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Gold Recovery from Waste Printed Circuit Board Iodide Leachates Using a Novel Membrane-Based Electrowinning Process

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Abstract

The hydrometallurgical recovery of gold from waste printed circuit boards is considered a promising recycling technique. This paper presents the findings from a study investigating the feasibility of a hydrometallurgical technique to recover gold from waste printed circuit boardiodide leachates using a novel membrane-based electrowinning process. Findings showed that the investigated novel process has the potential to be technically feasible if employed in a real life industrial-scale waste printed circuit board recycling operation. This is primarily because high purity (>99%) Au was effectively recovered using the investigated process with a 95.50 % yield under the following conditions: 6.0 hrs time, constant cell voltage of 12.9 V, temperature of 25 °C, 250 rpm agitation speed, average current of 5 A, 1429 A/m² equivalent current density, and 8.4×10^{-6} mol/m2.s Au flux. Overall, the simultaneous recovery of iodide lixiviant with Au makes the recovery process simple as well as potentially cost-effective. Further studies on the process scale-up are required to accurately evaluate the overall costing of the gold recovery process via the proposed technology.

Keywords: Electrowinning, Hydrometallurgy, Gold, Recycling, Membrane

1. Introduction

Gold (Au) is arguably the most important metal which is recovered during an industrial scale waste printed circuit board (WPCB) recycling operation. This is because Au is a highly economically valuable metal which is present in WPCBs in appreciable concentrations, often tens to hundreds of times greater than in naturally occurring ore deposits. In terms of economic value per kg of WPCB, Au accounts for 60-80 % of the total economic value of WPCBs [1]. Hence, the profitability of most industrial-scale WPCB recycling operations is intrinsically linked to successful Au recovery. Unfortunately, significant value loss and resource wastage occurs when conventional processes utilising expensive pyrometallurgical techniques are used for Au recovery from WPCBs. In response to these drawbacks, there has been increased research into the development of efficient, environmentally sustainable, and economically feasible hydrometallurgical processes for Au recovery from WPCBs [2]. Most hydrometallurgical processes for Au recovery from WPCBs commence with the leaching of a size-reduced WPCB particulate in a strongly oxidising and complexing aqueous lixiviant. A strongly oxidising lixiviant is required for Au leaching since Au possesses a large positive reduction potential, owing to the relativistic contraction of its 6s orbitals, while a

strongly complexing lixiviant is required since leached Au (as Au³⁺ and/or Au⁺ ions) is thermodynamically unstable unless it is complexed by a suitable ligand [3]. During the aforementioned leaching process, Au, and other metals (namely Ag, Pd, Fe, Ni, Zn, Sn, Sb, Mn, and Al) present in the WPCBs encounter the lixiviant and are subsequently leached. This leaching step produces a leachate from which Au is recovered after a refining process utilizing techniques such as solvent extraction and electrowinning [2,4].

Previous investigations into the hydrometallurgical recovery of Au from WPCBs and other secondary sources have focused on the use of cyanide lixiviants for the leaching step [5,6]. However, due to the environmental and human health risks associated with cyanide toxicity, there has been a great deal of research on the replacement of cyanide with alternative lixiviants such as thiosulfate, thiourea, and halides [7,8,9]. Among them are iodide lixiviants which are known to be recyclable, non-toxic, and highly efficient Au leaching lixiviants.

Iodide lixiviants used for Au leaching are typically aqueous solutions containing a diiodine (I2) oxidising agent, iodide (I–) complexing agent such as potassium iodide (KI) or sodium

iodide (NaI), and a hydrogen peroxide (H_2O_2) auxiliary oxidising agent. Au is typically leached in iodide lixiviants as AuI_2^- at acidic pH (2-7) according to equation 1[8,10,11].

 $2Au_{(s)} + I_{(aq)}^{-} + I_{3}^{-}{}_{(aq)} \rightarrow 2AuI_{2}^{-}{}_{(aq)}$ (1)

Although iodide lixiviants have been reported to be highly effective at leaching Au from WPCBs, there remains an issue on how to recover Au from WPCB iodide leachates in an efficient and economically feasible manner [11,12]. Previous authors have reported the high yield (> 99 %) recovery of Au from WPCB iodide leachates by reductive precipitation under highly acidic (pH < 1.6) and alkaline conditions (pH > 13) induced by 0.1 M ascorbic acid and 0.1 M sodium hydroxide respectively. However, considering the complex metal compositions of WPCB iodide leachates, extensive co-precipitation of impurities is expected to occur, resulting in the need for further beneficiation [11]. Moreover, strongly alkaline, and acidic precipitation reagents are highly corrosive and thus necessitate the use of expensive corrosion resistant equipment [13,14].

Hence, there is a need to develop a more efficient and less problematic process for Au recovery from WPCB iodide leachates. The present work uses a laboratory experiment to investigate the feasibility of Au recovery from WPCB iodide leachates using a novel membrane-based electrowinning process.

2. Experimental

Electrowinning Cell. The laboratory experiment was conducted in a novel custom-made membrane-based electrowinning cell comprised of a feed tank containing a test WPCB iodide leachate, together with a catholyte and anolyte tank containing an iodide electrolyte solution. Figure 1 schematically illustrates the electrowinning cell.

The electrowinning cell was comprised of three distinct, hollow, open-top cubic Pyrex tanks (feed, catholyte, and anolyte tank) with inner volumes of 2197 mL. Each tank was equipped with a 20/20 cm gasket. A circular hole with a 10 cm diameter was machined into the left and right lateral faces of the catholyte tank, left lateral face of the anolyte tank, and right lateral face of the feed tank. A square 12/12 cm Au selective ion exchange extractant membrane (Au-IEM) was positioned radially between the right lateral face of the feed tank and the left lateral face of the anolyte tank and the left lateral face of the anolyte tank and the left lateral face of the catholyte tank, while a square 12/12 cm anion exchange membrane (AEM) was positioned between the left lateral face of the anolyte tank and the right lateral face of the catholyte tank. Eight nut and bolt fasteners were used to join the three tanks of the electrowinning cell together. These fasteners passed through the gasket of the tanks.

The electrowinning cell utilised a 35 cm2 contact surface area mesh titanium anode and stainless-steel plate cathode spaced 13 cm apart. The cathode was connected to the negative pole of a DC power supply, while the anode was connected to the positive pole. The DC power supply had a maximum voltage >24 V, was capable of operating under constant current or voltage mode and had the capability to be power factor corrected. To control agitation speed, each tank of the electrochemical cell used in this project was equipped with an overhead stirrer.



Figure 1: Schematic Illustration of Side View (Bottom) And Isometric View (Top) Of Experimental Electrowinning Cell, Where 1 = Feed Tank, 2 = Catholyte Tank, 3 = Anolyte Tank, 4 = Au Selective Ion-Exchange Extractant Membrane, 5 = Cathode, 6 = Anion Exchange Membrane, 7 = Anode, 8 = Dc Power Supply, 9 = Overhead Stirrer, 10 = Gasket with Nut and Bolts

WPCB Iodide leachate, and Iodide Electrolyte Solution. The WPCB iodide leachate and iodide electrolyte solution used in the laboratory experiment was prepared in a low-form glass beaker on a heating plate, equipped with a 3D printed system closure, an overhead mechanical agitator, and a thermometer. Figure 2 shows the leaching set-up.



Figure 2: Scheme of Leaching Set-Up

The iodide electrolyte solution used in the laboratory experiment was an aqueous solution prepared with a 2.5 g/L I2 (Merck) and 10 g/L KI (Merck) concentration. While, the WPCB iodide leachate was an aqueous solution prepared by sequentially leaching pulverised WPCBs (2/2mm cm particle size) in 0.2 M nitric acid/ HNO, lixiviant (for 45 min at 500 g/L S/L, 18 rpm agitation rate, and 90 °C temp), 3.5 M hydrochloric acid/ HCl lixiviant (for 120 min, at 500 g/L S/L, 18 rpm agitation rate, and 90 °C temp), 2M sulphuric acid/ H₂SO, and 2M hydrogen peroxide/ H₂O₂ lixiviant (for 3 hrs, at 500 g/L S/L, 18 rpm agitation rate, and 25 oC temp), and finally 2.5 g/L I, 10 g/L I⁻, and 10 g/L H₂O₂ lixiviant (for 4 hrs at 500 g/L S/L, 18 rpm agitation rate, 25 °C temp, and pH 6). The observed metal leaching yields and metal concentrations in the leachates produced after each respective leaching stage are presented in Table 146 1 and 2. These yields and concentrations were determined through ICP-OES chemical analysis. It is important to note that all leaching stages were performed 3 times under a single set of operating conditions, and the reported metal concentrations are set averages, with their repeatability evaluated through standard deviation.

Images of the WPCBs (prior to pulverisation) used in the preparation of the WPCB iodide leachate are shown in Figure 3. The WPCBs were originally 50 g in mass and 10 cm x 15 cm in size and their metal composition is shown in Table 3. Metal composition of the WPCBs used was determined by leaching 50 g of pulverised and homogenised WPCBs in aqua regia at a S/L ratio of 25 g/L, for 24 hrs, and then chemically analysing the leachate produced by ICPOES. It is important to note that the WPCBs used in this study were unpopulated (not covered by a solder) ideal/model boards which were specifically designed for this project by Trax Interconnect.



Figure 3: 10 Cm X 15 Cm WPCB Used in The Laboratory Experiment (Left = Front, Right = Back)

2.1 Au Selective Ion Exchange Extractant Membrane (Au-IEM)

The Au-IEM used in the laboratory experiment was a heterogenous membrane comprised of 40 wt.% poly-vinylchloride (PVC) polymer-backbone and 60 wt.% Purogold S992 extractant carrier. This membrane composition was based on data provided by Křivčík et al. who suggested that extractant membranes with a 40/60 polymer-backbone/ extractant carrier ratio exhibit good selectivity, permselectivity, and conductivity [15]. This is important considering that selectivity determines how well the Au-IEM separates Au ions from competing base metal (BM) impurity ions, permselectivity determines the efficiency (yield) in which the Au-IEM will transport Au ions through its body, and conductivity determines the rate at which Au ions will be transported through the Au-IEM body.

Synthesis of the Au-IEM commenced with the preparation of a casting solution in the form of a 100 % 1.0 L tetrahydrofuran (THF) solution (Merck) containing approximately 20 g of dissolved S5718 PVC (Sasol) and 30 g of undissolved Purogold S992 extractant (Purolite). This casting solution was mixed for 30 min to obtain a uniform distribution of PVC-THF solution and S992 extractant. Once prepared, the casting solution was poured into a 150/15 mm petri dish (sitting on a flat glass plate and covered with filter paper and a watch glass), which was placed in a flat box under inert nitrogen (N₂) atmosphere for 24 hrs to allow for THF to evaporate slowly and have little contact with air humidity, thus, to avoid excessive formation of pores

(which would result in decreased selectivity and permselectivity of AuI_2^{-}). After complete evaporation of THF, a few droplets of distilled H₂O was added into the petri dish, and thereafter the remaining solid (Au-IEM) was peeled off.

It must be noted that the Purogold S992 extractant carrier was not ground prior to mixing with THF to ensure that the individual resin beads which made up the puro gold S992 extractant carrier retained their mechanical strength and performance. Furthermore, it was observed that the individual resin beads of the puro gold S992 extractant carrier did not disintegrate when mixed with THF as expected. Instead, the PVC acted as a binding agent which joined/ cast the individual resin beads of the S992 extractant carrier together. puro gold S992 has a macroporous structure and a styrenic backbone which is crosslinked with divinylbenzene (DVB). This means that it contains a network of pores within a gel matrix. Light is scattered by the pores, making the Purogold S992 extractant appear opaque. Proprietary functional groups that are attached to the backbone and located throughout the Purogold S992 extractant interact with Au ions in solution. The rate of reaction (kinetics) between Au ions in solution and the Purogold S992 extractant is controlled by the rate of ion diffusion through the thin film of H2O surrounding each individual bead of the Purogold S992 extractant (film diffusion) and through each bead of the Purogold S992 extractant itself (particle diffusion). In a dilute solution/ during the adsorption phase, the rate of film diffusion is slower than the rate of particle diffusion. However, during the subsequent stripping/ elution phase, the rate of film diffusion is faster than the rate of particle diffusion [16].

It is important to note that in the present study, resin beads of Purogold S992 are suspended within a PVC matrix. Hence, a PVC layer is present over the surface of each individual S992 resin bead in the Au-IEM employed. This PVC layer increases the diffusion path length of ions, and as a result, may interfere with and reduce the rate of ion diffusion, since ions must travel through the outer PVC later before they can interact with the S992 resin beads and their outer H_2O film. Furthermore, it is possible that when the S992 resin beads were mixed in PVC, their outer H_2O film may have been damaged, thus resulting in further interference and reduction of ion diffusion rate.

2.2 Anion-Exchange Membrane (AEM)

The AEM used in the laboratory experiment was the AFX membrane (Neosepta and Astom Corporation). The AFX membrane is a homogenous AEM comprised of a poly-styrene-

co-divinylbenzene base polymer with positively charged ionexchange functional groups (which are typically propriety quaternary ammonium bases (CH_3)₃N⁺)). The AFX membrane is also reinforced and interpenetrated with PVC [17].

3. Method

The laboratory experiment investigating the feasibility of Au recovery from WPCB iodide leachates using a novel membranebased electrowinning process commenced by loading 2.0 L of WPCB iodide leachate into the feed tank of the experimental electrowinning cell and loading 2.0 L of iodide electrolyte solution into the catholyte and anolyte tanks. Thereafter, electrical power was provided to the DC power supply connected to the electrodes in the cell, at a constant cell voltage of 12.9 V, temperature of 25 °C, 250 rpm agitation speed, average current of 5 A, and 1429 A/m² equivalent current density. After 6.0 hrs, the supply of electrical power was terminated, and the cathode was removed from the electrowinning cell. Thereafter, the darkbrown Au powder deposit on the cathode was scrapped off, weighed, and stored in a 100 mL container. A 25 mL sample solution was also collected from the feed and catholyte tanks after 2, 4, and 6 hrs. These sample solutions were placed in 50 mL containers for storage. The Au and BM concentration of the deposited Au and sample solutions from the feed tank and catholyte tank was determined through ICP-OES chemical analysis. Au recovery yield was then determined using equation 2. Where Y is the Au recovery yield, C_1 is the amount of Au (in mg) in the WPCB iodide leachate before cell voltage is applied, and C₂ is the amount of Au (in mg) deposited on the cathode.

$$Y = (C_2/C_1) \times 100(\%)$$
(2)

A 50 mL sample solution was also collected from the anolyte tank after 6 hrs, and the I2 and I- concentration of this solution was determined through chemical analysis using UV-Vis spectroscopy.

It is important to note that all experiments were done in 3 sets under a single set of operating conditions and all concentrations reported are set averages, with repeatability evaluated through standard deviation.

4. Results and Discussion

Au and BM Concentrations in WPCBs. Chemical analysis results in Table 1 show that each WPCB used in the preparation of WPCB iodide leachate was comprised of 0.11 wt.% Au, 55.7 wt.% Cu, 7.00 wt.% Al, 1.60 wt.% Ni, and 3.31 wt.% Fe.

	Au	Cu	Al	Ni	Fe
Metal wt.%	0.11	55.7	7.00	1.60	3.31

Table 1: Au and BM Concentrations in WPCBs

It is important to note that the high Au and Cu concentration of the model/ideal WPCBs used in this work is over 3 times higher than that of typical/non-ideal WPCBs. Ideal/model WPCBs were used since they allowed for the simple preparation of homogenous WPCB feed particulate. The use of ideal homogenous WPCB feeds with high Au and Cu concentrations was also beneficial since it allowed for accurate mass balancing of Au and Cu after leaching and electrowinning. Effective mass balancing of Au and Cu is important since these metals hold most of the economic value of WPCBs and they are the only metals which reported into the WPCB iodide leachate in appreciable concentrations. The use of typical/non-ideal WPCBs was not considered in this work due to scope constraints, and because the use of typical/non-ideal WPCBs may have resulted in a decreased statistical significance of results, due to increased variability/imprecision between test results. This is because the use of typical WPCBs is expected to result in a more inconsistent/ variable WPCB feed composition between the various leaching trials (due to the complex and variable chemistry of typical/ non-ideal WPCBs), and as a result increase the variability of the laboratory experiment results and thus decrease statistical significance.

4.1 Au and BM Concentrations in Leachates and Leaching Yields

Chemical analysis results of the Au and BM (Cu, Al, Ni, and Fe) concentrations in the leachates produced after each sequential leaching stage during the preparation of WPCB iodide leachate are presented in Table 2.

Results in Table 2 show that after the final iodide leaching stage, 1.07 ± 0.009 g of Au reported into 2.0 L of WPCB iodide leachate, with Cu the only BM impurity which reported into in the WPCB iodide leachate, present in a concentration of 0.121 ± 0.001 g.

		HNO3	HCl	H2SO4-H2O2	I2-I–-H2O2
Metal con- centration (g) in 2.0 L leachate Metal con-	Au	0.00	0.00	0.00	1.07 ± 0.009
	Cu	185 ± 0.742	95.1 ± 0.753	156 ± 1.25	0.121 ± 0.001
	Al	31.4 ± 1.07	15.9 ± 0.310	2.21 ± 0.118	0.00
	Ni	6.56 ± 0.438	1.66 ± 0.044	1.23 ± 0.027	0.00
	Fe	20.7 ± 0.295	1.86 ± 0.224	1.55 ± 0.20	0.00

Table 2: Au and BM Concentrations in Leachates

Chemical analysis results of Au and BM leaching yields after each respective leaching stage are summarised in Table 3. These results show that the total Au leaching yield obtained after the preparation of the WPCB iodide leachate was 97.78 %. The high Au leaching yield observed following the preparation of the WPCB iodide leachate is comparable to the Au leaching yield observed by Batnasan et al. who used a similar conventional iodide lixiviant to leach Au from WPCBs at a >95 % yield [11].

		Leachate					
		HNO3	HCl	H2SO4-H2O2	I2-I–-H2O2	Cumulative	
Metal	Au	0.00	0.00	0.00	97.78	97.78	
leach-	Cu	33.20	17.06	28.28	0.02	78.56	
ing	Al	43.99	22.64	3.08	0.00	69.70	
yield/	Ni	39.11	10.32	7.58	0.00	57.02	
%	Fe	63.11	5.62	4.32	0.00	73.04	

Table 3: Au and BM leaching Yields

Results in Table 3 also show that the BM leaching yields observed during the preparation of the WPCB iodide leachate (with cumulative yields ranging between 57.02 % to 78.56 %) are significantly lower than the WPCB BM leaching yields reported by previous authors who used either HNO₃, HCl, or H₂SO₄-H₂O₂ lixiviants to leach BMs from WPCBs with >95 % yields [18-22]. The lower BM leaching yields observed can be attributed to the high 500 g/L S/L ratio employed, which is significantly greater than the 100 g/L to 333 g/L S/L ratio employed by previous authors [18-22]. Although the high 500 g/L S/L ratio employed resulted in less feasible BM leaching yields, a high Au leaching yield was observed at this S/L ratio. The high 500 g/L S/L ratio employed in this work is also supported by Yannopoulos who reported that Au is effectively leached at high S/L ratios between 350 g/L to 500 g/L in industrial operations [17]. A higher S/L ratio can also be beneficial since it may result in a reduced leaching reactor size requirement and cost.

Considering that the investigated process is concerned with only Au recovery, the obtained leaching results suggest that the utilised leaching process is technically feasible for its intended purpose since interfering/competing BM impurities did not readily report into the WPCB iodide leachate, while Au was leached with high yield.

4.1.1 Au Recovery

Based on the findings of previous authors[1,12], it can be expected that the application of the 12.9 V cell voltage during the laboratory experiment resulted in the transport of dissolved Auiodide ions (primarily as AuI2⁻) from the WPCB iodide leachate in the feed tank to the surface of the cathode in the catholyte tank where they underwent a reduction reaction (equation 2) which produced iodide ions and the solid Au deposit which was observed on the cathode surface as a brown powder. While iodide ions in the catholyte tank are believed to be transported (through the AEM) to the surface of the anode in the anolyte tank where they were oxidized to di-iodine (equation 3). This iodide oxidation reaction was presumed to be coupled by the simultaneous electrolysis of H₂O at the anode surface to O₂ and H⁺ (equation 4).

$\operatorname{AuI}_{2^{-}(aq)} + e \rightarrow \operatorname{Au}_{(s)} + 2I^{-}_{(aq)}$	$E1 = 0.561 + 0.0592lg[a(AuI_2^{-})/a^2(I^{-})]$	(2)
$2I_{(aq)}^- 2e \rightarrow I_{2(aq)}$	$E3 = 0.544 + 0.0592 lga(I^{-})$	(3)
$2H_2O_{(aq)} - 4e \rightarrow O_{2(g)} + 4H_{(aq)}$	$E4 = 1.229 + 0.0592 lga(H^+)$	(4)

It is important to note that An AEM was used in the laboratory experiment for the inhibition of proton (H⁺) leakage and concentration into the catholyte tank. In theory, the inhibition of protons into the catholyte tank of the experimental electrowinning cell improves Au recovery efficiency, because an increase in proton concentration decreases the reduction potential for the Au-iodide ion (AuI,⁻) [12].

Chemical analysis results of the sample solutions collected from the electrowinning cell's feed and catholyte tanks after 2, 4 and 6 hrs (Table 4), show that there was no Au concentrated in the feed tank after 2 hrs. However, after 2 hrs, there was $0.498 \pm$ 0.021 g of Au (~47 wt.% of initial Au) which was concentrated in the catholyte tank, thus indicating that Au recovery as a deposit on the cathode was incomplete. Similarly, it was observed that

after 4 hrs, Au recovery was incomplete, since
$$0.174 \pm 0.015$$
 g of Au (~16.4 wt.% of initial Au) was found to be concentrated in the catholyte tank. However, after 6 hrs it was found that no Au was concentrated in the catholyte tank, thus indicating that Au recovery as a deposit on the cathode was complete after 6 hrs. Chemical analysis results of sample solutions also found that no BMs reported into the catholyte tank, thus confirming that Au was selectively transported through the Au-IEM and into the catholyte tank. However, it is important to note that the chemical analysis results of the Au-IEM found that 0.03 ± 0.004 g of Cu was adsorbed onto the Au-IEM after the laboratory experiment together with 0.051 ± 0.004 g of residual Au. However, this adsorbed Cu was not stripped and transported into the catholyte tank.

		0 hrs	2 hrs	4 hrs	6 hrs
Metal concentration/ g in feed tank con- taining 2.0 L WPCB iodide leachate	Au	1.07 ± 0.009	0.00	0.00	0.00
	Cu	0.121 ± 0.001	0.118 ± 0.001	0.091 ± 0.002	0.089 ± 0.002
Metal concentration/ g in catholyte tank	Au	0.00	0.498 ± 0.021	0.174 ± 0.015	0.00
containing 2.0 L io- dide electrolyte solu-	Cu	0.00	0.00	0.00	0.00

 Table 4: Metal Concentrations in Electrowinning Cell

Chemical analysis results of the 1.02 ± 0.006 g Au deposit obtained after the laboratory experiment showed that Au was the only metal, which was quantitatively detected in the deposit, constituting >99%. Hence, the total Au recovery yield observed was 95.50 %. The high Au recovery yield observed is comparable to, but slightly lower than the 98.75 % Au recovery yield observed by Meng et al.[12]. The lower Au recovery yield observed can be attributed to the loss of approximately 4 % of Au to the Au-IEM through incomplete elution/ stripping (this was confirmed by the presence of residual Au on the Au-IEM). In this instance, Au loss could be reduced by increasing the temperature or pH of the electrolyte solution in the catholyte tank. However, an increase in temperature and/or pH may have resulted in the degradation of the Au-IEM, due to a loss of mechanical strength in response to temperature changes, or osmotic shock in response to changes in pH [17].

In addition to the loss of Au to the Au-IEM, other drawbacks/ limitations of the investigated electrowinning process, include the high cell voltage employed and the long electrowinning time. The high 12.9 V cell voltage employed in the present work is significantly higher than the 2.1 - 5 V cell voltage applied in many industrial Au electrowinning processes (however it must be noted that these industrial electrowinning processes are rarely focused on Au recovery from leachates with dilute Au concentrations as in the present work). The long 6 hr. electrowinning time required to achieve a maximum 95.50 % Au recovery yield in the present work is triple the amount of time that was required by Meng et al. who used a similar electrowinning process to concentrate and recover Au from

J App Mat Sci & Engg Res, 2024

an iodide leachate with a maximum yield of 96% after 2 hrs [12]. The investigated membrane-based electrowinning process could be improved by overcoming the aforementioned drawbacks through process and configuration optimization. Process and configuration optimization could be achieved by conducting additional experiments investigating how Au recovery yield is affected by changes in operating conditions (e.g., cell voltage, temperature, etc.) or electrowinning cell configuration (e.g., electrode spacing, membrane contact surface area, etc.). Findings from these additional experiments could be used to design a more improved process which is able to concentrate and recover Au with a greater yield (>95.50 %), using a lower cell voltage (< 12.9 V), and in a shorter time (< 4 hrs).

4.1.2 Iodide Recovery

One of the key advantages of using electrowinning techniques for Au recovery from iodide leachates, is that such techniques allow for the simultaneous recovery of Au and iodide solution, in a few process steps, and in an environmentally benign manner, without the use of hazardous chemical regents or the production of hazardous waste [12]. Recovered iodide solution can then be re-used in subsequent process cycles to reduce costs. In the present work, approximately 2.0 L of iodide (I₂ and I) solution was recovered after the laboratory experiment from the anolyte tank, as a dark purple liquid. Chemical analysis results found that the recovered iodide solution had a 6.11 \pm 0.85 g/L I₂ concentration and 26.77 \pm 3.73 g/L I- concentration. This implies that approximately 82 % of initial I2 and 89 % of initial I⁻ was recovered (this finding is comparable to the 83 % I₂ recovery yield obtained by Meng et al., 2021). In addition to the recovered iodide solution, 2.0 L of clear aqueous solution/ water with a Cu (as Cu_2^+) concentration of 89.07 ± 1.79 mg was recovered from the feed tank, while 2.0 L of clear di-ionised water was recovered from the catholyte tank.

4.1.3 Flux

The laboratory experiment results revealed that the investigated novel membrane-based electrowinning process was able to concentrate and recover a maximum of 1.02 g of Au from 2.0 L of WPCB iodide leachate after 6.0 hrs, as a deposit on the stainlesssteel cathode in the electrowinning cell. Consequently, the average flux of Au ions through the 12 cm \times 12 cm Au-IEM was determined to be 3.27 mg/m².s or $8.4 \times 10-6$ mol/m².s. This flux is comparable to the $5.5 \times 10-6$ to $14.8 \times 10-6$ mol/m2.s Au flux reported by Sun et al., who studied the flux of Au ions through an ion exchange membrane comprised of a polyvinylidene fluoride back bone and [A336][SCN] carrier, during the recovery of Au in cyanide solution using a technique similar to the investigated membrane based electrowinning process (the process applied by Sun et al. is primarily differentiated by the absence of an AEM, and by the use of only a single electrolyte solution tank which hosted both the anode and cathode) [23]. Any future scale-up of the novel membrane-based electrowinning process must take this flux into account. Hence, there is a need to develop a more efficient and less problematic process for Au recovery from WPCB iodide leachates. The present work uses a laboratory experiment to investigate the feasibility of Au recovery from WPCB iodide leachates using a novel membrane-based electrowinning process.

5. Conclusions

The present paper reported a feasible process for Au recovery from WPCB iodide leachates using a novel membrane-based electrowinning process incorporating an Au selective ion exchange extractant membrane. It was concluded that:

• High purity (>99%) Au can be effectively recovered from WPCB iodide leachates with a 95.50 % recovery yield using the novel membrane-based electrowinning process.

• Au recovery was achieved under the following conditions: 6.0 hrs time, constant cell voltage of 12.9 V, temperature of 25 °C, 250 rpm agitation speed, average current of 5 A, 1429 A/m² equivalent current density, and 3.27 mg/m2.s or 8.4×10^{-6} mol/m2.s Au flux.

+ 82 % of initial $\rm I_2$ and 89 % of initial $\rm I^-$ was recovered simultaneously with Au.

Data availability

Data will be made available on request.

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Author Contributions

M.M. and J.P. designed the research; M.M. performed the research, analysed the data, and wrote the paper; J.P. supervised the research; and M.T co-supervised the research.

Competing Interests

The authors declare no competing interests.

References

- 1. Gloe, K., Mühl, P., & Knothe, M. (1990). Recovery of precious metals from electronic scrap, in particular from waste products of the thick-layer technique. *Hydrometallurgy*, 25(1), 99-110.
- 2. Ashiq, A., Kulkarni, J., & Vithanage, M. (2019). Hydrometallurgical recovery of metals from e-waste. In *Electronic waste management and treatment technology* (pp. 225-246). Butterworth-Heinemann.
- Zhang, H., Jeffery, C. A., & Jeffrey, M. I. (2012). Ion exchange recovery of gold from iodine-iodide solutions. *Hydrometallurgy*, 125, 69-75.
- 4. Singh, S., & Prasad, B. L. V. (2007). Nearly complete oxidation of Au° in hydrophobized nanoparticles to Au3+ ions by N-bromosuccinimide. *The Journal of Physical Chemistry C, 111*(39), 14348-14352.
- Galva, 2023. Galva, Bibliographic References, http://www.galva.com.br> (access on
- 6. 20.01.23).
- Kasper, A. C., Veit, H. M., García-Gabaldón, M., & Herranz, V. P. (2018). Electrochemical study of gold recovery from ammoniacal thiosulfate, simulating the PCBs leaching of mobile phones. *Electrochimica Acta*, 259, 500-509.
- 8. Petter, P. M. H., Veit, H. M., & Bernardes, A. M. (2014). Evaluation of gold and silver leaching from printed circuit board of cellphones. *Waste management*, *34*(2), 475-482.
- Sahin, M., Akcil, A., Erust, C., Altynbek, S., Gahan, C. S., & Tuncuk, A. (2015). A potential alternative for precious metal recovery from e-waste: iodine leaching. *Separation Science and Technology*, 50(16), 2587-2595.
- 10. Zheng, S., Wang, Y. Y., & Chai, L. Y. (2006). Research status and prospect of gold leaching in alkaline thiourea solution. *Minerals engineering*, *19*(13), 1301-1306.
- 11. Xu, Q., Chen, D. H., Chen, L., & Huang, M. H. (2010). Gold leaching from waste printed circuit board by iodine process. *Nonferrous Metals*, 62(3), 88-90.
- Altansukh, B., Haga, K., Huang, H. H., & Shibayama, A. (2019). Gold recovery from waste printed circuit boards by advanced hydrometallurgical processing. *Materials Transactions*, 60(2), 287-296.
- Meng, Q., Li, G., Kang, H., Yan, X., Wang, H., & Xu, D. (2019). A study of the electrodeposition of gold process in iodine leaching solution. *Metals*, 10(1), 50.
- 14. Medina, D., & Anderson, C. G. (2020). A review of the cyanidation treatment of copper-gold ores and concentrates. *Metals*, *10*(7), 897.
- 15. Deschenes, G. (2005). Advances in the cyanidation of gold. *Developments in Mineral Processing, 15*, 479-500.
- Křivčík, J., Neděla, D., & Válek, R. (2015). Ion-exchange membrane reinforcing. *Desalination and Water Treatment*, 56(12), 3214-3219.

- 17. Deventer, J. V. (2014). New Developments in Ion Exchange Resins for the Recovery
- 18. of Gold in Complex Ores. Originally presented at Hydrometallurgy 2014. *Canadian Institute of Mining, Metallurgy & Petroleum.* 677-687.
- Zaheri, P., & Davarkhah, R. (2020). Selective separation of uranium from sulfuric acid media using a polymer inclusion membrane containing alamine336. *Chemical Papers*, 74, 2573-2581.
- Mecucci, A., & Scott, K. (2002). Leaching and electrochemical recovery of copper, lead and tin from scrap printed circuit boards. *Journal of Chemical Technology* & *Biotechnology: International Research in Process, Environmental & Clean Technology*, 77(4), 449-457.
- Jha, M. K., Kumari, A., Choubey, P. K., Lee, J. C., Kumar, V., & Jeong, J. (2012). Leaching of lead from solder material of waste printed circuit boards (PCBs). *Hydrometallurgy*,

121, 28-34.

- 22. Behnamfard, A., Salarirad, M. M., & Veglio, F. (2013). Process development for recovery of copper and precious metals from waste printed circuit boards with emphasize on palladium and gold leaching and precipitation. *Waste management*, 33(11), 2354-2363.
- Ficeriová, J., Baláž, P., & Gock, E. (2011). Leaching of gold, silver and accompanying metals from circuit boards (PCBs) waste. *Acta Montanistica Slovaca*, 16(2), 128.
- 24. Yang, H., Liu, J., & Yang, J. (2011). Leaching copper from shredded particles of waste printed circuit boards. *Journal of hazardous materials*, 187(1-3), 393-400.
- 25. Sun, Y., Wang, Z., Wang, Y., Liu, M., Li, S., Tang, L., ... & Ji, S. (2020). Improved transport of gold (I) from aurocyanide solution using a green ionic liquid-based polymer inclusion membrane with in-situ electrodeposition. *Chemical Engineering Research and Design*, 153, 136-145.

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