

## Selective separation of gallium from zinc using flotation: Effect of solution pH value and the separation mechanism



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### ABSTRACT

Zinc is one of the impure elements in acidic solutions obtained from gallium sources; therefore, flotation studies were performed to investigate the removal of Ga(III) from Zn(II) using sodium dodecyl sulfate (SDS) as an anionic surface-active agent. In this study, the effect of different pH values on the percentage removal of Ga(III) and Zn(II) was studied. It was observed that the distribution of hydrolysis species and change in pH, had a significant effect upon the separation of Ga(III) from Zn(II). Also, the selective separation of Ga(III) was dependent on the recovery of water from the froth. The results showed that the optimal selective separation could be obtained at low pH values when the species of Ga<sup>3+</sup> was predominant and the precipitation of Ga(III) was at a minimum. The effect of pH on the characterization of the sublate (insoluble complexes formed in scum layer) and the complexation of SDS with hydrolysis species were studied by scanning electron microscopy coupled with energy dispersive X-ray (SEM/EDX) spectroscopy and Fourier transform infrared (FTIR) spectroscopy. In order to obtain more information about the mechanism of selective separation, the interaction of Ga(III) and Zn(II) with SDS was investigated using turbidity technique.

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### 1. Introduction

Gallium is in group 3A of the periodic table and a trace-metallic element in the Earth's crust (approximately 16.9 ppm), which was discovered by Lecoq de Boisbaudran in 1875 (Moskalyk, 2003). This element has wide application in the electronic industry. Gallium combined with arsenic [gallium arsenide (GaAs)] is used as a semiconductor in photovoltaic cells, super computers, light emitting diodes (LED), microwave transceivers, DVDs, solar cells, and other electronic devices (Font et al., 2007; Zhao et al., 2012). Other applications of gallium are in medicine, for example, the <sup>67</sup>Ga isotope is used as a diagnostic tool for detecting tumor tissues (Ahmed et al., 2013). Gallium does not exist as a concentrated deposit in nature and is usually present in very small concentration in ores of other metals. It is mainly obtained as a byproduct from Bayer solutions of aluminum and from acidic solutions in the manufacture of zinc (Flamini et al., 2007; Xu et al., 2007; Dumortier et al., 2005). Other sources of gallium are coal fly ash (Fang and Gesser, 1996; Gutierrez et al., 1997; Tsuboi et al., 1991) and industrial electronic scrap (Sturgill et al., 2000; Lee and Nam, 1998). Several techniques have been reported to recover gallium from

its resources, such as fractional precipitation, electrochemistry, solvent extraction (Kinoshita et al., 2004; Thakare and Malkhede, 2014), ion exchange (Rao et al., 2003), and liquid membrane (Kumbasar and Tutkun, 2006). Owing to the limitations of the aforementioned techniques in the extraction of gallium and the increasing demand for gallium in recent years, the recovery of gallium through a simple and effective method is necessary. For this reason, present studies have been focused on the development of a gallium-extraction method using an ion-flotation technique. The concentration of gallium in its resources is relatively low (liquors from the Bayer process include 0.19 ± 0.02 g/L Ga, 80 g/L Al<sub>2</sub>O<sub>3</sub>, and 450 g/L Na<sub>2</sub>O; a typical composition of zinc refinery residue includes 10% Zn, 7.6% Fe, 5.3% Al, 2.1% As, 1.1% In, 0.3% Cu, and 0.03% Ga, as well as coal fly ash that includes 26% Al<sub>2</sub>O<sub>3</sub>, 10% Fe<sub>2</sub>O<sub>3</sub>, 4% CaO, 0.16% Zn, and 0.013% Ga). Such an ion-flotation technique has the ability to concentrate extremely dilute solutions. Therefore, this technique is likely to be preferred over other methods for the recovery of gallium from acidic and basic leach liquors in terms of energy consumption and reagent losses. In this study, since Zn(II) is as an impurity in the acidic solutions obtained from gallium sources, the selective extraction of Ga(III) from Zn(II) using SDS was investigated by ion flotation. Ion flotation introduced by Sebba has been known as a separation process since 1960 (Sebba, 1962). This technique has a great number of

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advantages, including simplicity, rapid operation, economical, good separation yields (for treating solutions with low concentrations of metal ions ( $10^{-6}$ – $10^{-2}$  mol L<sup>-1</sup>) and high volume), and low energy requirements (Stoica et al., 1998; Shakir et al., 2007). All of these factors make it an effective and low-cost method for the recovery of valuable and rare metals with low concentrations from industrial wastewater. During ion-flotation, a solution is initially homogeneous, but, after the addition of a surfactant (collector) with the opposite ionic charge of the colligend, the solution becomes heterogeneous, and insoluble complexes (sublate) are formed. The sublate (the colligend–collector product) attaches preferentially to the bubbles passing through the solution. Then bubbles are transferred to the top of the column (Wang et al., 2006). One or more of the several mechanisms may occur during the ion flotation, relating to the solubility of the collector–colligend product and variation in pH. For example, the term “foam fractionation” (homogeneous ion flotation) is applied to the process in which the colligend–collector product is an ion pair or soluble complex (Grieves, 1975). Whereas, the term “precipitate flotation” is applied to the process in which the ionic species is first precipitated by pH variation or a non-surface-active agent (i.e. precipitating agent), and precipitated particles are then floated using a surfactant (surface-active reagent) (Wang et al., 2006; Grieves, 1975). Generally, these processes are identified by the kinetics of sublate removal and the solubility of the collector–colligend product (Filippov, 2000). Ion flotation is the most selective process in foam separation. Previous studies have indicated that many hydrodynamic and operation parameters affect the ion-flotation performance, such as ionic strength, metal ion concentration, solution pH, collector concentration, and gas flow rate (Reyes et al., 2012; Reyes et al., 2009; Kinoshita et al., 2013; Lemlich and Arod, 1972; Scorzelli et al., 1999; Zouboulis and Matis, 1987). Although these factors are important, the composition of the aqueous solution is the most important parameter. The selectivity and recovery of the ion of interest is low when the aqueous solution includes same charge ions (Ulewicz et al., 2001). In the ion-flotation technique, ions with charges of the same sign and the same valence compete for the adsorption to the collector; there-

fore, the separation efficiency of the metal values reduce (Sebba, 1962; Lemlich and Arod, 1972). An earlier study by Baarson and Ray showed that the effective selective separation of different metal ions is possible by pH control and stage flotation. The aim of this study is to investigate the selective extraction of Ga(III) from Zn(II) at different pH values (Baarson and Ray, 1963).

## 2. Materials and methods

### 2.1. Reagents and sample preparation

Gallium(III) nitrate hydrate 98+ [Ga(NO<sub>3</sub>)<sub>3</sub> · xH<sub>2</sub>O] and zinc nitrate [Zn(NO<sub>3</sub>)<sub>2</sub> · 4H<sub>2</sub>O] were purchased from Merck. SDS (Merck, 98%) was used as the collector and the frother. Caustic soda (NaOH) and hydrochloric acid (HCl), obtained from Merck, were used to adjust the solution pH. These reagents were prepared using double-distilled water and all solutions were left for 12 h before use. Ion flotation experiments were carried out in a Denver laboratory flotation machine with 1 L capacity cell. All operational and hydrodynamic parameters, except pH, were kept constant during all experiments and the pH range of 0–7 was studied. 2 L of a  $1.5 \times 10^{-3}$  M Ga(III) stock solution was prepared from gallium nitrate dissolved in hydrochloric acid, and to preserve the Ga(III) precipitate, the solution was adjusted to pH 2. The experiment processes are presented in Fig. 1. The cell and all laboratory glassware were thoroughly rinsed with distilled water three times at the end of any test. The percentage removal of ions (R) was calculated according to the relationship (Ehrampoush et al., 2011):

$$R (\%) = [1 - (C/C_0)] \times 100 \quad (1)$$

where C is the residual concentration of the metal ion in the treated solution and C<sub>0</sub> is the initial concentration of the ion in the feed solution. In the flotation tests, the initial concentration of ions (Ga(III) and Zn(II)) was the same and constant at  $1.5 \times 10^{-4}$  M.

The selectivity coefficient corresponding to the charge exchange equilibrium at the interface was also calculated using the following equation (Micheau et al., 2015):

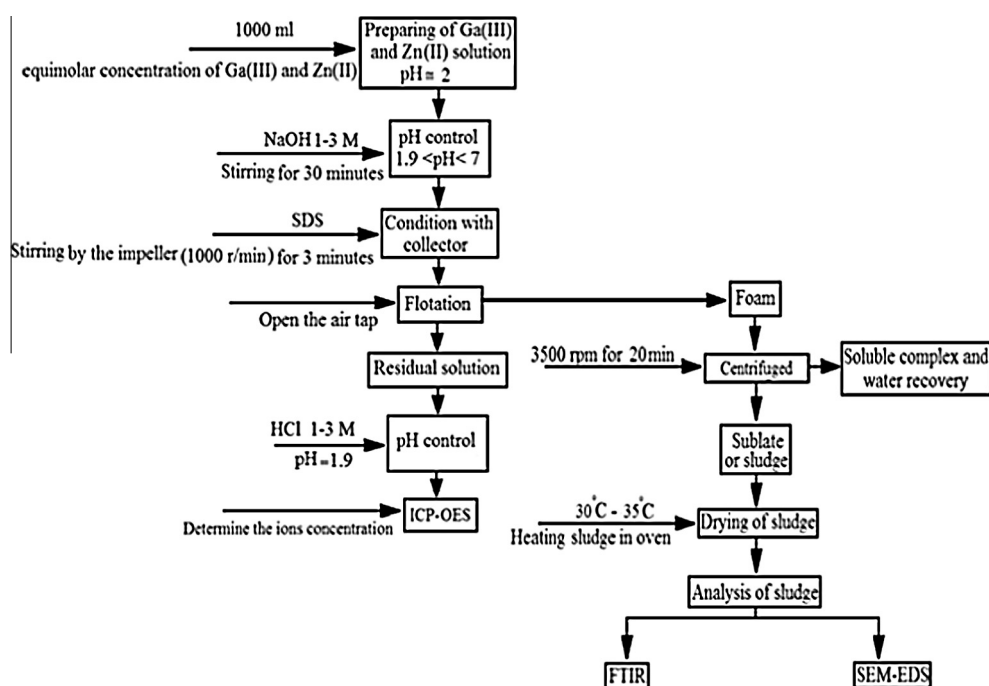


Fig. 1. Processing of the experiment.

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