



Sono-electrochemical recovery of metal ions from their aqueous solutions



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HIGHLIGHTS

- Sono-electrochemical method is effective in the recovery of Pd, Ga and Pb.
- Recovery efficiency depends on the type of metal ion and megasonic frequency used.
- Pd is recovered mainly in metallic form while Ga and Pb show presence of oxide.

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ABSTRACT

Metal recovery from aqueous waste streams is an important goal for recycling, agriculture and mining industries. The development of more effective methods of recovery have been of increasing interest. The most common methods for metal recovery include precipitation, electrochemical, ion exchange, flocculation/coagulation and filtration. In the current work, a sono-electrochemical technique employing sound field at megasonic frequency (500 kHz or 1 MHz) in conjunction with electrochemistry is evaluated for enhanced recovery of selected metal ions (palladium, lead and gallium) with different redox potentials from their aqueous solutions. The surface morphology and elemental composition of the metal deposits were characterized using scanning electron microscopy, energy dispersive X-ray spectroscopy, and X-ray photoelectron spectroscopy. The percent recovery was found to depend on the type of metal ion and the megasonic frequency used. Palladium was recovered in its metal form, while lead and gallium were oxidized during or after the recovery process.

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1. Introduction

Cost effective and energy efficient recovery of metals from aqueous waste streams is an important goal for recycling, agriculture, and mining industries [1,2]. Metals, such as lead, can contaminate the environment when mining operations are performed or electronic waste is processed [3]. Since rare earth and heavy metals are frequently toxic even at very low concentrations, and their refined forms are industrially valuable, the development of more effective methods of recovery have been of increasing interest [4]. The prevailing methods for metal recovery from aqueous sources include precipitation, electrochemical, ion exchange, flocculation/coagulation and filtration [4,5]. These tra-

ditional methods suffer from drawbacks such as non-selectiveness in precipitation process, susceptibility to suspended solids during flocculation/coagulation, pH sensitivity during ion exchange, and poor removal rate in filtration [5,6].

Metal recovery methods can be categorized into two major groups based on the working mechanisms: physical and chemical processes [4–8]. Physical processes separate liquid from solid phase in a mixture, which can be accomplished in a variety of ways such as filtration and/or coagulation/flocculation [7]. Filtration is a physical remediation method that can remove heavy metal ions from solution using techniques like reverse osmosis (RO), although the filter can get quickly clogged, which leads to a decrease in membrane performance and higher costs due to eventual membrane replacement [5,8]. The other drawback with RO is that it is only effective with aqueous solutions containing low concentrations of contaminate [6]. Coagulation/flocculation in wastewater treatment process destabilize smaller particles by removing repulsive elec-

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trostatic forces between them [9] causing them to agglomerate and settle faster. While useful at removing particles from aqueous solutions, coagulation/flocculation has its drawbacks since it is unable to remove solutes from solutions directly [10].

Compared with physical methods for recovering metals from aqueous solutions, chemical methods operate by causing chemical or electrochemical reactions of metals in solution or by changing the solubility limit of dissolved species [6,8]. Chemical precipitation is the most widely used method for heavy metal remediation, mainly due to its low cost and high removal efficiency [4,5], although it is plagued by the large amount of consumption of reagents to bring pH to the required level and environmental problems associated with its sludge production after the process [8,11].

Ion exchange is another option for metal recovery, which substitutes heavy metal ions with other ions [12]. High cost of installation and maintenance make it a limited usage, it has problems with suspended solids in aqueous solution as well [8]. An alternative chemical method is the electrochemical process, which has good efficiency and can be used with high concentrations of dissolved heavy metals [8]. It is also associated with lower cost compared to conventional chemical precipitation [13–19]. One advantage of this technique is that dissolved metal ions can be directly deposited in their metallic form [16]. Metals that have already been demonstrated to be removed by electrodeposition process include silver, copper, lead, gallium, nickel, zinc and cadmium [16,17,19]. Further the technique offers selectivity, which can greatly simplify the recycling process [20]. If the performance and efficiency of this technique can be further augmented through assistance of a low cost and a green technology, it will likely be of great value to industries interested in metal recovery.

In this work, we employ a sono-electrochemical technique utilizing sound field at megasonic frequencies in conjunction with electrochemistry for enhanced recovery of selected metal ions, with widely differing redox potentials, from their aqueous solutions. Electrochemical potentials were applied in the range where hydrogen gas was produced, which was stabilized in the form of oscillating bubbles using the megasonic field. The extreme temperatures and highly reducing environment inside the bubbles were used for the reduction of metal ions to their zero state solid form. The metals chosen for this study were palladium, gallium and lead. Palladium is used as a catalyst in automobile and other industries, and is critical for conservation of resources [21]. Lead is widely used in the industry, and it is an environmental contaminant above threshold level and can cause irreversible health effects to human beings [5]. Gallium is an important element for manufacturing electronic products, and its sources are limited in nature [22]. The paper is organized as follows: in Section 2, we describe the experimental set-up and conditions and electrochemical, analytical and surface characterization techniques used in this study. In Section 3, we have presented and discussed the key results of this work. Finally, the main conclusions of this research are provided in Section 4.

2. Experimental

2.1. Chemicals used in this study

Aqueous solutions were prepared using high resistivity deionized (DI) water (18 MΩ-cm), and the chemicals used were of

Table 1
Composition of the examined aqueous solutions.

Metal	Electrolyte composition		pH
Palladium (Pd)	Pd(NO ₃) ₂	3.5 mM	~0.9
	HNO ₃	1 M	
Lead (Pb)	PbCl ₂	100 mM	~3.8 (adjusted using HAc)
	CH ₃ COONH ₄	0.5 M	
Gallium (Ga)	GaCl ₃	10 mM	~2.5 (adjusted using HCl)
	NaCl	0.5 M	

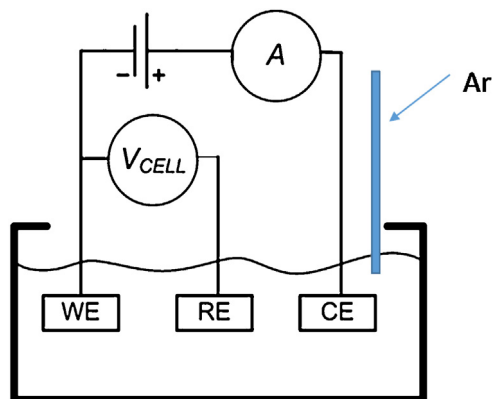


Fig. 1. Schematic of the three electrode system used in the study. WE, CE and RE refer to working, counter and reference electrodes.

analytical reagent (AR) grade. Palladium nitrate (Pd(NO₃)₂, 99.9%), lead chloride (PbCl₂, >99%), gallium chloride (GaCl₃, 99.9%), ammonium acetate (CH₃COONH₄, ≥97%), nitric acid (HNO₃, 70%), acetic acid (HAc, ≥99.7%), hydrochloric acid (HCl, 37%) and sodium chloride (NaCl, ≥99%) were purchased from Thermo Fisher Scientific (New Jersey). The chemical compositions and pH values of the solutions used in the study are listed in Table 1.

2.2. Voltammetry study of different metals

The electrochemical setup consisted of a glass vessel (~50 mL liquid volume) sealed with Parafilm with provision for inserting the electrodes. Three electrode system was used for both cyclic voltammetry (CV) and chronoamperometry studies, as illustrated in Fig. 1. During CV measurements, the potential of working electrode was scanned at 20 mV/s in the potential range of –2.0 to 1.0 V (cathodic first) (versus Ag/AgCl, sat. KCl), while in the chronoamperometry study, the potential was maintained at a constant value (versus Ag/AgCl, sat. KCl) by means of a Gamry Reference 3000™ Potentiostat (Warminster, PA). The electric current was measured as a function of voltage in the CV study, and as a function of time during the chronoamperometry study. Sonication experiments were performed by using: (a) Mini-meg[®] tank (PCT Systems Inc.) of volume ~4.5 L and consisting of a 125 cm² transducer affixed at the bottom with an operating frequency of 1 MHz and power density of 8 W/cm², and (b) a cubical reactor (12 L), custom made by Weber Ultrasonics Inc. (Clarkston, MI, USA), with 500 kHz transducer (73 cm²) on the side wall operating at 8 W/cm².

In all the experiments performed, the aqueous solutions were saturated with ultrahigh purity argon (Ar, 99.999%) by bubbling

Table 2
Half reactions and details of three electrode electrochemical cell.

Half reaction	E ₀ (V)	Working electrode	Counter electrode	Reference electrode
Pd ²⁺ + 2e ⁻ → Pd (s)	+0.987	Pt mesh	Pt mesh	Ag/AgCl (sat. KCl)
Pb ²⁺ + 2e ⁻ → Pb (s)	-0.126	Copper sheet	Pt mesh	Ag/AgCl (sat. KCl)
Ga ³⁺ + 3e ⁻ → Ga (s)	-0.530	Vitreous carbon rod	Pt mesh	Ag/AgCl (sat. KCl)

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