



Separation of rare metals using silica gel modified with a surfactant micelle containing an extractant



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ABSTRACT

Solvent extraction is widely used as a universal method to extract and separate metals in industry. However solvent extraction has some disadvantages, for example, the use of a large amount of organic solvent which could be an environmental hazard. Therefore a solid-phase extraction method is proposed in this research. First of all, a silica gel modified with a nonionic surfactant (TritonX-100) micelle including 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester (EHPNA) as a metal extractant has been prepared. The amount of extractant contained in the modified silica gel was measured to evaluate the characteristics of the modified silica gel. Then the stability of the modified silica gel was examined. Next, the adsorption equilibrium and the adsorption rate of the metals (aluminum, gallium and indium) were measured to estimate the adsorption mechanism of the metals. Finally, the mutual separation of aluminum, gallium and indium using the modified silica gel packed in a column was investigated. The amount of EHPNA impregnated in the modified silica gel was 0.205 mmol/g-silica gel. The modified silica gel can be used as a stable adsorbent below pH 2.0 without leaking EHPNA. The results of adsorption equilibrium showed that the higher the pH, the higher was the extent of metal adsorbed. And among aluminum, gallium and indium, only indium was recovered with a three-metal mixture solution ranging pH 1.0 to pH 1.5. In contrast, as a result of examination of the separation of each metal using the modified silica gel packed in a column, the mutual separation of aluminum, gallium and indium was achieved by repeating the adsorption and desorption processes.

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1. Introduction

In recent years, the demand for rare metals has increased, because of their use as constructional, electronic, magnetic and functional materials. In particular, gallium and indium are nowadays used for semiconductor materials. Especially indium and its compounds have numerous industrial applications and they are currently used in the manufacture of liquid crystal displays, semiconductors, low-temperature solders and infrared photo-detectors (Alfantatazi and Moskalyk, 2003). Gallium and indium are very widely dispersed on the earth's surface, but little of these elements occur naturally in Japan. They are often found in industrial wastes because the amount of consumption of these metals is still increasing in Japan. From this point of view, it is necessary to recover and separate these metals from waste materials. Today solvent extraction is widely used as a universal method in industry to extract and separate metals. However solvent extraction has some disadvantages such as the heavy use of organic solvents which could be an environmental hazard.

As the main techniques used to remove metals other than the solvent extraction method, there have been ion exchange, chemical precipitation, metal complex formation, membrane separation, advanced oxidation processes, electrolysis, reverse osmosis, and biological treatment. However, these methods may be ineffective or expensive, especially when the heavy metal ions are present at low concentrations in the wastewater.

In order to overcome these disadvantages, several methods with respect to solid-phase extraction have been developed. Silica gel modified chemically (Jal et al., 2001; Tokman et al., 2003; Cui et al., 2007; Ngeontae et al., 2007; Zhu et al., 2007; Madrakian et al., 2008; Puanngam and Unob, 2008; Aeungmaitrepirom et al., 2009; Martendal et al., 2009; Repo et al., 2009; Mahmoud et al., 2010; Pereira et al., 2010; Tian et al., 2010; Abdel-Fattah et al., 2011; Donia et al., 2011; Sharma et al., 2013; Wang et al., 2013) and nanometer-size alumina particles modified chemically (Afkhami et al., 2010; Ezoddin et al., 2010) were used for the extraction and separation of transition and heavy metals. And there have been some researches (Garcia-Valls et al., 2001; Filho et al., 2006; Qu et al., 2008; Sadeghi and Sheikhzadeh, 2009; Sharma et al., 2012) as to rare and toxic metals. Especially for gallium and indium extraction, Nishihama et al. (1998) studied only the extraction and separation of the metals using organophosphorus extractants to determine the extraction mechanism. They also

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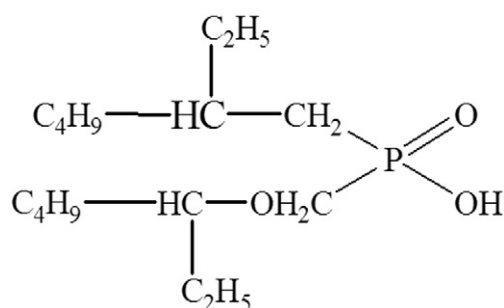


Fig. 1. Structure of EHPNA.

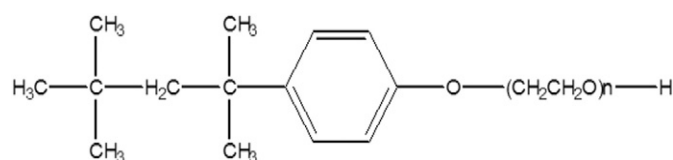


Fig. 3. Structure of TX-100.

presented new ideas to separate and recover gallium and indium (Nishihama et al., 1999). We have studied the equilibrium and kinetics of gallium extraction (Kondo and Matsumoto, 1996), the separation of gallium and indium by a supported liquid membrane (Kondo et al., 1997), the separation and concentration of indium by an emulsion liquid membrane (Kondo and Matsumoto, 1998) and the extraction mechanism of gallium and indium by microcapsules containing organophosphorus extractants (Kamio et al., 2002). Other than the studies mentioned above, the extraction of gallium and indium has been studied using several methods (Lee et al., 2002; Liu et al., 2006; Ohashi et al., 2006; Uezu et al., 2006; Kurahashi et al., 2008; Adhikari et al., 2011). There have been few researches so far with respect to adsorption of gallium and indium onto chemically modified silica gel.

In the light of the research described above, we report here a solid-phase extraction method for gallium and indium from an aluminum co-existing solution. We used silica gel as a solid support and 2-ethylhexyl phosphonic acid mono-2-ethylhexyl ester (abbreviated as EHPNA) as the metal adsorbent. EHPNA cannot be combined directly to silica gel due to its strong hydrophobicity. So we proposed the use of a surfactant micelle. Through this method, EHPNA can stay in the micelle because the interiors of the micelles are hydrophobic. Our research reported here is also a work wherein the limit of application of EHPNA to gallium or indium separation using various kinds of method is tested.

First of all in the work, silica gel modified with nonionic surfactant (TritonX-100) micelle including EHPNA was prepared and the stability of the modified silica gel was examined. Next, the adsorption behavior of gallium, indium, and aluminum onto the modified silica gel was investigated to clarify the adsorption mechanism as well as to study this industrial application. Finally the separation of each metal using the modified silica gel packed in a column was examined. The mutual separation of aluminum, gallium and indium was achieved by repeating adsorption and desorption processes.

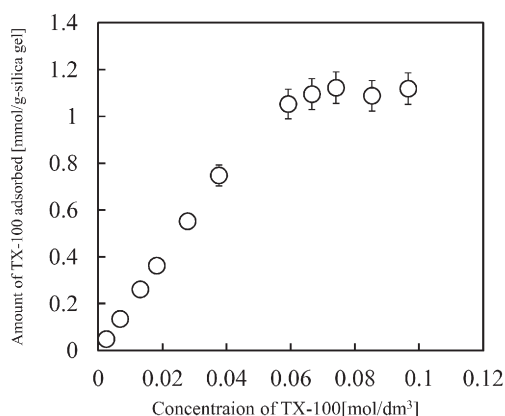


Fig. 2. Relationship between amount of TX-100 adsorbed on silica gel and concentration of TX-100.

2. Experimental

2.1. Reagents

Silica gel 60, with a particle size of 70–230 mesh, and the nonionic surfactant, Triton® X-100 (p-(1,1,3,3-tetramethylbutyl)phenoxy poly(oxyethylene)glycol, abbreviated as TX-100) and ethanol (99.5%) were obtained from Nacalai Tesque Inc., Japan and they were used without further purification. EHPNA was kindly supplied from Daihachi Chemical Industry Co., Ltd. (Osaka, Japan) and was used without further purification. The structural formula of EHPNA is shown in Fig. 1.

Aqueous solutions were prepared using 0.1 mol/dm³ HCl-CH₃COONa buffer solution containing the desired amount of metal ions. The pH of the aqueous solutions was adjusted by means of a pH meter (Horiba F-23). As the metal sources, special grade metal chlorides (GaCl₃, InCl₃ and AlCl₃) were used.

2.2. Adsorption experiments of TX-100 to silica gel

Adsorption experiments for TX-100 on silica gel were carried out batch wise as follows. Silica gel, (0.5 g), and 10 cm³ of an aqueous solution containing TX-100 were contacted in a vial. The vials were shaken at 100 rpm in a thermostat bath at 303 K for 24 h to attain equilibrium. Then TX-100 solution and silica gel were separated by filtration. The TX-100 concentrations in the aqueous solutions before and after equilibrium were measured using a UV-VIS spectrophotometer (UV-2500, Shimadzu Co. Ltd., Japan) at 275 nm. The TX-100 concentration adsorbed onto the silica gel was calculated from the mass balance before and after equilibrium.

2.3. Preparation of modified silica gel

The immobilization of EHPNA into the micelles of TX-100 was carried out as follows. An ethanol solution containing EHPNA and a 0.08 mol/dm³ aqueous solution containing TX-100 prepared using the buffer solution (pH 7.0) were mixed and the final concentration of EHPNA was so controlled as to be 30 mmol/dm³ in the EHPNA-TX-100 solution. This EHPNA-TX-100 solution (0.2 dm³) and 10 g silica gel were mixed in a vial, and then stirred in a thermostat bath at 303 K for 24 h. Compounds

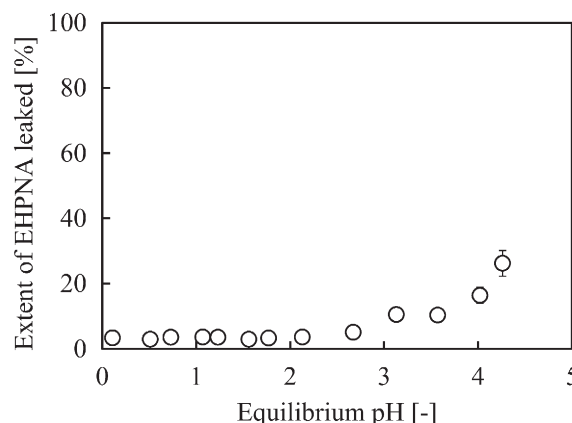


Fig. 4. Relationship between extent of EHPNA leaked and equilibrium pH.

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