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SILVER LEACHING FROM PRINTED CIRCUIT BOARD USING DEEP EUTECTIC SOLVENT- OXALINE, MALINE, AND SUCCILINE

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Abstract

The exponential growth in printed circuit boards (PCBs) waste, presents an urgent environmental and economic challenge. PCBs contain silver which has a high potential economic value. The high production of waste and the potential economic value make it important to recycle PCB waste. Recycling can be conducted by leaching silver using Deep Eutectic Solvent (DES). DES is environmentally friendly and able to form complexes with various types of metals. This research aims to study the effect of the HBD carbon chain length of oxalic acid, malonic acid, and succinic acid from choline chloride-based eutectic solvents (DES) on the efficiency of leaching silver from Printed Circuit Board (PCB) waste. Optimization of the solid/liquid ratio, time, and temperature is carried out to obtain optimum leaching conditions. The synthesis results show that DES oxaline 2:1, malin 1:1, and succilin 2:1 have good stability, forming a colorless, clear, and viscous solution. FTIR analysis shows a shift in the wave number of the -OH group due to hydrogen bond interactions between HBA and HBD molecules in the formation of DES. The leaching yield of DES against standard silver oxide was measured using AAS. DES oxaline 2:1 has the greatest efficiency compared to other DES variations. The results of optimizing standard silver leaching with DES oxaline showed that the S/L ratio was 40 mg/mL, with a time of 18 hours, at a temperature of 60°C which resulted in a recovery percentage of 90.90%, while for the PCB waste sample yielded an efficiency of 99.38%. The eutectic solvent oxaline has the potential to be an environmentally friendly solvent that is efficient in leaching silver from PCB electronic waste.

Keywords:
Deep Eutectic Solvent;
Printed Circuit Board;
Oxaline; Leaching; Silver.

INTRODUCTION

The United Nations Institute reports that there are 1.2 million tons of PCB waste in the world, of which only 0.4 million tons can be handled. PCBs contain about 30% plastic, 30% ceramic, and 40% metal. PCBs have a very diverse metal content such as iron (Fe) 8- 38%, copper (Cu) 10-27%, aluminum (Al) 2-19%, tin (Pb) 0.3-2%, silver (Ag) 200-3000 ppm, gold (Au) 50-500 ppm, and palladium (Pd) 10-200 ppm. [1-2].

One way to recycle PCBs is to extract the valuable metals contained in PCBs, namely silver. In addition to addressing environmental concerns, recycling is also driven by economic interests. Moreover, the scarcity of precious metal reserves and their significant economic value serve as further motivations for intensifying the research of precious metals recovery from PCBs [3].

Metal recovery methods in PCBs generally use pyrometallurgy, hydrometallurgy, and solvometallurgy methods. Pyrometallurgy has the disadvantage that it can only be applied to metals with high concentrations and is carried out at a temperature of 1200°C [4]. Hydrometallurgy is more popularly used with the use of strong acid solutions because the extraction rate is high but not environmentally friendly [5-6]. Solvometallurgy is similar to hydrometallurgy in that the metal separation or extraction process is performed at low temperatures. Solvometallurgy uses anhydrous or low water content materials [7].

Ionic liquids (ILs) and Deep Eutectic Solvent (DES) are solvents used in solvometallurgy. Solvometallurgy is the solution to this problem. Deep Eutectic Solvent (DES) is a type of green solvent but greatly capable to extract metals [8]. DES are solvents formed by eutectic mixtures of Lewis or Brønsted acids and bases that can contain various anionic and/or cationic species, but also neutral compounds. A eutectic mixture is a specific composition of at least two solid components that results in a phase change to liquid at a certain temperature. This point is referred to as the eutectic point [9].

Various literatures show the ability of DES to dissolve various types of metal oxides with a large percentage of leaching [10]. DES based on ChCl:citric acid was able to leach cobalt (Co) by 99.6% [11], ChCl:ethylene glycol leached 89.91% of lithium (Li) and 94.14% of cobalt (Co) [12] ChCl:oxalic acid leached 90% of lithium (Li) and 96% of manganese (Mn) [13].

DES composed of Hydrogen Bond Acceptors (HBA) and Hydrogen Bond Donors (HBD) that form a liquid phase at room temperature and is able to form complexes with various types of metals or metal oxides. HBAs in DES are generally quaternary ammonium salts or phosphonium salts and HBDs are sugars, alcohols, carboxylic acids, or amino acids [10]. Based on the literature review, there are not many studies related to the use of choline chloride-based DES for silver metal leaching from PCB waste. This research aims to synthesize and determine the characteristics of choline chloride-based DES with oxalic acid, malonic acid, and succinic acid; study the effect of different carbon chain lengths of HBD on silver leaching; and determine the effect of solid/liquid ratio, leaching time and temperature on silver leaching efficiency.

EXPERIMENT

Material

Materials used in this research include choline chloride ($[(CH_3)_3NCH_2CH_2OH]^+Cl^-$ ≥99%), oxalic acid ($C_2H_2O_4{\cdot}2H_2O$), malonic acid ($C_3H_4O_4$ 99%) and succinic acid ($C_4H_6O_4$ ≥99.0%), silver oxide >99%, and end-used computer PCB.

Instrumentation

Instrumentation used in this research include FTIR Spectroscopy 8400s (Shimadzu), AAS – GBC Scientific – Avanta, XRF Portable Thermo Scientific Niton type XL3t 500 Analyzers.

Procedure

To achieve the optimum condition, DES of ChCl:Oxa, ChCl:Mal, and ChCl: Suc synthesized with mole ratios of 1:1; 1:2; and 2:1 and heated at 100°C with a stirring speed of 500 rpm until a homogeneous solution formed [14]. DES which is liquid at room temperature, clear and viscous will be selected as the silver leaching agent on Ag_2O and PCB. A total of 20 mg of standard Ag_2O was dissolved in 1 mL of DES solution and then heated at 50°C for 24 hours with a stirring speed of 500 rpm [15]. The leachate was subsequently filtered and pH-adjusted for quantitative assessment using atomic absorption spectroscopy (AAS) calibrated with standard silver solutions. Detection involved analyzing absorbance at 328.1 nm via flame or graphite furnace AAS, validated by blanks and interpreted through a calibration curve to see the concentration of leached silver. The DES with the highest percent leaching was then selected and optimized with variables of solid/liquid ratio, leaching temperature, and leaching time.

RESULT AND DISCUSSION

Synthesis of DES-Oxaline, Maline, and Succiline

DES are composed of choline chloride (ChCl) as hydrogen bond acceptor (HBA) with carboxylic acid as hydrogen bond donor (HBD) [10]. DES formation is based on the phase equilibrium between HBA and its constituent HBDs. This phase equilibrium is called the eutectic point, where the eutectic point is the point where HBA and HBD will form a homogeneous liquid with a low melting point. The type of DES chosen in this study is a type 3 DES with the formula Cat^+X^-zY, where Cat^+ is a cation (e.g., ammonium, phosphonium, sulfonium) with X^- being a Lewis base that serves as an opposing anion (mostly halide anions) and Y is a Lewis or Brønsted acid from the z molecule that interacts with the X^- anion [16].

The HBA used is a quaternary ammonium salt in the form of choline chloride (ChCl) with carboxylic acid HBD in the form of oxalic acid, malonic acid, and succinic acid. These three compounds have different chain lengths. This chain length difference was analyzed to see its effect on the silver leaching efficiency of the standard and sample. DES was successfully synthesized at 100°C for 2 hours of heating with a stirring speed of 500 rpm [17]. In the heating process, all types of DES are clear, colorless, and viscous liquids except for succinic acid which is a white, turbid and viscous liquid. Oxaline 2:1, maline 1:1, and succiline 2:1 DES are stable at room temperature. Meanwhile, oxaline 1:1 and 1:2, maline 2:1, and succiline 1:1 and 1:2 were unstable at room temperature, formed crystals and solidified after 1-7 days. The instability of DES indicates that its melting point is higher than DES which is more stable at room temperature (25°C). **Table 1** shows the DES synthesis results of the three types of DES.

Table 1. Synthesis of DES using Choline Chloride with various carboxydioic acids at various mole ratio.

HBA	HBD	Mole ratio	Physical Properties
Choline Chloride	Oxalic Acid	1:1	Crystal-formed
	Malonic Acid		Clear, slightly yellow-colored and highly viscous solution
	Succinic Acid		Solid
	Oxalic Acid	1:2	Solid
	Malonic Acid		Crystal-formed
	Succinic Acid		Solid
	Oxalic Acid	2:1	Clear colorless and viscous solution
	Malonic Acid		Solid
	Succinic Acid		Clear colorless and viscous solution

The component's composition becomes an important factor in this synthesis as it can affect the properties of DES such as density, viscosity, electrical conductivity, thermal stability, and the reaction mechanism [18]. However, the composition of DES components depends on the combination of HBA and HBD. This is because the individual combinations produce DES with specific properties and characteristics [16]. Furthermore, DES-oxaline with a mol ratio of 2:1, DES-Maline with a mol ratio of 1:1, and DES-Succiline with a mol ratio of 2:1 was used for further steps and characterizations in this study.

Density of DES

The density of these DES was analyzed using a pycnometer. Most eutectic solvents have a higher density than water with values ranging from 1.0 to 1.3 g.cm^{-3} at 25 °C [19] as well as ranging from 1.1 to 1.25 g.cm^{-3} from computational prediction results [20]. **Table 2** shows the experimental DES density. The density of DES is depent on temperature and decreases linearly with increasing temperature [5,20-21]. In addition, the density also depends on the selection of hydrogen bond donor (HBD) [5,21-22]. and mole ratio [23]. Density also affects the efficiency of the leaching process. The higher the density, the lower the leaching efficiency [24].

Table 2. Measurement result of DES density (T =25 ^{0}C).

DES	Mole ratio	Densitas (g/cm^3)	
		Experimental result	Reference
Oxaline	2:1	1.21	-
Maline	1:1	1.26	1.234 [8,25-26]
Succiline	2:1	1.24	-

Fourier Transform Infra-Red Characterization of DES

The FTIR study was performed to elucidate the interaction between ChCl and glycerol after the mixing and heating processes. Based on the FTIR spectrum depicted in **Figure 1**, the formation of DES can be seen from the -OH group in oxalic acid that interacts with Cl$^-$ ions forming intermolecular hydrogen bonds. In oxalic acid -OH is at a wavelength of 3396.94 cm^{-1} while in DES there is a shift in wave number to 3369.75 cm^{-1}. It can also be seen that the -OH bond in malonic acid is at a wavelength of 3437.03 cm^{-1} and in DES it shifts to 3375.54 cm^{-1}. in succinic acid which was originally at a wave number of 3394.54 cm^{-1} to 3348.54 cm^{-1}.

Hydrogen bonding can make the electrostatic force slightly weakened which results in a shift in absorption. Hydrogen bonding can also result in band widening and decrease the intensity of absorption [27]. The shift in wave number is due to intermolecular interactions that occur in the formation of hydrogen bonds. If the absorption shifts towards a smaller wave number, it means that the bond to the group is getting stronger, while if the absorption shifts towards a larger number, it means that the bond strength is weakening. In the process of forming DES, this wave number shift occurs due to the strengthening of the OH----Cl bond and the weakening of the COOH bond in oxalic acid [28].

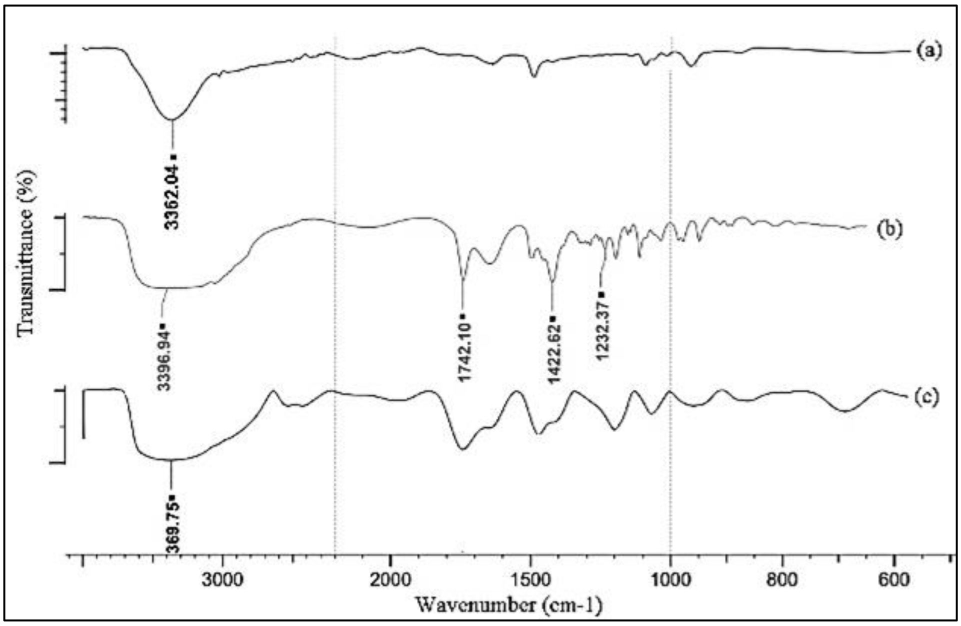

Figure 1. FTIR spectra of 2:1 oxaline DES, (a) choline chloride, (b) oxalic acid, and (c) oxaline DES.

This wave number shift also affects the stability of liquid phase liquids, decreasing the melting point and viscosity of oxaline [29]. The N$^+$ and Cl- parts form electrostatic bonds. From the results of the IR spectra analysis, an estimate of the interaction of hydrogen bond formation in oxaline DES is shown in **Figure 2** [30-31].

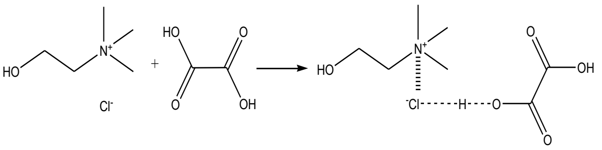

Figure 2 Reaction mechanism of hydrogen bond formation in Oxaline DES formation.

The same thing also occurs in the process of DES formation between choline chloride with malonic acid and succinic acid. It can be seen (**Figure 3**) that the -OH bond in malonic acid is at a wavelength of 3437.03 cm^{-1} and in DES it shifts to 3375.54 cm^{-1}. The C=O functional group in malonic acid is at a wavelength of 1734.19 cm^{-1}, C-O at 1185.70 cm^{-1} and -OH bending at 1401.96 cm^{-1}. Then in choline chloride the C-N functional group is at a wavelength of 1084.94 cm^{-1}. **Figure 4** shows the estimated interaction of hydrogen bond formation in malonic acid with choline chloride. Just like DES oxaline part of N$^+$ and Cl$^-$ ions form electrostatic bonds.

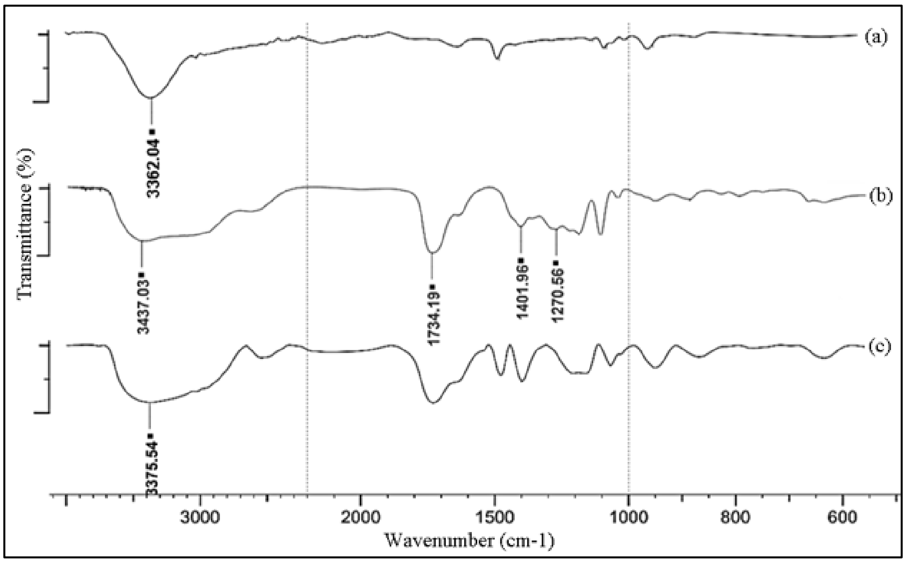

Figure 3. FTIR spectra of 1:1 Malin DES, (a) choline chloride, (b), malonic acid and (c) Malin DES.

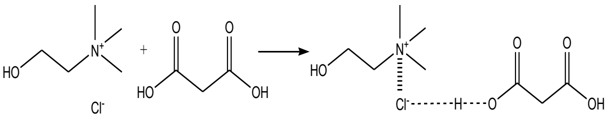

Figure 4. Reaction mechanism of hydrogen bond formation in the formation of DES Malin

The process of succinyl DES formation is also based on the intermolecular interaction between succinic acid and choline chloride on the OH group on succinic acid with Cl^- ions on choline chloride (**Figure 5**). This can be observed in the shift of the wave number of the -OH functional group on succinic acid which was originally at wave number 3394.54 cm^{-1} to 3348.54 cm^{-1}.

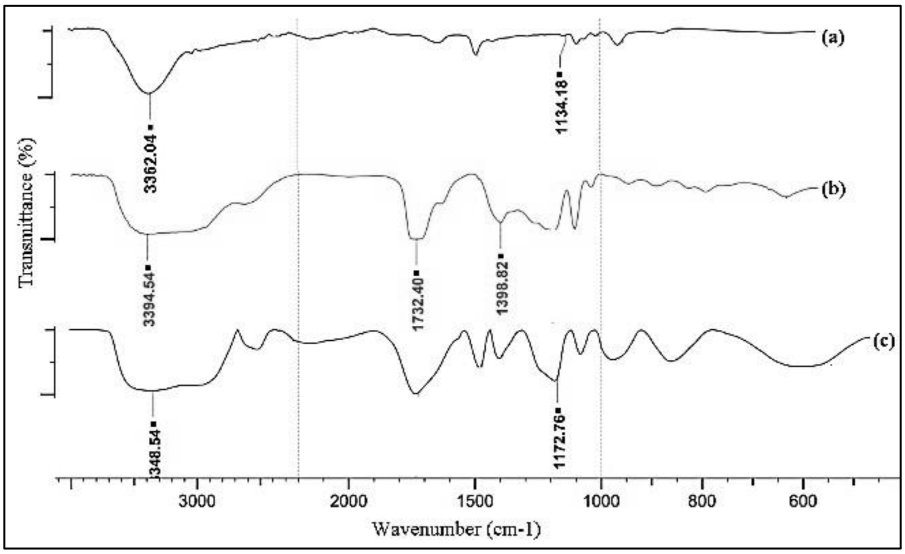

Figure 5. FTIR spectra of 2:1 Succilin DES, (a) choline chloride, (b) succinic acid, and (c) Succilin DES.

The -OH group was at wave number 3373.49 cm^{-1} for choline chloride, 3394.54 cm^{-1} in succinic acid, and 3348.54 cm^{-1} in succilin DES. Then in succinic acid, the C=O group is at 1732.40 cm^{-1} and -OH bending at 1398, cm^{-1}. **Figure 6** shows the estimated interaction of hydrogen bond formation in succinic acid with choline chloride.

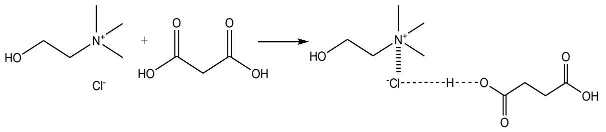

Figure 6. Reaction mechanism of hydrogen bond formation in the formation of succilin DES.

Application of DES to Standard Silver Oxide Leaching

DES that are stable at room temperature are then selected to leach silver oxide. This data will be used for analysis to see which DES is most efficient in leaching silver and to study the effect of carbon chain length of the hydrogen bond donor (HBD) on the percentage of silver oxide leached. The leaching conditions used were at 50°C, 24 hours with a solid/liquid ratio of 20mg/mL. The concentration of DES was measured using AAS, then the % leaching efficiency was calculated by the equation:

$$\%efisiensi = \frac{M_t - M_o}{M_t} \times 100 \qquad (1)$$

Where M_o is the measured silver concentration and M_t is the initial total silver concentration [32]. The percentage of leaching efficiency is shown in **Figure 7.**

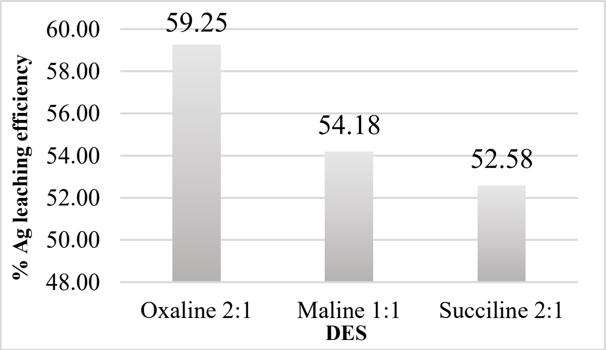

Figure 7. Silver leaching efficiency on Ag_2O using carboxylic acid DES.

Based **Figure 7**, 2:1 oxaline DES has the best leaching efficiency with % efficiency of 59.25%. This data shows that HBD with the shortest chain length has the best leaching ability. This is attributed to the proton activity and acidity of Oxalic Acid. The solubility of metal oxides is affected by proton activity in DES [33]. The solubility of metal oxides is also affected by the hydrogen bond donor coordination (HBD) strength of DES, where oxalate has the ability to release the most H protons and affects the acidity of DES [33-34]. Oxalic acid has a high acidity level compared to malonic acid and succinic acid. This acidity can be seen from the pKa values of the three acids, which are 1.25 for Oxalic Acid, 2.54 for malonic acid, and 4.21 for succinic acid.

Besides being influenced by the acidity of the DES constituent components, the leaching efficiency is also influenced by the DES density. Metal leaching efficiency decreases as the density increases due to an increase in diffusion resistance of metal ions within the solution, and a decrease in the cleavage of metal oxides by the leaching agent [35-37]. These factors reduce the rate and extent of metal dissolution, which results in lower leaching efficiency. Therefore, lower densities are usually more efficiently used for metal leaching. This statement is consistent with the experimental results which show that oxaline has the least density value compared to maline and succiline but has the greatest leaching efficiency.

Optimization of Silver Oxide Standard Leaching Conditions

Optimization of leaching conditions is carried out to obtain optimum conditions which will then be used when leaching PCB samples. Optimization of metal leaching using eutectic solvents aims to leach and separate metals from various sources in a more economical and environmentally friendly way compared to conventional solvents [7-8]. Optimization of metal leaching using eutectic solvents is performed for the best conditions such as temperature, time, concentration, and solvent composition to achieve the highest metal extraction efficiency [38-39]. There are three stages of leaching optimization, namely solid/liquid ratio, leaching time, and leaching temperature. Silver concentration measurements were made using AAS and the leaching efficiency was calculated by **equation 1**.

The leaching mechanism was obtained by plotting the leaching percentage (%) with different S/L ratios, time and temperature [11]. Solid/liquid ratio optimization is performed to obtain standard conditions used for extraction and is associated with process effectiveness [32]. In general, the higher the ratio of solids to liquid, the lower the metal leaching efficiency, due to a decrease in the decrease in contact area between the solid and liquid phases. These factors reduce the strength of the metal dissolution opportunity, which results in lower leaching efficiency. Therefore, the L/S ratio is usually preferred in metal leaching optimization [40-41]. The S/L optimization results shown in **Figure 8**, gave silver leaching efficiency results in the range between 93 and 98% with the optimum point of S/L ratio at 40mg/ml. This indicates that DES is able to leach silver maximally at a ratio of 40 mg/ml.

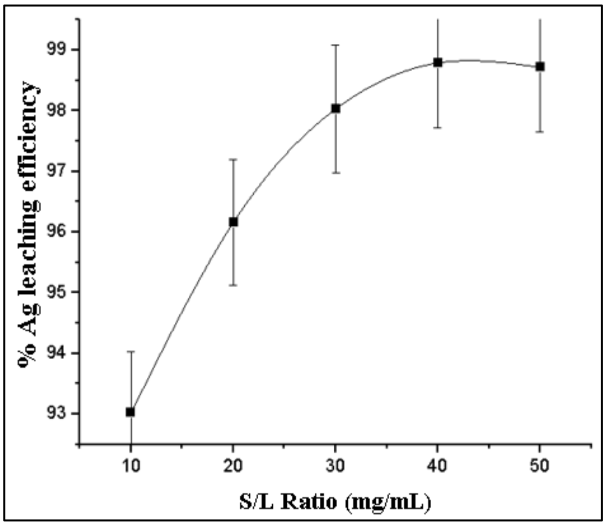

Figure 8. Silver S/L optimization results using oxaline DES.

This optimum point of S/L ratio is then used in time optimization. The optimum leaching time using eutectic solvents depends on various factors such as solvent type and composition, leaching temperature and pressure, and metal oxide concentration [19]. Generally, the longer the leaching time, the higher the metal leaching efficiency, until an optimum time is reached which slopes down indicating that no more metal can be extracted/leached [24,42]. **Figure 9** shows the results of the leaching time estimation with the

%leaching efficiency that increases with increasing time and begins to slope after 18 hours of leaching process with a %efficiency of 99.88% so that the 18-hour time is determined as the optimum time for silver leaching.

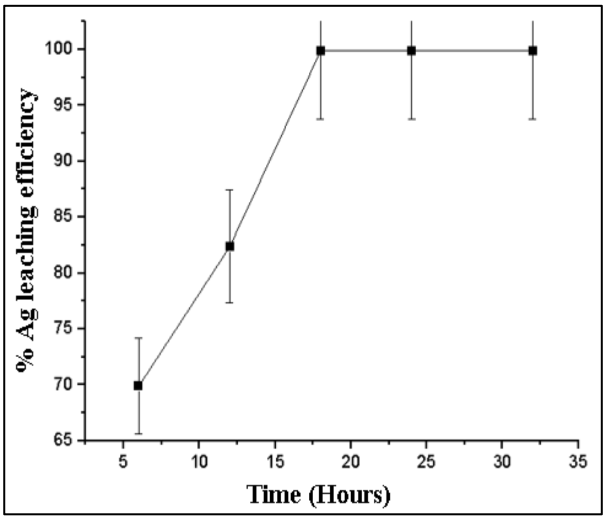

Figure 9. Results of silver leaching time optimization using oxaline DES.

The optimum points of S/L ratio and leaching time were then applied in the temperature optimization process. Temperature optimization was varied at 40, 50, 60, 70, and 80°C to see the relationship between temperature and the increase in leaching efficiency. In general, the higher the temperature the higher the metal leaching efficiency, due to an increase in the reaction rate between the solid and liquid phases, an increase in the solubility of metal ions in solution, and an increase in complex formation from metal oxidation [43-44]. It can be seen in **Figure 10** that the leaching efficiency increases with increasing temperature. At 60°C the leaching efficiency was 99.90% and at 70°C it only increased by 0.01% to 99.91%. this indicates that the optimum point of leaching is at 60°C.

In addition to the S/L ratio, leaching time and temperature, the stirring rate also affects the efficiency. The higher the stirring rate, the higher the metal leaching efficiency, due to an increase in mass transfer between the solid and liquid phases and a decrease in the diffusion resistance of metal ions in the solution [45-46]. These factors increase the rate and extent of metal dissolution, resulting in higher leaching efficiency. However, very high stirring rates can have a negative effect on metal leaching as they reduce the contact time between the target metal and the leaching agent [47-48]. In this study, the stirring rate refers to several previous studies, namely using a rate of 500 rpm [15]. In this strirring rate, optimal leaching effectiveness is achieved. From the optimization results it can be concluded that the optimum conditions for silver leaching are at a ratio of 40mg/mL with a leaching time of 18 hours at 60°C.

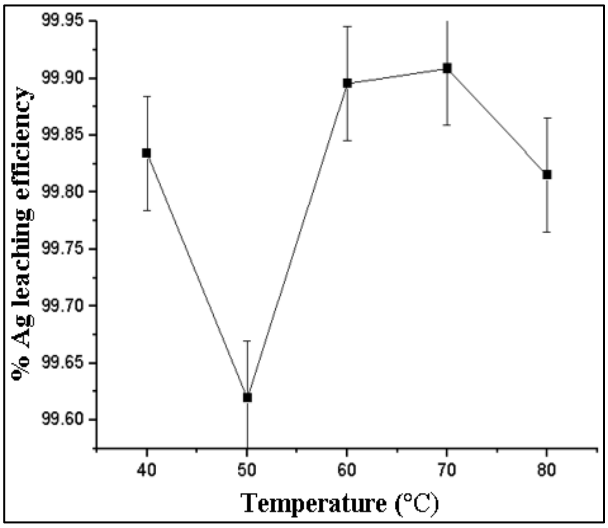

Figure 10. Silver leaching temperature optimization results using oxaline DES.

Application of DES in PCB Leaching

PCB samples that will be analyzed are first prepared by removing components, cut PCB boards into smaller sizes, followed by calcination at 600° C until the PCB sample is black powder. This PCB sample was analyzed using XRF (X-ray fluorescence) to see the metal content in PCB. **Table 3** shows the results of PCB sample analysis.

Table 3. XRF analysis results of PCB samples.

Element	Mass percent (%)
Copper (Cu)	32.64
Aluminum (Al)	8.26
Silver (Ag)	3.13
Iron (Fe)	0.29
Calcium (Ca)	20.12
Silicon (Si)	35.54

Silver leaching was conducted using PCB samples to find out whether there is alignment between the leaching results on Ag_2O standards and

PCB samples using optimum conditions due to PCB samples containing a more complex matrix because they contain various heavy metals. The leaching process was conducted using DES Oxaline 2:1, Maline 1:1 and Succiline 2:1 at optimum conditions, namely with a S/L ratio of 40 mg/mL at 60°C for 18 hours and a stirring speed of 500 rpm.

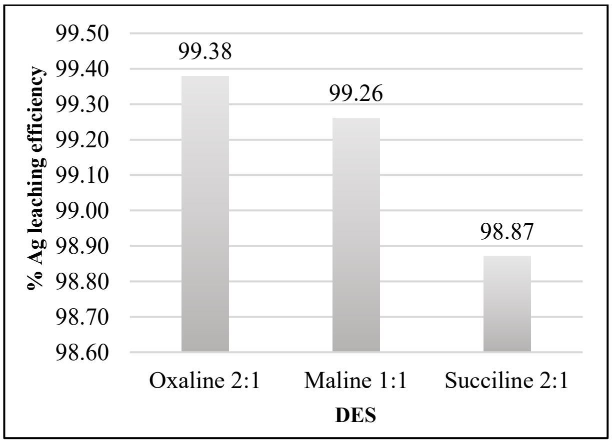

Figure 11. Silver leaching efficiency of PCB samples using DES.

DES oxaline 2:1, this result is in line with silver leaching efficiency data on Ag_2O standards which show that the length of the HBD chain greatly affects the silver leaching efficiency. The results of leaching using DES oxaline had a % efficiency of 99.38%, while in Malin 1:1 it was 99.26% and succilin 98.87%. This data shows that in a more complex matrix, DES still functions well as a silver leaching agent. It can be concluded that DES has the potential as an efficient environmentally friendly solvent to leach valuable metals specifically from PCB waste.

CONCLUSION

Based on the research that has been done, it can be concluded, DES synthesis was successfully conducted at 100°C for 2 hours with stirring speed of 500 rpm. DES with the best stability are Oxaline DES with 2:1 ratio, Malin DES with 1:1 ratio, and Succilin DES with 2:1 ratio. The leaching result towards Ag_2O and PCB standard, DES with the best % leaching is oxaline DES with 2:1 ratio, it shows that the chain length affects the leaching efficiency. Optimization results showed that the optimum S/L ratio was 40mg/mL, leaching time was 18 hours at 60°C with leaching efficiency of 99.90% on Ag_2O standard and 99.38% on PCB.

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