



Adsorption behaviour of copper and gold glycinate complexes in alkaline media onto activated carbon. Part 1: Isotherms



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ARTICLE INFO

Keywords:

Gold
Adsorption
Activated carbon
Glycine solutions

ABSTRACT

Alkaline glycine/glycinate solutions have been shown to have significant potential in the leaching of gold and copper from ores, concentrates, and wastes bearing these metals. An attempt was made to understand the adsorption behaviour of the gold glycinate complex onto activated carbon in the presence of copper for a pure alkaline glycine/glycinate system, after the gold has been dissolved using hydrogen peroxide as an oxidant and caustic soda as pH modifier. The adsorptive behaviour under various process conditions was evaluated using equilibrium loading isotherms. The linear regression (R^2) values for the Freundlich isotherm for all tested parameters are close to unity, suggesting the model to be well suited for gold glycinate adsorption onto activated carbon. The maximum adsorption capacity of the gold complex on the activated carbon was determined as a function of free glycine concentration, solution pH, initial gold and copper concentration and calcium chloride salt concentration using synthetic solutions. The adsorption capacity increased with increasing free glycine, copper and calcium chloride concentrations, but decreased with increasing solution pH and initial gold concentration. This information can be used (with adsorption kinetics) to design and evaluate Carbon-in-Pulp (CIP), Carbon-in-Leach (CIL) and Carbon-in-Column (CIC) circuits for gold when copper may be present.

1. Introduction

The increased interest in non-cyanide technology for gold ore processing can be attributed to a number of reasons. From an environmental perspective, cyanide is toxic, and the adverse environmental impacts resulting from its discharge have led to stringent jurisdiction regarding its usage (Eisler et al., 1999; Jeffrey et al., 2002). In some parts of the world, the use of cyanide for gold mining is prohibited while in other places approval for any gold cyanidation project is proving to be extremely difficult (Mudder and Botz, 2004). Geologically, due to the growing demand for gold, exploitation is shifting from cyanide-amenable ores to increasingly refractory or polymetallic ores, increasing the need for alternative processing routes (Hiskey and Atluri, 1988; La Brooy et al., 1994; Breuer et al., 2005; Dai et al., 2012; Jeffrey et al., 2002). Ammonia, thiocyanate, thiourea and thiosulfate are some of the alternative lixiviants studied thus far (Pyper, 1981; Aylmore and Muir, 2001; Grosse et al., 2003; Molleman and Dreisinger, 2002; Rezai and Peikary, 2002; Muir and Aylmore, 2004). More recently, the ability of glycine to leach gold and gold-copper ores was shown by a number of researchers (Perea and Restrepo, 2018; Eksteen and Oraby, 2015;

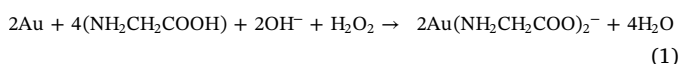
Oraby and Eksteen, 2014; Eksteen et al., 2017a,b; Tanda et al., 2017; Barton et al., 2018). As a leaching agent, glycine has environmental, technical and economic advantages over cyanide. It is environmentally benign and stable, yet easily biologically destructible under neutral and mild acidic conditions, but quite stable compared to sodium cyanide under alkaline conditions. From an economic perspective, apart from it being cheaper than cyanide (the bulk cost of glycine and NaCN respectively is around USD 1000–1800 per tonne (FOB, Feed Grade glycine) versus USD 2000 - USD2500 per tonne (FOB, Industrial grade sodium cyanide, 98%)) at the time of writing and compared to many other alternative plausible lixiviants, glycine can easily be recovered, regenerated and reused. Cyanide tends to convert to cyanate, thiocyanate, volatile HCN, and ferrocyanide, making recovery and reuse uneconomical in many cases. To reduce cyanide consumption of oxide gold-copper ores, these ores are conventionally first acid leached prior to neutralisation and cyanidation. However, being an alkaline leach, glycine leaching of gold ores is less susceptible to acid consuming gangue and co-leaching of Fe, Mn, Mg and Al (Eksteen and Oraby, 2015; Tanda et al., 2017). Glycine therefore allows the leaching of copper and gold at the same pH.

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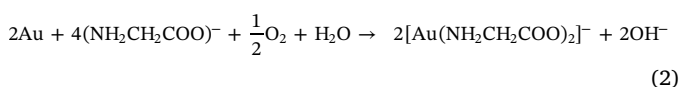
E-mail address: jacques.eksteen@curtin.edu.au (J.J. Eksteen).

The alkaline glycine system is characterised by simple chemistry (i.e. not prone to the side reactions as cyanide). Glycine, also known as aminoethanoic/aminoacetic acid, is the simplest neutral amino acid made up of one amine (–NH) and one carboxyl (COOH[–]) functional groups. It can, depending on solution pH, exist as either a glycinium cation (⁺H₃NCH₂COOH), a glycinate anion (H₂NCH₂COO[–]) or zwitterion (⁺H₃NCH₂COO[–]). In the glycine leach system, owing to the alkaline nature of the leach, at pH 10.5–11, the most predominant specie is the glycinate anion (Rega et al., 1998). The leaching process involves metal dissolution in a neutral to alkaline heated (23–60 °C) glycine solution in the presence of a soluble oxidant (Eksteen and Oraby, 2015). The oxidants successfully employed in previous studies include dilute hydrogen peroxide (Eq. (1)) and dissolved oxygen (Eq. (2)). The possible mechanism of gold dissolution in glycine has been proposed as:

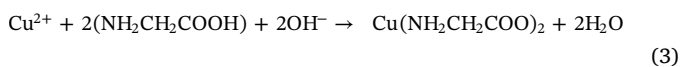
a) Using hydrogen peroxide as oxidant



b) Using oxygen as oxidant



The leaching behaviour of various copper oxide minerals in alkaline glycine systems has also been studied and is reasonably well understood. Results from studies indicate that, at optimum conditions (pH 11, glycine to copper molar ratio of 8:1), the copper extraction was over 80.0% for azurite, malachite and cuprite and < 20.0% for chrysocolla in 24 h. The copper complexing mechanism is shown in Eq. (3) (Tanda et al., 2017):



Methods for gold recovery from resultant polymetallic leachates include precipitation, ion exchange, solvent extraction and adsorption. Preliminary studies have confirmed that activated carbon adsorb gold glycinate complexes extremely well, possibly better compared to gold cyanide (Oraby and Eksteen, 2015). NaSH precipitation and solvent extraction were exploited for copper glycinate recovery and it was found out that it was possible to selectively recover over 99% copper using either method (Eksteen et al., 2016). Based on these results, activated carbon adsorption, currently being the main technique applied in cyanide systems for recovery of gold from cyanide leach liquors, is the starting point of our consideration for the newly developed glycine system. Industrially, it is preferred over other methods since it is cost effective, simple and has a high removal capacity owing to the large surface area of the carbon (Wan and Miller, 1990; Tu et al., 2011). Metals recovery from glycine leachates is still at an early stage and to date, fundamental understanding concerning the adsorption behaviour of metal glycinate complexes on activated carbon under different process conditions is lacking.

The interaction between an adsorbate and an adsorbent for any new system is best understood through the use of adsorption equilibrium information. Adsorption isotherms, their proper understanding and interpretation, are critical in the effective design and optimisation of any adsorption system. The Freundlich and Langmuir adsorption isotherms have been used to describe gold cyanide adsorption onto activated carbon from synthetic solutions with the former giving a much more accurate description of the equilibrium gold adsorption behaviour (Davidson, 1974; Cho et al., 1979). Previous studies have shown that these isotherms are well-suited for low concentration systems (Dixon

et al., 1976; Van Deventer, 1986; Woollacott, 1990) as encountered in most gold leaching operations from its ores. This is usually the case in the carbon-in-pulp (CIP) and the carbon-in-leach (CIL) processes, the two most dominant downstream processes used for gold extraction after cyanide leaching.

The main objective of the current study was to model the adsorption behaviour of metals onto activated carbon from a pure glycine system by generating isotherm parameters. The effects of pH, glycine concentration, ionic strength, initial gold and copper concentrations on the competitive loading of gold and copper glycinate complexes onto activated carbon are evaluated below.

2. Material and methods

2.1. Sorbent characterisation

The adsorbent used in this work was fresh activated carbon: PICA-GOLD® G210AS produced by Jacobi Carbons. The sorbent was manufactured from raw high grade coconut shell as per specifications from the manufacturer. Prior to use, the activated carbon was pulverised, mechanically sieved to –45 µm, oven dried at 70 °C for 16 h and preserved at ambient temperature in a desiccator. The surface area of the carbon as determined by the N₂BET method was 840.6881 ± 46.5496 m²/g. The BET surface area of the carbon enables the loading capacity to be presented as grams of gold per metre squared of carbon (g/m²). The activated carbon is pulverised to reduce kinetic effects and allow the carbon to reach equilibrium as quickly as possible. The resultant isotherm results can then be used to calculate the theoretical amount of granular activated carbon needed to meet the treatment objectives industrially.

2.2. Adsorbates

All experiments were carried out using synthetic solutions prepared from Millipore water and analytical grade reagents. The gold glycinate solution was prepared by leaching a 24 ct (99.99%) gold sheet (10 cm × 2.5 cm × 0.03 cm) supplied by A&E Metals in an alkaline glycine solution. Gold dissolution was done in a glass beaker which contained 15 mL hydrogen peroxide (30% w/v, Rowe Scientific), 15 g/L glycine (> 99%, Sigma-Aldrich) dissolved in 500 mL of distilled water and buffered to a pH of 11 by adding NaOH (ThermoFisher Scientific). The alkaline solution was heated to 60 °C and stirred at 400 rpm using Teflon coated magnetic stirrer bars. The beaker was covered to prevent evaporation. Copper was added to the prepared gold glycinate solution as CuSO₄·5H₂O (Asia Pacific Specialty Chemicals).

2.3. Adsorption experiments

The traditional bottle-on-rolls method was employed for the metal glycine complexes adsorption onto activated carbon. The experimental runs were carried out in sealed 2.5 L Winchester bottles (OD 44 cm and L 30 cm) at room temperature (approx. 23 °C). The development of equilibrium isotherms was effected by contacting weighted samples of 0.040, 0.128, 0.256, 0.404 and 0.640 g/L powdered activated carbon with 250 mL of gold and copper synthetic solution. The initial pH of the solutions was adjusted to 11 and controlled during the experiments by using NaOH or H₂SO₄. The mixture was bottle rolled for 24 h at 105 rpm. Preliminary tests indicated that 24 h was sufficient to reach equilibrium conditions. When equilibrium was reached, samples were filtered using a 0.22 µm FilterBio® CA syringe filter to remove any present carbon fines and prevent further adsorption. Total metal concentration in solution was determined by atomic adsorption spectrophotometry by using an Agilent 55B AAS.

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