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# Treatment of gold mining effluent in pilot fixed bed sorption system



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## ARTICLE INFO

Article history: Received 2 December 2012 Received in revised form 7 October 2013 Accepted 21 October 2013 Available online 2 November 2013

Kevwords: Heavy metal Sorption Pilot system Coconut shell Iron-oxide-coated sand

## ABSTRACT

This paper studied the removal of heavy metals from gold mining effluent (GME) of the AngloGold Ashanti mine (Obuasi, Ghana) in continuous down-flow fixed bed columns using coconut shell and iron oxide-coated sand as sorbents operated at a temperature of 28  $\pm$  2 °C and a constant flow-rate of 150 mL min<sup>-1</sup>. The two-stage treatment system targeted the removal of copper and arsenic, but other heavy metals (iron, lead and zinc) present in the GME in very low concentrations were also removed, with removal efficiencies exceeding 98% for all metals in all the cases studied. A total of 14.8 m<sup>3</sup> of GME was treated in 1608 h before arsenic breakthrough occurred in the system. At that point, copper, iron, lead and zinc were still completely removed, leaving no traces of the metals in the treated effluent. Copper uptake amounted to 16.11 mg  $g^{-1}$ , which is 2.23 times higher than the value obtained in a single ion laboratory column study. Arsenic and iron uptake amounted to 12.68 and 5.46 mg  $g^{-1}$ , respectively. The study showed that the two-stage treatment configuration is an ideal system for the simultaneous removal of copper and arsenic from low concentration GME, in addition to other heavy metals present at low concentrations.

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#### 1. Introduction

Heavy metals are found in wastewater discharged from industries such as mining, metal processing, electroplating, textile, tannery and petroleum refinery (Izquierdo et al., 2010; Kumar et al., 2011; Martín-Lara et al., 2012; Oguz and Ersoy, 2010; Vilar et al., 2008). Heavy metals such as As, Cu, Pd, Zn, Fe, Cr, Ni and Hg are toxic (Kaczala et al., 2009; Liu et al., 2011; Saad et al., 2011) and therefore the discharge of untreated metal-laden effluent poses serious environmental challenges (Benavente et al., 2011: Mondal, 2009). Sorption using natural and agricultural materials has been proposed as a cost effective and environmentally friendly alternative to chemical treatment (Acheampong et al., 2010, 2011a; Fu and Wang, 2011; Mohan and Chander, 2006; Sousa et al., 2009).

Although there are many industrial processes using sorbents such as activated carbon, the development of biosorption processes is still mainly at the stage of laboratory studies in spite of enormous progress made over the last decade (Park et al., 2010). Most studies on continuous biosorption systems have focused mainly on the optimisation of operating conditions and breakthrough curves using synthetic metal solutions (Izquierdo et al., 2012; Kleinübing et al., 2011, 2012; Muhamad et al., 2010; Naja and Volesky, 2008; Ramírez Carmona et al., 2012; Salamatinia et al., 2010; Singh et al., 2011; Sousa et al., 2010). Many of these studies have limited industrial application because industrial effluents are more complex, containing several metal ions and other contaminants (Singh et al., 2012). Singh et al. (2012) indicated that treatment of industrial effluent in continuous flow through systems allows the use of sorbents for multiple sorption and desorption applications. In this way, process sustainability is achieved through a substantial reduction in the fresh sorbent requirement, operational cost and solid waste materials needing disposal or containment (Kumar et al., 2011: Park et al., 2010). Little effort has been made on heavy metal removal from industrial effluents using agricultural and natural materials in continuous systems. For this reason, more research is needed at the pilot scale using real wastewater to demonstrate the applicability of a biosorption process to treat this type of wastewaters at the industrial scale.

The present work is a continuation of our previous studies, which showed the ability of coconut shell to sorb Cu(II) under batch (Acheampong et al., 2011a) and continuous flow through (Acheampong et al., 2013a) conditions, and its reusability during multiple sorption and desorption applications (Acheampong et al., in press). The SEM-EDX characterisation of coconut shell before and after sorption showed irregular surface texture of the biosorbent and the presence of K<sup>+</sup> and Mg<sup>2+</sup> that were replaced by copper during sorption, while the FTIR analysis suggested the involvement of amino, amide, carboxylic, hydroxyl and carbonyl groups in metal binding (Acheampong et al., 2011a). Kinetic studies indicated that intraparticle diffusion controls the rate of sorption in the coconut shell biosorption of Cu(II); however film diffusion cannot

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<sup>0304-386</sup>X/\$ - see front matter © 2013 Elsevier B.V. All rights reserved. http://dx.doi.org/10.1016/j.hydromet.2013.10.013

be neglected, especially at the initial stage of sorption (Acheampong et al., 2011b).

In our previous work, the biosorption of Cu(II) onto coconut shell was done at the laboratory scale with synthetic wastewater. This study evaluates the sorption efficiency of coconut shell (CS) and iron oxide coated sand (IOCS) for, respectively, copper (Cu) and arsenic (As) in gold mining effluent in a two-stage pilot fixed bed column continuous flow system operated from December 2011 to June 2012 with real GME (Temperature range: 25.7–27.6 °C, pH range: 7.6–8.1) from the AngloGold Ashanti mine (Obuasi, Ghana). Though the focus of this work was on the removal of Cu and As, the removal of other metals present in the gold mining effluent was considered as well.

### 2. Materials and methods

#### 2.1. Sorbent preparation

The coconut shell (CS) used in this study was obtained from Kumasi (Ghana) and its Cu(II) biosorption properties were described in detail by Acheampong et al. (2011a). The material was washed with distilled water and dried at 105 °C for 24 h. The coconut shells were ground using a Peppink hammer mill. The biosorbent was then sieved to obtain the 0.5-1.4 mm fraction used throughout this study. The iron oxidecoated sand (IOCS) was obtained from the water treatment plant at Zwolle Engelse Werk (The Netherlands) as a spent product of the sand filtration process. The IOCS was used as received; with a particle size range of 1.0-3.0 mm. Coating analysis performed on the IOCS shows the presence of Mn (19.4 mg g<sup>-1</sup>), Mg (0.81 mg g<sup>-1</sup>), Ca (3.33 mg g<sup>-1</sup>), Fe (151.85 mg g<sup>-1</sup>) and Si (2.43 mg g<sup>-1</sup>), among other constituents. In handling the IOCS before and during the experiments, we were careful not to destroy the iron oxide coatings on the sand particles. The physical characteristics of both sorbents used are presented in Table 1. SEM-EDX, FTIR and point of zero charge (PZC) analysis of the coconut shell prior to and after Cu(II) sorption were presented in Acheampong et al. (2011a).

#### 2.2. Gold mining effluent

The GME (Table 2) was taken from the tailings pond at a location described in Acheampong et al. (2011a, 2011b). The effluent was pumped into a 2500L PVC storage tank (Polytank, GH-Pt2500) located at the tailings pond site. The tank was first cleaned thoroughly using a detergent, 10% HNO<sub>3</sub>, triple rinsed with distilled water, and finally triple rinsed with the GME. The effluent was kept in the storage tank over night to allow solid particles to settle at the bottom and the effluent to equilibrate. The supernatant was discharged by gravity into two 100 L plastic tanks (720011 Getrankesfass 100 L) and transported to the laboratory, located at 25 km from the tailings pond, where the pilot plant was installed. The pH and the temperature of the effluent were checked prior to being fed to the sorption columns, but no pH adjustment was made. Table 2 shows the characteristics of the GME used for the experimental runs. Experimental runs I, II and III were conducted consecutively with GME collected on different dates from the same location but with different characteristics: the GME temperature, pH and metals concentrations were different for each run (Table 2). All GME samples were analysed three times and the values reported as the mean ( $\pm$  standard deviation).

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Physical characterisation of the sorbents used in this study (Acheampong et al., in press).

Sorbent	Total pore volume	Porosity	Bulk density	Surface area ( $S_{BET}$ )
	(cm <sup>3</sup> g <sup>-1</sup> )	(%)	(g cm <sup>-3</sup> )	( $m^2 g^{-1}$ )
CS	0.900	55	1.35	0.4
IOCS	0.012	51	2.38	11.4

Table 2	2
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Parameters	Run I	Run II	Run III	Ghana EPA standard
Temperature (°C) pH Copper (mg $L^{-1}$ ) Arsenic (mg $L^{-1}$ ) Lead (mg $L^{-1}$ ) Zinc (mg $L^{-1}$ )	$\begin{array}{c} 26.1 (\pm 0.00) \\ 7.6 (\pm 0.02) \\ 5.67 (\pm 0.03) \\ 8.81 (\pm 0.01) \\ 0.03 (\pm 0.00) \\ 0.01 (\pm 0.00) \end{array}$	$\begin{array}{c} 25.7 (\pm 0.02) \\ 7.7 (\pm 0.01) \\ 6.21 (\pm 0.01) \\ 8.72 (\pm 0.02) \\ 0.05 (\pm 0.00) \\ 0.02 (\pm 0.00) \end{array}$	$\begin{array}{c} 27.6 (\pm 0.01) \\ 8.1 (\pm 0.01) \\ 7.98 (\pm 0.01) \\ 12.11 (\pm 0.03) \\ 0.07 (\pm 0.01) \\ 0.01 (\pm 0.00) \end{array}$	40.0 6.0-9.0 1.00 0.20 0.10 2.00
Iron (mg L <sup>-1</sup> ) Cyanide (mg L <sup>-1</sup> )	$1.92 (\pm 0.01)$ $1.56 (\pm 0.03)$	$0.78 (\pm 0.01)$ $3.64 (\pm 0.04)$	$2.16 (\pm 0.02)$ $2.58 (\pm 0.01)$	2.00 0.20

#### 2.3. Pilot plant set-up

The experimental set-up (Fig. 1) was constructed out of two identical Perspex columns (each of 10 cm inner diameter and 200 cm long). Column I was packed with CS (particle size 0.5–1.4 mm) to give a total effective bed depth of 150 cm (5.17 kg of sorbent), while column II was packed with IOCS (particle size 1.0–3.0 mm) to give a total effective bed depth of 150 cm (10.06 kg sorbent). Each column used in this study had a packing volume of 11.8 L. The sorbent in each of the columns was placed in between a layer of glass beads at the top and a thin layer of sieve at the bottom. The sieve was to prevent sorbent particles from falling through the effluent outlet. Packing was done under wet conditions to prevent trapping of air within the bed. Table 3 shows the pilot plant operational parameters. Prior to the start of the experimental run, distilled water was passed through each of the columns until a clear liquid was obtained at the outlet. This was done to remove powdered sorbent particles from the columns.

The heavy metal removal efficiency was studied by pumping the GME from a 200 L plastic tank (L4506 Getrankesfass 200 L) into the two columns in series using a pump (Masterflex model 77521-57, Range: 1-100 rpm) equipped with two pump heads (Masterflex Easy-Load II, model 77201-60). Piping was done with PVC rubber tubing (Masterflex L/S 17 TYGON Tubing, Inner diameter = 6.4 mm, Outer diameter = 9.6 mm) with a flow range of 5–250 mL min<sup>-1</sup>. The flow-rates were measured with a flow metre (OMEGA Direct Read Rotameter, model FLDW3210G, Maximum Flow Rate 250 mL min<sup>-1</sup>) placed between the pump and each column (Fig. 1). For regulating the liquid flow from the storage tanks, control valves were installed at the outlet of each storage tank as shown in Fig. 1. Metal ion concentrations at the inlet of column I were measured prior to each run. Samples were collected from the outlet of each column twice daily for the first 5 days and then once a day for the remaining of the run time for metal ion analysis with AAS. Metal concentrations in the feed were checked regularly for fluctuations during the experiments; no significant variations were observed.

#### 2.4. Experimental design

The pilot plant columns were operated at constant flow rate  $(150 \text{ ml min}^{-1})$  and fixed bed height (150 cm) during each run. Three experimental runs (I, II and III) were conducted with GME having different characteristics (Table 2). All the GME were collected from the same intake point but at different times. The reasons for running three different experiments with effluent having varied characteristics are to study the treatment plant response to fluctuations in metal concentrations as well as to verify the reproducibility of the results. All continuous operations with the pilot columns were performed under downflow mode. The plant was operated until the dynamic transfer zone reached the end of the column (Fig. 2). Until that time the effluent leaving the column had no trace of the sorbate in it. When the transfer zone reaches the column end, the sorbate concentration in the effluent starts to gradually increase and, for practical purposes, the working life of the column is over; the "breakthrough point" is reached, marking the usable

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