a surface with a low surface energy, and unlike to the glass surface is hydrophobic. The difference in degree of hydrophobicity of infill material will affect cell adhesion micro-organisms to these areas. This hypothesis will be tested in future experiments.

The changes in copper recovery values during 14 days of bioleaching experiments are shown at Fig. 3. At first, substantial increase of metal recovery was obtained when the supported materials were added to the column bead. The results presented at Fig. 3 indicate that the copper recovery was very similar to glass beads and polyethylene beads.

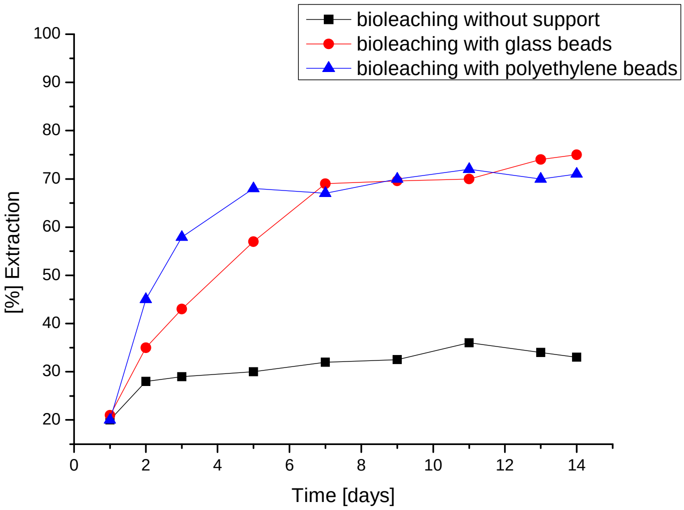

Fig. 3. Bioleaching behaviours of shale with and without supported materials (Copper recovery during bioleaching).

Before the bioleaching, the permeability was estimated to be on the order of 14.10^{-4} cm^2. This value was reduced to about 4.10^{-4} cm^2 after bioleaching.

4. Conclusions

In this paper, we have proposed a new approach to the column bioleaching of fine black shale ore. The development procedure was able to improve the copper recovery by adopting two support materials. These materials were glass small beads and polyethylene beads. It was shown that the addition of these materials to the porous bed, caused an improvement of hydrodynamic parameters. It is important to note that using the conventional leaching method it is not possible to obtain the copper recovery on the level of 35 %.

Nomenclature

A – the cross section area [m^2]
a – the empirical parameter (a = 150)

C_0 – the Kozeny's coefficient
C_d – the drag coefficient
c – microbial cells concentration [cells/volume]
D – the hydrodynamic dispersion coefficient
d_p – the diameter of particle [m]
h_1, h_2 – upper and button levels [m]
l – the bed height [cm]
M_b – the specific surface of the porous bed [m^2/g]
P_i – the wetted parameter
Δp – the pressure drop [Pa]
R_H – the hydraulic radius [m]
Re – the Reynold's number
r – the sphere radius [m]
t – time [s]
x – the distance [m]

Greek symbols

ε – the porosity
ε_g – the gas holdup
μ – the kinetic viscosity [m^2/s]
φ – the permeability of porous bed [m^2]

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