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Effect of Reaction Temperature on Noble Metal Regeneration from E-Waste Through Hydrometallurgical Process by using HCl-H_2O_2 Leaching Agent

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Abstract: Noble metal comes with many applications, including in catalysis, electronic devices and jewellery. However, its limited resources are becoming depleted, in order to secure future needs and cost-saving, it was necessary to process spent noble metal-containing materials, such as electronic scraps (e-waste) i.e. computer, mobile phone and radio. This research carried out to study the recycling noble metal from e-waste. It was believed that hydrometallurgical processes through hydrochloric acid and hydrogen peroxide (HCl-H_2O_2) as a leaching agent, potentially enhance the noble metal regeneration rate. For that purpose, the reaction temperature effect on the optimum leaching operation condition and noble metal yield % per batch of e-waste raw of extracted Platinum (Pt), Ruthenium (Ru) and Rhodium (Rh), needs to be defined in order to evaluate the contribution of hydrochloric acid-hydrogen peroxide (HCl-H_2O_2) as a leaching agent in hydrometallurgical process. The experimental works are covering from e-waste preparation stage (selection, segregated, crushed and ground), leaching process at reaction temperature from 50 °C to 80 °C, a retention time of 2 hours at concentration ratio HCl-H_2O_2 of 10:1. Followed by the purification stage of filtration, washing and drying. The Scanning Electron Microscope with Energy Dispersive X-Ray (SEM_EDS) analysis were used to characterized the composition and weight %. From SEM_EDS analysis, Pt showed the highest recovery yield of 1.6 weight % and regenaration rate of 0.196 g/s. The Rh showed the thermodynamic activation energy pattern for extraction which begin with the lag phase and moving forward for the exponential phase. The outcome of this work has the potential to scale up for noble metal regeneration of future demands especially in any region with the abundant of e-waste.

Keywords: Noble Metal, Hydrometallurgical, Leaching Agent, Reaction Temperature

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1. Introduction

Electronics waste such as computer, mobile phone and radio, commonly known as e-scrap or e-waste, is the trash we generate from surplus, broken, and obsolete electronic devices. Electronics contains various toxic and hazardous chemicals and materials that are released into the environment if do not dispose of them properly. E-waste or electronics recycling is the process of recovering material from old devices to use in new products. Figure 1 shows studies conducted in Malaysia the quantity of e-waste generate in 1995 reach 15 million pieces and increase 44 million pieces in 2015 and estimated to reach 53 million pieces in 2020, which is 3.5 times higher than 1995 [1-3].

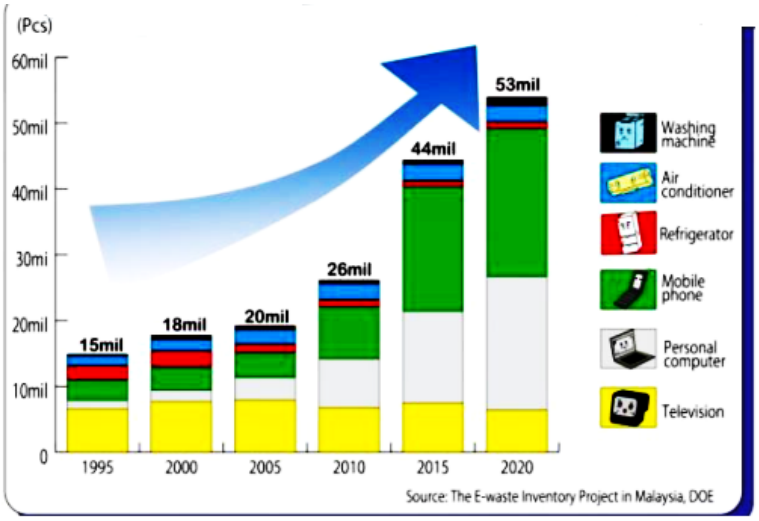

Figure 1: Data of E-waste from 1995-2020 [1]

Platinum group metals (PGMs) play a key role in modern society, as they are of specific importance for clean technologies and other high-tech equipment. Important applications beyond the well-known areas of chemical process catalysis and automotive emissions control include information technology (IT), consumer electronics, and sustainable energy production such as photovoltaics (PV) and fuel cells, among others Figure 2. Driving forces for the booming use of pgms are their extraordinary and sometimes exclusive properties, which make them essential components in a broad range of applications which can play a part in building a more sustainable society [2-4].

The following Table 1 presents an overview of the current end-of-life recycling rates for 60 metals. These ranges of values represent the global situation ,and include all users of the metals. The recycling situation for precious metals (platinum, palladium, gold, silver) and cobalt is significantly better with rates above 50.00 % [5-8].

Table 1 : Important application for PGMs [2]

Some Important Application Areas for Platinum Group Metals

Application area	Platinum group metal				
	Platinum	Palladium	Rhodium	Iridium	Ruthenium
Catalysts	✓	✓	✓	✓	✓
Electronics	✓	✓		✓	✓
Fuel cells	✓	✓	✓		✓
Glass, ceramics and pigments	✓		✓		
Medical/dental	✓	✓		✓	
Pharmaceuticals	✓	✓			✓
Photovoltaics					✓
Superalloys					✓

Various investigators studied the extraction of PMs, copper, lead and zinc from e-waste using hydrometallurgical routes. These routes are based on traditional hydrometallurgical technology of metals extractions from their primary ores. Similar steps of acid or caustic leaching are employed for the selective dissolution of PMs from e-waste. The pregnant solution is separated and purified for the enrichment of metal content thereby impurities are removed as gangue materials. The isolation of metal of interest is conducted through solvent extraction, adsorption, and ion exchange enrichment processes. Finally, metals are recovered from the solution through electrorefining (electrometallurgy) or chemical reduction processes. Hydro- and pyrometallurgical processes were evaluated and discussed. It has been reported that hydrometallurgical processes have additional benefits compared to pyrometallurgical processes because they are more exact, predictable, and easily controllable [7-10].

Other investigators have been carrying out studies on the recovery of PMs from e-waste at laboratory scale. It is noted that HNO_3, H_2SO_4, and HCl-based solutions are commonly employed for dissolving PMs from e-waste. From the leachants, PMs are recovered employing methods similar to those used in the mineral industry [11-12].

2. Materials and Methods

The precious metal is recovered and purified via chemical (metallurgical) processing to ensure that the extracted yield can be acceptable for its original usage. Recovered precious metals via chemical (metallurgical) techniques process, including pyrometallurgical, hydrometallurgical and biometallurgical processing for removal of impurities. A few series of the chemical reaction is involved during this metallurgical process [5-6].

2.1 Leaching in Aqua Regia Solutions

The leaching of metal is mainly governed by the formation of a complex under the potential and pH of the solution in the presence of ionic species. As PGMs forms stable chloro-complexes in chloride solution at high acid concentration, PGMs could be leached at a higher potential using an oxidizing agent. The other associated metals (Pd, Rh, etc.) also form chloro-complexes at lower potential and will also be leached, but the Rh leaching rate is relatively low. The oxidizing agent, HNO_3, is suitable for Pt leaching because of its high oxidation potential of 0.96 V according to Equation 1. In the case of metal dissolution with aqua regia, the formed nascent chlorine (Cl_2) and nitrosyl chloride (NOCl) provide high oxidation potential and the high chloride-ion concentration acts as the complexing agent. HNO_3 and HCl undergo through Equation 2 and Equation 3 [7,10 -11].

$$NO_3^- + 4H+ + 3e- \leftrightarrow NO + 2H_2O \quad \text{(Eq.1)}$$

$$HNO_3 + 3HCl \leftrightarrow NOCl + Cl_2 + 2H_2O \quad \text{(Eq.2)}$$

$$2NOCl(g) \rightarrow 2NO(g) + Cl_2(g) \quad \text{(Eq.3)}$$

2.2 Leaching within Hydrogen Peroxide

Replacing aqua regia with HCl and H_2O_2 lixiviant for the dissolution of Pt from spent catalytic converters, can achieving a recovery of 95.00 % Pt under the optimized lixiviant concentration with the oxidizing agent in a 10 HCl:1 H_2O_2 solution. The process could be easily adaptable for industrial applications under moderate experimental conditions. the process for the dissolution of PGMs, including Pt, from different types of car catalytic converter catalysts with honeycomb monolith, supports with wash-coats onto which the PGMs are adsorbed. The ground sample was thermally pre-treated at 250 °C for 22 h before leaching in HCl and H_2SO_4 in the presence of oxidizing agents, hydrogen peroxide and nitric acid. The fluoride ion was also employed by adding NH_4F, which improved the dissolution efficiency by attacking the interface between the PGMs and the catalyst substrates. A recovery of 95.00 % was obtained by leaching at 90 °C for 6 h after thermal pretreatment

with both oxidizing agents. Other associated metals, Pd and Rh are also leached in the presence of H_2O_2. The presence of H_2SO_4 as a proton source also improves the leaching of Rh. The process is relatively environmentally friendly because Pt dissolution in this solution yields less hazardous gases [6-7,11].

$$Pt + 6HCl + 2H_2O_2 \leftrightarrow [PtCl_6]^{2-} + 2H+ + 4H_2O \quad \text{(Eq.4)}$$

2.3 Material

The platter HDD that was chosen for this research. The selection of the platen because of the noble metal element that has in the material. This material has been compared to other e-waste materials by studying the percentage of Pt, Rh, and Ru present in the e-waste. This material also has a high potential for the leaching process [6-7,11].

2.4 Intrumentation Analysis

The research applied X-ray testing to determine the mechanical properties in the sample. X-ray testing made to know whether that sample has platinum, ruthenium and rhodium or not. Using this testing we can know the element, mass of element, component that has in the sample and gets analysis of the sample. From this research, the sample has to inspection with instrumentation analysis it is EDS.

3. Results and Discussion

The result covered the experimental and analysis of operating temperature effect on e-waste sample recovery by using hydrometallurgical process. The characterization analysis of noble metal yield and extraction recovery rate using SEM_EDS. For this works, Platinum (Pt), Ruthenium (Ru) and Rhodium (Rh) is the main focused of noble metal's element due to its various application and at most expensive price.

3.1 Effect of Reaction Temperature on Noble Metal Yields

The temperature is the most important parameters for metal leaching. Increase in the temperature of the solution increases its reactive activity. The noble metal extraction kinetic studies were conducted at four temperature levels of 50 °C, 60 °C, 70 °C, and 80 °C. For the operating condition at a constant time of 2 hours and leaching agent concentration ratio of 10:1 hydrochloric acid and hydrogen peroxide. The result of this extraction of noble metal Pt, Ru and Rh were presented as in Figure 2.

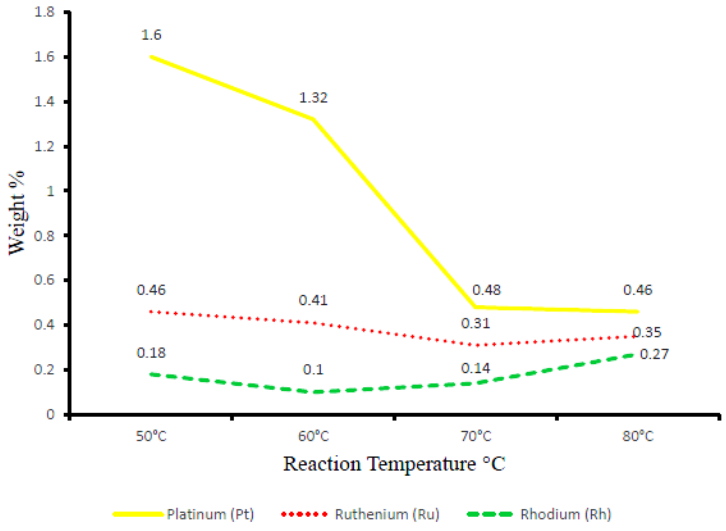

Figure 2: Effect of Reaction Temperature on Pt, Ru and Rh Yield

Figure 2 clearly shows that increasing temperature affect the extraction kinetics and recovery of Pt, Rh, and Ru. The large temperature impact must be due to relatively high activation energies for the

reaction. This experiment showed that the Platinum recovery is slightly high compare to the Ruthenium and Rhodium, due to the HCl - H_2O_2 leaching agent reaction properties selectivity. However, the reduction of increasing temperature would have an adverse effect on the sedimentation of the solid after leaching. The Platinum and Ruthenium recovery trend proportionally decreases with operating temperature starting from 60 ºC, this indicates that both element start decomposing at a higher temperature. These temperatures elevation showed that the free cyanide is hydrolysed and oxidized, and decomposed more quickly than the complexed cyanide Platinum and Ruthenium appeared to be in an easy to dissolve within leaching agent in the high temperature. Platinum yield slightly decreases from 1.60 % to 0.46 % at increasing temperature. Same goes to the Ruthenium, the yields decrease from 0.46 % to 0.35 % at temperature elevation.

Compare to the Rhodium, recovery trend is increasing proportionally to the operating temperature at 60 ºC, its believe that thermodynamic activation energy for extraction begins with the lag phase and moving forward for the exponential phase. From Figure 2, showed that during the reaction temperature is 50 °C, yield 0.18 %, and elevation until 80 °C, yield 0.27 %. The temperature elevation believed to affect the surface chemical reaction process where Rhodium and Oxygen need to absorbed on the surface sample as reported by [7] and 2014 [12].

3.2 Effect of Reaction Temperature on Noble Metal Regeneration Rate

Figure 3 shows the regeneration rate of Pt, Ru, and Rh. The regeneration rate of Platinum was decreased from 0.196 g/s at a temperature 50 °C to 0.056 g/s at a temperature 80 °C. Meanwhile the regeneration rate of Ruthenium also slightly degrease from 0.056 g/s at temperature 50 °C to 0.033 g/s at temperature 80 °C. Both elements is showing the same pattern because of the effect of solid sedimentation after leaching and straightly easy to decompose at higher temperatures.

For the regeneration rate of Rhodium, it decreases from 0.022 g/s at temperature 50 °C to 0.012 g/s at temperature 60 °C, and slightly increase to 0.033 g/s at a temperature of 80° C. The temperature elevation believed to affect the surface chemical reaction process where Rhodium and Oxygen need to absorbed on the surface sample

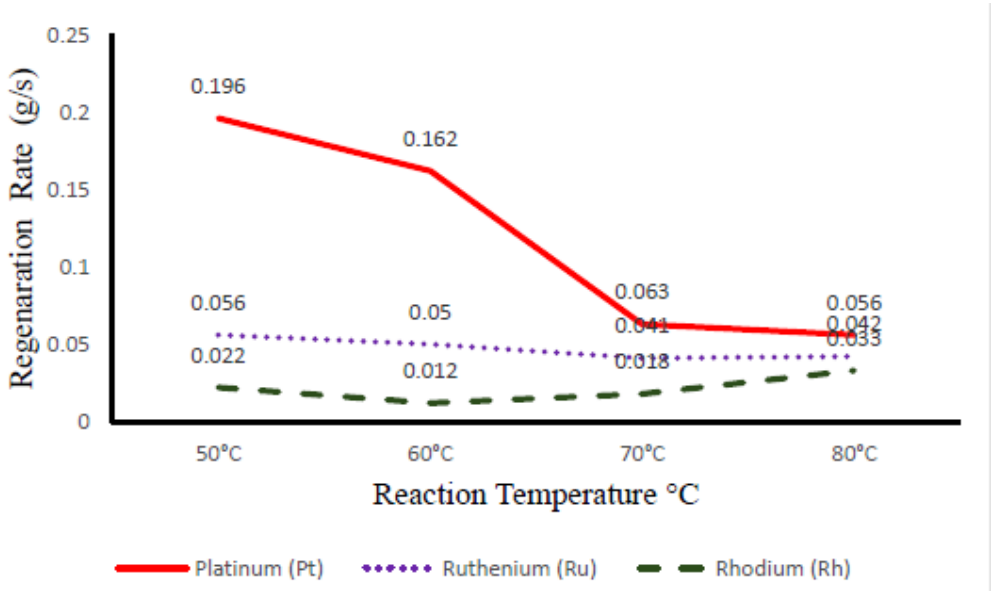

Figure 3: Effect of Reaction rate on Pt, Ru and Rh regeneration rate

4. Conclusion

From this research, the effect of reaction temperature on leaching operation condition of noble metal regeneration from e-waste via hydrometallurgical process by using HCl-H_2O_2 leaching agent was

successfully characterized. The main finding from this work is concluded. The Platinum showed the highest recovery yield of 1.6 weight % and regeneration rate of 0.196 g/s. The Rhodium showed the thermodynamic activation energy pattern for extraction begins with the lag phase and moving forward for the exponential phase. The outcome of this work has the potential to be further and scale-up for noble metal regeneration from e-waste via a hydrometallurgical process by using HCl-H_2O_2 leaching agent.

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