Removal of Precious Metals from Electronic-Waste by Using Composite Material

Raghad Hamid Hilal
Department of Production Engineering & Metallurgy University of Technology – Baghdad-Iraq
E-mail: raghadhilal4@gmail.com

Abstract: In this research, in-situ polymerization is used to prepare composite material at the same ratio of (polyacrylic acid to bentonite) and (10ml) aquaregia (1:3 HNO₃ : HCl), accurately about (5g) of the electronic waste (e-waste) powder is dissolved. Adsorption efficiency of (Polyacrylic acid - bentonite) composite to precious metals from neutral (e-waste) solution via forth flotation method, taking various weights (2.5, 5, 10 g.l⁻¹) of adsorbent material (polyacrylic acid - bentonite) composite at different temperatures (25, 35 and 45) °C. Results show that adsorption efficiency of Au⁺ is higher than Ag⁺ from adsorbent material and the highest value is (53.238%, 43.3904%), (51.778%, 41.99%) and (51.278%, 39.595%) for (Au⁺, Ag⁺) at (25, 35 and 45) °C respectively. But it is decreased slightly with increasing temperature.

1. Introduction
Environmental pollution by heavy metals has accelerated dramatically during the last few decades because the link minerals with industrial development such as mining, metallurgical, electronic, electroplating and metal finish [1]. Therefore, it is necessary to treat metal contaminated prior to its discharge to the environment. In recent years; adsorption has become one of the alternative treatments to search for low-cost adsorbents that have metal-binding capacities have intensified [2]. Clay minerals are added to polymers and the challenge in the synthesis of this kind of composites is combining a hydrophilic layered material with a hydrophobic organic part [3]. Precious metals, such as gold (Au), silver (Ag), platinum (Pt) are widely used in various industries such as: jewelry, electronics and others. Also, it has appeared different properties, like high resistance to chemical attack, excellent high-temperature characteristics [4]. Several methods based on pyro metallurgy and hydrometallurgy is currently used for the recovery of metals from waste printed circuits boards (PCBs). Pyro metallurgical processes require heating the waste at high temperatures to recover valuable metals [5]. Hydrometallurgical is a successful processes to leaching, then ion-exchange to gold adsorbent from electronic waste solution [6, 7]. Lahtinen et al [8] investigated the selective nylon-12-based three-dimensional (3D)-printed scavenger objects for gold recovery directly from an aqua regia extract of a printed circuit board waste. Using the easy to handle and reusable 3D printed meshes or columns, gold can be selectively captured both in a batch and continuous flow processes by dipping the scavenger into the solution or passing the gold containing solution through the column.
[8]. The recycling of e-waste is important for resource and waste management. Pyro metallurgical, hydrometallurgical or a combination of both routes is used for recovering precious metals (PMs) from e-waste. Fundamentally, hydrometallurgical routes are similar to those used in the mineral industry, which include leaching and metal extractions from leachates. Pyro metallurgical routes are economical and eco-efficient for the recovery of precious metals (PMs) [9]. Chehade et al [10] used variable extraction process involved: Physical separation to size reduction to~1 mm diameter then corona electrostatic separator and the hydro-cyclone to separated metals from non-metals. Metal recovery by hydrometallurgical processing by dissolution of metals in sulphuric acid and aqua regia, results show the metal purities of up to 99.99%. Angela et al [11] studied the aims to characterize different types of PCBs from mobile phones in relation to the amount of gold contained, the results showed that the content of gold varied from 142 to 700 g/ton. The cyanide-based solution was able to extract 88% of the gold, while sodium and ammonium thiosulfate extracted 70 and 75% of the gold, respectively. The electro winning tests showed a 94% recovery of the gold present in thiosulfate solutions. Piotr N et al [12] in-situ polymerization technique was applied to synthesize various polymer composites of Poly (acrylic acid)/ montmorillonite (MMT) composites. In accordance with XRD and FTIR spectroscopy, limited quantities of hydrogel were inserted among the clay layers. Means part of polymer remained precipitated on the outer surface of clay particles. This resulted in the improvement of the adsorption ability of MMT to remove of Fe$^{3+}$ ions from aqueous solution. According to the chemical structure of the polymer and the clay type available methods have been developed to prepare polymer/clay nanocomposites by melt compounding, which is the easiest and most cost effective method to produce nanocomposite materials. The technique induces the formation of nanocomposites structure by subjecting shearing force at high temperature [13, 14]. Polyacrylic acid (PAA) possesses many polar carboxylic groups with ability to form strong complexes with metal ions in solution, although limited used in remove heavy metal ions from wastewater [15]. It is well known that an increase in temperature enhances the adsorption of the collector in chemisorbing-types of flotation systems. This is perhaps due to enhanced free energy in the solid–liquid system. Therefore, more active areas on the surfaces would be available for hydroxamate ions to adsorb [16]. Fe´ris et al [17] were found high separation values for oil droplets—emulsified in water by adsorptive particulate flotation using mineral particles as carrier. Treated solutions had very low content of oil, lower than emission limits. Best carriers were the hydrophobic adsorbents. These carriers showed to possess good adsorbing and flotation characteristics for oils, dyes and heavy metal ions removal such as copper, zinc and nickel. The aim of this search is to improve the effect of variable temperature on adsorption efficiency of (polyacrylic acid - bentonite) composite to Au$^{+1}$, Ag$^{+1}$ from neutral electronic waste solution by forth flotation method.

2. Materials

Electric waste powder, calcium bentonite clay, acrylic acid (AA) monomer, $N$, $N'$-Methylenebis - acylamide (MBA) as cross linking agent, ammonium persulfate (APS) as initiator were products of Sigma-Aldrich, deionized water, Nitric acid (HNO$_3$), Hydrochloric acid (HCl) were supplied CDH company and diluted (1:1) and forth flotation process agents ((PAA- bentonite) composite as a collector , activator, depressant and forther).

3. Methods

3.1. (PAA- bentonite) composite

In-situ polymerization[12], used to prepare composite material at the same ratio of polymer to clay acrylic acid (AA) monomer (5ml) was mixed with deionized water (45ml) by magnetic stirrer to obtain (50ml) mass solution. During the mixing, bentonite was added, heat was applied to reach mixing temperature (60±2°C), (0.01g) from clay rate, $N$, $N'$-Methylenebis-acylamide (MBA) was added. Pump inert gas (nitrogen N$_2$) for (10min) to remove oxygen. The Ammonium persulfate (APS) initiator (0.01g) from clay rate was added. The time required to complete the polymerization was nearly (2-3hrs).
3.2. E- waste solution by aqua regia (1HNO$_3$: 3HCl)

By (10ml) aquaregia (1:3 HNO$_3$: HCl), accurately about (5g) of the e- waste powder was dissolved in a beaker with stirring for (24hrs). The beaker content was filter paper. The filtrate was quantitatively transferred to (100ml) volumetric flask and the volume was completed with distilled water.

3.3. Adsorption of (Au$^{+1}$, Ag$^{+1}$) from E-waste by (PAA- bentonite) composite used forth flotation method

Forth flotation method was used to determine the efficiency of (PAA - bentonite) composite to adsorption of precious metals, taking various weights (2.5, 5, 10 g.l$^{-1}$) of adsorbent material at different temperatures (25, 35 and 45) °C. Flame Atomic Absorbent spectrometer was used to analyze and determine the remaining concentration of the output solution. Then calculate the adsorption efficiency (%) for the adsorbed materials by applying the following equations [18]:

$$\text{Adsorption efficiency} \% = \left(\frac{C_i - C_f}{C_i}\right) \times 100$$

Where $C_i$, $C_f$= initial and actual concentration of Ag$^{+1}$, Au$^{+1}$ respectively.

4. Results and discussion

In this part of the research, the results will be discussed according to the graphs of the adsorption efficiency of the (PAA - bentonite) composite as a function of the variables weight (g.l$^{-1}$) and temperature (°C) and comparing them with previous studies closest to the research.

![Figure 1](image-url)

Figure 1. Adsorption efficiency %Vs Weight (g.l$^{-1}$) at Temperature = 25 °C

Figure 1, showed that the adsorbent material (poly acrylic acid – bentonite) composite has the same behavior to adsorption precious metal Ag$^{+1}$, Au$^{+1}$ respectively. At room temperature (25 °C), the effect of amount of composite material clearly appears in this figure, where the adsorption efficiency for Au$^{+1}$ being approximation (47.9%), increased to (53.238%) and slightly decreased to (51.221) % at variable amount of adsorbent material. This indicates that the adsorption efficiency do not depend on the amount of adsorbent material. For Ag$^{+1}$, adsorption efficiency increased with adsorbent material increase, until (wt.=5g.l$^{-1}$), then it was decrease (37.581%) at (wt.=10g.l$^{-1}$), this is due to de-adsorption process. Therefore adsorption efficiency depends on the amount of (PAA - bentonite) composite. (PAA – bentonite) composite presence within a flotation cell to float depends upon its surface properties and stability of the froth phase through the interaction of the bubble surface and the particle during true flotation or entrainment.
Figure 2, note that the absorbent material has the same behavior for absorbing the precious metal (Ag + 1, Au + 1), respectively. The adsorption efficiency of the absorbing materials of the precious metals increased with the increase of the amount of the composite materials up to a weight = 5g.l⁻¹ and then decreased due to the de-adsorption process. However, the adsorption efficiency depends on the amount of composite materials. As for the effect of temperature on the properties of the e-waste solution, this is due to the difference in the size of the bubble bearing of the metal granule, surface tension, viscosity and density. The high temperature greatly affects the viscosity by increasing the mobility of the particles, and reducing the size of the hydrate particles.

Figure 3 indicates that the temperature increased has contributed to stabilizing the adsorption efficiency of adsorbent material. This effect is evident in (Ag + 1), where its value exceeded the previous value at 35 °C (after weight = 5g.l⁻¹) and became a linear pathway similar to (Au + 1) until (10 g.l⁻¹) via increase the carriers were the hydrophobic adsorbents, this shows that the adsorption efficiency is not dependent on the amount of composite material.
Reference [17] is the closest to this research by studying the removal of heavy metals from wastewater by forth flotation process and reference [12] by studying the effect of temperature on the composite material.

![Surface Plot of adsorption efficiency (Ag⁺) % Vs Weight (g.l⁻¹), Temperature °C](image)

**Figure 4.** Surface plot of adsorption efficiency (Ag⁺) % vs Weight (g.l⁻¹), Temperature °C

![Surface Plot of adsorption efficiency (Au⁺) % Vs Weight (g.l⁻¹), Temperature °C](image)

**Figure 5.** Surface plot of adsorption efficiency (Au⁺) % vs Weight (g.l⁻¹), Temperature °C

Figures (4, 5) refer the surface plot appears the effect of the parameters (amount of the adsorbent material and the temperature) on adsorption efficiency % for (Ag⁺, Au⁺) respectively. Which shows the clear gradient and convergence in the adsorption efficiency values of (Ag⁺, Au⁺), which increase with the increase of the adsorbent material and temperature, but at (5 g.l⁻¹) the adsorption efficiency begins to decrease, reaching the stability condition at (10 g.l⁻¹) and the temperature (45°C).
5. Conclusions
1- This research shows the effect of temperature differences on the adsorption efficiency of the composite material for precious elements (Ag+1, Au+1) in a neutral electronic waste solution.
2- The effect of high temperatures reduces the adsorption efficiency, especially at 45 °C.
3- The amount of superposition has an important role in the efficient adsorption of precious elements.

References
[1] JadHAV U., HochENG H., 2012, A review of recovery of metals from industrial waste, Journal of Achievements in Materials and Manufacturing Engineering, 54(2).
[2] Barakat M.A., 2011, New trends in removing heavy metals from industrial wastewater, Arabian Journal of Chemistry, 4, 361–377.
[3] KiP N., Alas A. B., 2012, The synthesis of poly (acrylic acid)- bentonite and polyacrylamide-bentonite composites for adsorption applications, Chemi K, 66, 7, 742-749.
[4] Homchuen P., 2017, Hydrometallurgical recovery of precious metals using magnetic adsorbents, thesis (doctoral), Division of Sustainable Resources Engineering, Graduate School of Engineering, Hokkaido University, Japan, March.
[5] JadHAV U., HochENG H., 2015, Hydrometallurgical Recovery of Metals from Large Printed Circuit Board Pieces, Sci. Rep. 5(14574).
[6] Kim E.Y., Lee M.S. and Pandey J.C., 2011, Selective Recovery of Gold from Waste Mobile Phone PCBs by Hydrometallurgical Process, J. Hazard. Mater. 198, 206-215.
[7] Tuncuk, A., Stazi, V., Akcil, A., Yazici, E. Y. and Deveci, H.; 2012, Aqueous Metal Recovery Techniques from E Scrap: Hydrometallurgy in Recycling, Miner. Eng., 25, 28-37.
[8] Lahtinen E., Kivijärvi L., Tatikonda R., Väisänen A., Rissanenand K. and Haukka M., 2017, Selective Recovery of Gold from Electronic Waste Using 3D-Printed Scavenger, ACS Omega, 2, 7299-7304.
[9] Abdul Khaliq M.A., Rhamdhani G. Brooks and Masood S., 2014, Metal Extraction Processes for Electronic Waste and Existing Industrial Routes: A Review and Australian Perspective, Resources, 3, 152-179.
[10] Chehade Y., A.Siddique, H. Alayan, N.Sadasivam, S. Nusri and T. Ibrahim., 2012, Recovery of Gold, Silver, Palladium and Copper from Waste Printed Circuit Boards, International Conference on Chemical, Civil and Environmental Engineering (ICCEE’2012), Dubai. Planetary Scientific Research Center
[11] Angela C. K. and Hugo M. V., 2018, Gold recovery from printed circuit boards of mobile phones scraps using a leaching solution alternative to cyanide, Braz. J. Chem. Eng. 35(3).
[12] Piotr N., Piotr K., Anna B. And Janusz S., 2013, Effect of Fe3+ ions present in the structure of poly(acrylic acid)/montmorillonite composites on their thermal decomposition, J Therm Anal Calorim, 113, 335–342.
[13] Li, X., Ha, C.-S., 2001, Nanostructure of EVA / Organ clay Nanocomposite: Effects of Kinds of Organ clays and Grafting of Maleic Anhydrides onto EVA, J. Appl. Poly. Sci., 87, 1901-09.
[14] Ren, J., Yanxia, H., Yan L., and Xiaozhen, T., 2005, Preparation, Characterization and Properties of Poly(vinyl chloride) / Compatibilizer / Organophilic-Montmorillonite Nanocomposites by Melt Intercalation, Polymer Testing, 24, 316-323.
[15] Rafiei H. R, Shirvani M. and Ogusseitan O. A., 2016, Removal of lead from aqueous solutions by a poly(acrylic acid)/bentonite nanocomposite, Applied water science, 6(4) 331–338.
[16] Zhang Y., 2015, Froth flotation of xenotime , thesis of Master of Science Metallurgical and Materials Engineering, Faculty and the Board of Trustees of the Colorado School of Mines.
[17] Fe´ris L.A., DeLeo’n A.T., Santande M. and Rubio J., 2004, Advances in the adsorptive particulate flotation process, Int. J. Miner. Process 14(1-4),101-106.
[18] Barbooti M.M, 2015, Simultaneous Removal of Chromium and Lead from Water by Sorption on Iraqi Montmorillonite, J. Environ. Protection, 6, 237-249.