



Determination of the equilibrium and film diffusion constants of the platinum cyanide anions during the elution from activated carbon



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ABSTRACT

Various leach approaches have been developed for the alkaline cyanide leaching of platinum group metals (PGMs) from ores, concentrates and from secondary wastes such as spent automobile catalytic converters. Recently, the adsorption and elution of PGM cyanide complexes onto activated carbon has also been demonstrated and is similar to the activated carbon process for gold. The stripping of adsorbed tetracyanoplatinate(II) ($[\text{Pt}(\text{CN})_4]^{2-}$) from activated carbon consists of a two-step batch process which involves the pre-treatment of the metal-loaded activated carbon with a relatively strong sodium cyanide and sodium hydroxide solution prior to the elution step with de-ionized water at 80 °C. This paper models the elution and describes the mechanism of platinum elution from activated carbon. The equilibrium and film transfer constants were determined by fitting the model to experimental results and then validated by comparing to additional experiments. It was found that the rate of release of the platinum ions is governed by the amount of platinum and sodium on the activated carbon and the concentrations of these ions in the bulk of the liquid. It was found that the adsorption-elution of the single component system can be adequately described by using a modified Freundlich isotherm and a mass transfer diffusion equation. The rate of platinum elution interchangeably depends on the equilibrium of the Pt ions at the carbon-liquid interface and the mass transfer of these Pt ions from the interface to the bulk liquid. As both of these rate limiting factors were found to depend on the sodium concentration, the dominant platinum elution rate limiting factor shifts as the sodium concentrations change as the elution progresses. The elution process could be modelled accurately and it is shown that the second step (elution) of the two-step batch process (pre-treatment and elution) can be broken down into a sequence of four consecutive stages. The benefits of fundamentally understanding this process can ultimately lead to improved elution, better process control, shorter elution times, smaller elution columns or assist in the development of a continuous elution process.

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1. Introduction

Numerous direct leach technologies, including a number of cyanide-based leach processes has been reviewed by Mpinga et al. (2015). The leaching of Platinum Group Metals (PGMs) with cyanide, in a manner similar to that used for gold extraction, has been investigated as an alternative PGM process option that is less energy intensive and better suited to the extraction of complex, lower grade ores (Mwase et al., 2012a,b, 2014a,b; Eksteen et al., 2012, 2014). After showing that direct cementation using Merrill-Crowe technology was unsuitable for PGMs (Mpinga et al., 2014b), carbon adsorption and consequently elution

(Snyders et al., 2012, 2013, 2014a,b; Mpinga et al., 2014a) and ion exchange (Schoeman et al., 2012) were considered as technologies for recovery and concentration upgrading from solution. Two of the major steps of the process, i.e. the adsorption of the metal cyanide complexes onto activated carbon and the elution of these complexes into a more concentrated stream, were investigated and the effects of operating variables on process performance were determined (Mpinga et al., 2014a; Snyders et al., 2013). Successful elution of the metal complexes from the adsorbent into a sufficiently concentrated solution is critical in the development of this process and it was indeed only following the development of a suitable elution process that the now universal carbon in pulp process for gold recovery became commercially viable.

In a previous paper by the authors (Snyders et al., 2013) adsorption experiments were performed on a pregnant alkaline leach

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Nomenclature

A	parameter in Freundlich isotherm expression	N	mass transfer ($\text{kg}/\text{m}^3 \text{ s}$)
B	parameter in mass flux equation and modified from the Freundlich isotherm expression	p	static pressure (Pa)
BMs	Base Metals (Copper, Nickel, Cobalt...)	PGMs	Precious Group Metals (Platinum, Palladium, Gold)
BV	bed volume (m^3)	t	time (s)
C	liquid phase concentration (mg/L)	T	temperature ($^{\circ}\text{C}$)
C_L	bulk liquid phase concentration (mg/L)	R	gas constant (J/mol K)
C_2	inertial resistance factor	V	superficial velocity (m/s)
D	mass diffusivity (m^2/s)	Y	mass fraction
F	external force vector (N)	α	permeability
g	gravitational acceleration (m/s^2)	Ω	computational domain
i	specie (subscript)	ε	porosity
J	diffusion flux ($\text{kg}/\text{m}^2 \text{ s}$)	ρ	density (kg/m^3)
k	film transfer coefficient (m/s)	τ	stress tensor (Pa)
K	mass transfer coefficient ($\text{kg}/\text{m}^3 \text{ s}$)	μ	dynamic viscosity ($\text{N s}/\text{m}^2$)
L	length (mm)	∇	del operator which represents the partial derivative of a quantity with respect to all directions in the chosen coordinate system
Q	concentration of adsorbent on carbon surface (mg/kg)		
m	mass (kg)		
n	parameter in Freundlich isotherm expression		

solution (0.15 mg/L Pt, 0.38 mg/L Pd, 0.1 mg/L Au) originating from a cyanide extraction performed in a column leach test. Initial adsorption rates of Pt, Pd and Au were found to be fast with recoveries at approximately 90 percent after 2 h. After 72 h, the recoveries were observed to be 100%, 97.4% and 99.9% respectively. The parameters that influence the extraction of PGMs and Au were also examined. It has also been shown in the same paper that through an analogue of the AARL process, platinum and palladium cyanide were found to elute from activated carbon almost to completion in 5–6 bed volumes at 80 °C while the elution of gold cyanide at this temperature is poor (50% elution after 15 bed volumes). The process involves the pre-treatment of the metal-loaded activated carbon with a relatively strong sodium cyanide and sodium hydroxide solution prior to elution with de-ionized water. For effective gold cyanide elution, higher temperatures (between 90 and 130 °C) and pressures (between 200 and 300 kPa) are necessary. Although effective elution of Pt and Pd can be achieved at lower temperatures, several similarities to the well-researched (Adams, 1991; Davidson and Duncanson, 1977; Davidson et al., 1979; Van Der Merwe, 1991; Van Deventer and Van Der Merwe, 1994) gold elution were found. As with aurocyanide elution, a shift towards faster and more effective elution occurs as the temperature of the de-ionized water is increased. A higher cyanide concentration in the pre-treatment step results in more efficient elution up to a point, beyond which the elution efficiency starts decreasing again. This is related to the increase in the ionic strength of the eluant, which has also been found to have a negative impact on platinum and palladium elution (Eksteen et al., 2012, 2014). The aim of the current paper is to study these existing Pt elution profiles and to determine the appropriate rate constant involved.

2. Gold elution modelling

Since no fundamental studies regarding the elution of platinum cyanide from activated carbon exist in the open literature, the AARL process, and to a lesser extent the Zadra elution process, for gold were studied as a basis for the modelling of the platinum elution process.

Adams and Nicol (1986) modelled the Zadra elution process and assumed the gold cyanide diffuses to an interface where

equilibrium with the liquid phase exists. From the interface the gold then diffuses through a liquid film into the bulk of the liquid. A linear isotherm was used to describe the equilibrium which was later shown by Adams (1990) to be only applicable for very short columns and high flow rates. The intra-particle diffusion was described by a single average mass transfer coefficient.

A model of the AARL process was developed by Van der Merwe and Van Deventer (1990), who used the same fundamental model to describe the AARL and the Zadra elution processes. This model is similar to the adsorption model developed by Van Deventer (1984) and incorporates a macro pore mass balance equation, a micro pore mass balance, a mass balance at the external particle surface, a mass balance over the interparticle solution and a Freundlich isotherm (Eq. (1)) to describe the equilibrium.

$$Q = A \cdot C^n \quad (1)$$

Importantly, A was found to change as conditions during the elution process change and was subsequently written as a function of temperature, pH, cyanide concentration and individual cation (K^+ , Na^+) concentration. From this model, Van der Merwe and Van Deventer (1990) concluded that the shape of the gold elution profile for an AARL process is a function mainly dependent on the removal of the cations that had been adsorbed during the pre-treatment stage. The dependence of A on the system conditions is not an isolated case. Woollacot and Nino de Guzman (1993) also found A to be depended on oxygen and cyanide concentration in gold cyanide adsorption systems while Özer et al. (1999) stated that A is a function of the energy of adsorption and temperature and a measure of adsorption capacity while n represents adsorption intensity.

Stange (1991) simulated the elution process as a binary system, containing Na^+ and aurocyanide and described the rate of desorption as a rate expression with kinetic and equilibrium parameters and assumed that film transfer is the rate limiting process for both Na^+ and Au. With a constant value (time invariant value at all points in the column during an elution) for the gold mass transfer parameter and the equilibrium parameter for gold assumed to be a function of the sodium solution tenor, an acceptable fit with a set of industrial elution data was obtained. When the predictive nature of the model was tested, however, it was found to be rather insensitive towards an increase in the sodium ion concentrations

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