INVESTIGATION OF ULTRASONIC INFLUENCE ON THE KINETICS OF EXTRACTING GOLD FROM ELECTRONIC WASTE

Received – Primljeno: 2022-10-24 Accepted – Prihvaćeno: 2022-12-26 Original Scientific Paper – Izvorni znanstveni rad

In this paper the effect of ultrasonic treatment on the apparent activation energy of gold leaching from e-waste in aqueous thiourea solution was studied. The leaching was carried out under conventional conditions and under ultrasonic treatment. The leaching in both cases was carried out at 25 °C, 35 °C, 45 °C and 60 °C. The apparent activation energy using ultrasonic activation decreases. In both cases the process of gold leaching from e-waste proceeds in the diffusion region. Thus, ultrasonic intensification is a means of increasing the efficiency of hydrometallurgical processes of e-waste processing by reducing the activation energy of the process.

Keywords: extraction, gold, electronic waste, ultrasound, thiourea.

INTRODUCTION

The issue of recycling (electronic waste) e-waste is an acute one in the world today. It is known that e-waste can be a source of various metals [1]. Both pyrometallurgical and hydrometallurgical methods are applicable to recycling of secondary electronic raw materials in order to extract metals. It is known that hydrometallurgical methods have a number of advantages over pyrometallurgical methods in terms of lower consumption of energy, resources and release of various harmful reaction products [2]. However, hydrometallurgical methods are not without disadvantages, such as incomplete extraction of useful components from various raw materials [3], or toxicity of reagents.

These shortcomings can be compensated for by using non-toxic reagents, and for the complete extraction of useful components, hydrometallurgical processes can be intensified in various ways, such as vibration, microwaves, ultrasound [4] and other methods [5].

The authors have previously studied the effect of microwaves on copper leaching with aqueous sulphuric acid solution from man-made waste, in order to further extract gold with thiourea [6]. Copper had to be extracted at the first stage to exclude thiourea decomposition in contact with copper. In the experiment it was determined that exposure to ultrasound significantly increases the extraction of copper and other metals into the productive solution.

Corresponding author: sapinov.r@teachers.tou.edu.kz

It is reasonable to assume that intensification by ultrasound would also affect the recovery of gold in the pregnant solution. To test this hypothesis and to understand the intensification processes, it was decided to investigate the effect of intensification on the activation energy of the process.

The activation energy is the level of energy that the molecules of the reactants must overcome in order to carry out a reaction. The activation energy is calculated according to the formula proposed by S.A. Arrhenius (formula 1).

$$k = A \cdot exp(-E_a/RT)$$
(1)

where A is the pre-exponential multiplier,

E_a – activation energy,

R – gas constant,

T – absolute temperature.

Unit of measure to J/mol [7].

The duration (τ /min) and process temperature (T/°C) as well as the application of the intensification were



Figure 1 Scheme of the experiment normal conditions (1) and with (2) ultrasonic impact treatment (UIT)

Zh. Shoshay, M. Sadenova: D. Serikbayev East Kazakhstan technical university, Kazakhstan

M. Suyundikov, R. Sapinov, D. Absolyamova: Toraighyrov University, Pavlodar, Kazakhstan

P. Varbanov: - Sustainable Process Integration Laboratory – SPIL, NETME Centre, Faculty of Mechanical Engineering, Brno University of Technology - VUT, Brno, Czech Republic

varied during the experiment. The reagent concentration and Solid/Liquid (SL) ratio were chosen on the basis of previous studies. In the first stage (pretreatment) copper was extracted to exclude harmful effects on thiourea by the method described in a previous article by the authors [8]. Then gold was extracted with aqueous thiourea solution under normal conditions and with ultrasound exposure. Scheme of the experiment is shownin Figure 1.

METHODS AND MATERIALS

All experiments were carried out in the laboratory of EKTU named after D. Serikbaev (https://www.ektu.kz). Electronic waste was used as raw material (Figure 2). Ball and vibrating mills were used to grind the materials. The elemental composition of the materials was determined using an ICP-MS 7500 cx inductively coupled plasma mass spectrometer from Agilenttechnologies (USA) (Table 1). A JSM-6390LV scanning electron microscope (SEM) manufactured by JEOLLtd. (Japan) was used to show the distribution of element profiles. All reagents were of analytical purity. Chemical heat resistant glassware was used for leaching. An ultrasonic bath PSB - 1335-05 produced by "PSB-Hals" (Russia) was used for activation of processes. Leaching was carried out with stirring on a magnetic stirrer. Solid and liquid phases were separated by filtration, and filtrates were analyzed for content of valuable components. The residues were dried and also analysed to determine their content of valuable components.



Figure 2 Electronic waste

Leaching stage

During the study, e-waste samples were leached in 2 ways.

1 Method. Raw materials were ground and leached in an aqueous thiourea solution with a concentration of 20 g/l, at 25 °C, 35 °C, 45 °C, and 60 °C. Solid to liquid S/L content (20), process duration min - (30, 45, 60, 90, 120), without ultrasonic treatment, with stirring on a magnetic stirrer.

2 Method. The raw material was subjected to ultrasonic treatment for 30, 45, 60, 90, 120 minutes. Leaching in 20 g/l aqueous thiourea solution at 25 °C, 35 °C, 45 °C, and 60 °C.

Solid to liquid S/L (20), for 60 minutes, with stirring. The dish with the solution and the suspension was placed in a sonicator at the beginning of leaching and then every 15 minutes for 60 seconds.

RESULTS AND DISCUSSION

The resulting chemical analysis of the e-raw material is shown in Table 1. Copper 26,4 %, aluminium 3,9 %, zinc 1,2 % and other elements are shown. The gold content is 0,0086 % (86 g/t).

Table 1 Average concentration of metals in the mixture samples in e-waste/wt.%

| Cu | AI | Sn | Ni | Zn |
|------|-----|-----|--------|-----|
| 26,4 | 3,9 | 3,2 | 2,3 | 1,2 |
| Pb | Fe | Ag | Au | Pd |
| 1,7 | 4,2 | 0,2 | 0,0086 | 0,1 |

Figure 3 shows images of the element distribution profiles in a selected area of the sample. The analysis also confirmed the presence of Au





Figure 3 SEM, normal condition (a), SEM of element distribution profiles in the selected area (b)

Leaching stage.

Leaching results are shown in Table 2 and Table 3. Table 2 shows the recovery of gold to the pregnant solution without the ultrasonic impact treatment (UIT) as a function of process time

Table 3 shows the recovery of gold in the pregnant solution with ultrasonic impact treatment (UIT) of the raw material.

| Min | 25 °C | 35°C | 45 °C | 60 °C |
|-----|-------|------|-------|-------|
| 0 | 0 | 0 | 0 | 0 |
| 30 | 20 | 22 | 23 | 29,6 |
| 45 | 32,4 | 35 | 38,4 | 46,7 |
| 60 | 37,9 | 43,7 | 58,6 | 71,2 |
| 90 | 40,3 | 46,4 | 65 | 78,3 |
| 120 | 40,6 | 49,8 | 67,2 | 79,4 |

Table 2 Gold recovery into the pregnant solution without ultrasonic treatment /wt. %

Table 3 Gold recovery into the pregnant solution with ultrasonic treatment /wt. %

| Min | 25 °C | 35°C | 45 °C | 60 °C |
|-----|-------|------|-------|-------|
| 0 | 0 | 0 | 0 | 0 |
| 30 | 28,6 | 32 | 32,5 | 34,7 |
| 45 | 48,3 | 60 | 50 | 67 |
| 60 | 57 | 67,6 | 77 | 84,6 |
| 90 | 63,2 | 75,4 | 83 | 87,2 |
| 120 | 65 | 77 | 84,2 | 88,6 |

Figure 4a shows plots of gold recovery under normal conditions and Figure 4b with ultrasonic impact treatment (UIT). It can be seen that the use of ultrasonic



Figure 4 Plots of recovery versus duration and temperature of the leaching process under normal conditions (a) and with UIT (b)

intensification increases gold recovery at all temperature ranges.

In order to study the effect of ultrasound on activation energy, the experimental plots were processed using the least squares method using Excel software. Horizontal lines were plotted to obtain extraction data of 20, 40 and 60 % at different temperatures (Figure 5).



Figure 5 Graphs of recovery vs. duration and temperature of the UIT gold leaching process

The values obtained and their treatment results under normal conditions and with ultrasound exposure, were used to estimate the apparent activation energy of the interaction of the material with aqueous thiourea solution.

For this purpose the logarithm of the time required to achieve the same degree of gold recovery in solution at different leaching temperatures was determined as a function of the inverse temperature (Figure 5). The duration of thiourea leaching of e-raw material providing a given gold extraction into a solution at different leaching temperatures. The logarithm of the time required to achieve the same degree of gold recovery in solution at different leaching temperatures under normal conditions is shown in Figure 6 a. The logarithm of the time required to achieve the same degree of gold recovery in solution at different leaching temperatures under conventional conditions 6a and ultrasonic intensification is shown in figure 6b.

The angular coefficients of straight $\lg \tau = f(1/T)$ (Figure 6) are related to the apparent activation energy by the relation $d(\lg \tau)/d(1/T) = E_a/(2,3R)$. The apparent



Figure 6 Dependencies of the logarithm of the leaching process duration on its return temperature under normal conditions (a), with UIT (b)

activation energy of the interaction of e-waste and aqueous thiourea solution without ultrasonic treatment was 31,087 kJ/mol. The apparent activation energy of interaction of e-waste and aqueous thiourea solution with ultrasonic treatment was 14,941 kJ/mol. Values of apparent activation energy corresponding to diffusion mode = 8-20 kJ/mol, and kinetic mode = 40-300 kJ/mol [9]. Thus, the value of apparent activation energy $E_a =$ 14,941 kJ/mol means that the process of thiourea leaching occurs in the diffusion region. In both cases. However, the activation energy decreases when exposed to ultrasonic.

CONCLUSIONS

Study of kinetics of e-waste thiourea leaching process is aimed at finding of leaching process activation parameters by determining dependence of leaching time and extraction on main technological parameters - temperature, process duration and setting stage regime. It has been found that using ultrasonic activation, at a temperature of 60 °C, the recovery of gold in the productive solution has increased. A kinetic model of thiourea leaching of electronic wastes was constructed. Obtained experimental data of activation energy without ultrasonic treatment Ea = 31,087 kJ/mol, and with ultrasonic treatment of raw material Ea = 14,941 kJ/mol. Thus, values of apparent activation energy in both cases Ea = 31,087 kJ/mol and 14,941 kJ/mol mean that the process of thiourea leaching occurs in the diffusion region. In addition, the ultrasonic lowered the activation energy from 31,087 kJ/ mol to 14,941 kJ/mol. This area of research is promising and requires further development.

REFERENCES

- J. V. Yken, N.J. Boxall, K.Y. Cheng, A.N. Nikoloski, N.R. Moheimani, A.H. Kaksonen. E-Waste Recycling and Resource Recovery: A Review on Technologies, Barriers and Enablers with a Focus on Oceania *Metals*. 11 (2021) 8, 1313.
- [2] D.A. Ray, M. Baniasadi, J.E. Graves et al. Thiourea Leaching: An Update on a Sustainable Approach for Gold Recovery from E-waste. J. Sustain. Metall (2022) 8, 597–612.
- [3] M.D. Rao, K.K. Singh, C.A. Morrison, J.B. Love Challenges and opportunities in the recovery of gold from electronic waste RSC Adv. 10 (2020) 8, 4300-4309.
- [4] G. Zhang, S. Wang, L. Zhang & J. Peng. Ultrasound-intensified Leaching of Gold from a Refractory Ore. ISIJ International 56 (2016) 4, 714–718.
- [5] Y. Yang; M. Lai, Q. Zhong, Q. Li, B. Xu, T. Jiang. Study on Intensification Behavior of Bismuth Ions on Gold Cyanide Leaching Metals (2019) 9, 362.
- [6] Z. Shoshay, R.V. Sapinov, M.A. Sadenova, P.S.Varbanov, Intensification of the Process of Extracting Non-Ferrous Metals from Kazakhstani Technogenic Raw Materials, Chemical Engineering Transactions (2021) 88, 1069-1074.
- [7] J. Kohout, ModifiedArrhenius Equation in Materials Science, Chemistry and Biology. Molecules (2021) 26, 7162.
- [8] Z. Shoshay, R.V. Sapinov, M.A. Sadenova, P.S.Varbanov, 2021, Hydrometallurgical Methods for Extracting Nonferrous Metals from Electronic Gadgets, Chemical Engineering Transactions (2021) 88, 139-144.
- [9] G.M. Voldman, A.N. Zelikman Theory of hydrometallurgical processes, Textbook for Universities. - 4-th edition, revised and supplemented - M.: Intermet Engineering (2003), 464 pp. (in Russian)
- **Note:** The responsible for English translate is Adam Daurbekov, Pavlodar, Kazakhstan.