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Recovery of gold nanoparticles from electronic solid waste: An experimental design and mathematical approach

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Abstract

Due to the growing consumption of gold metal and its various applications, gold recovery is very important. The element gold is an electron-bearing particle that can be used in a variety of applications such as medicine, engineering, and biology by changing its surface activity and particle size. The purpose of this study is to recover gold metal in the form of gold nanoparticles from electronic solid waste. For this purpose and to better investigate the effect of factors affecting the extraction of gold nanoparticles, the response method has been used in the experimental design. The innovation is to make gold nanoparticles with the smallest particle size with the help of reducing materials and in a cost-effective way while saving energy and also the highest recovery rate. In order to make gold nanoparticles, there are different methods such as physical, chemical and biological methods. Among these three methods, the use of the chemical method has been selected for this research. The material for extraction was royal acid, and parameters such as pH, the amount of royal acid and the temperature of the solution on the gold extraction efficiency were examined. The final nanoparticles were analyzed using XRD, FESEM, EDX and TEM analyzes. In order to determine the effectiveness of each variable and to determine which of the studied variables has the greatest impact on the gold recovery process, statistical methods were used. The variables of pH, solution temperature and the amount of Soltani acid had the greatest effect on the recovery of gold nanoparticles, respectively. According to the information obtained from Design-Expert software, at a temperature of 60 ° C and a pH of 8 and 200 ml of Soltani acid solution, the recovery rate of gold nanoparticles was 97.3%.

Keywords: Gold Nanoparticles, Recovery, Experimental Design, Solid Waste.
2020 MSC: 97M60

1 Introduction

Gold metal is very old; According to the available evidence, it dates back to 3300 BC. This metal is in the category of precious metals, which due to its color and beauty, has long been highly regarded. Gold with properties such as flexibility and malleability is used in jewellery [2]. Also, due to its physical and chemical state, it is widely used in high-tech industries and medical science. In recent decades, this metal has been used for the placement of joints and

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corrosion resistance of materials in the electrical and electronics industries due to its properties such as high electrical conductivity and low electrical resistance [7].

In the chart in Figure 1,you can see the consumption of gold metal in various matters in the period 2009 to 2018.

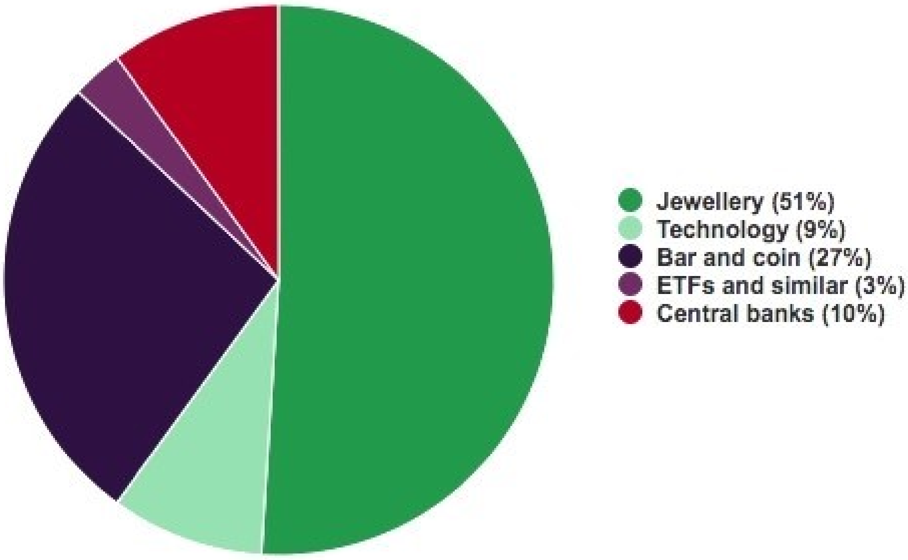

Figure 1: Consumption of gold metal in various matters in the period of 2009 to 2018

As can be seen, more than half of the gold consumed in the world is related to jewelry and ornaments, and in the discussion of technology and matters related to electronic and mechanical equipment, it is about 9% of the share of gold consumed in the world. That is why recovering gold from electronic and mechanical equipment and devices can be valuable.

In 2020, a study was conducted on the amount of recycled materials obtained from the recovery of electronic and electrical equipment [5], the results of which canbe seen in the diagram in Figure 2.

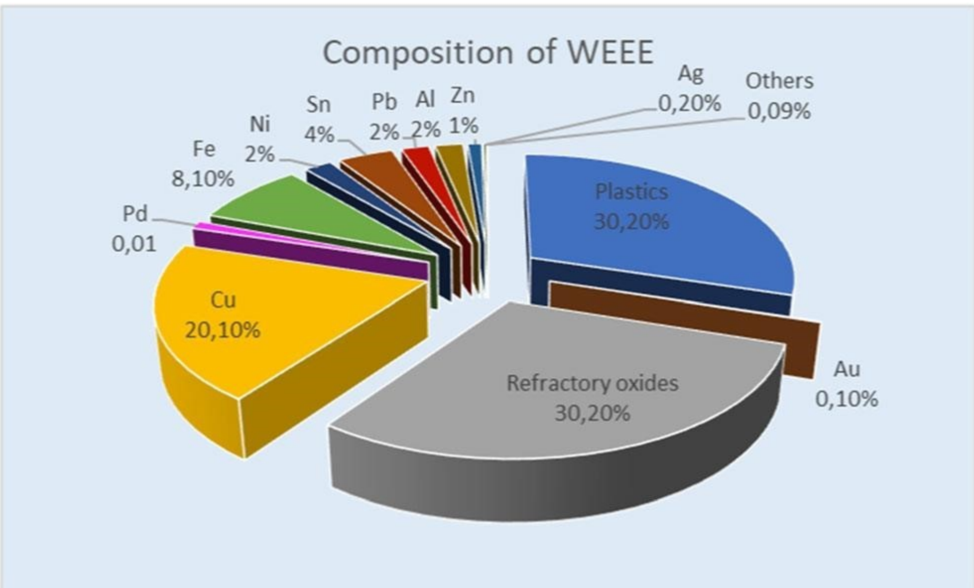

Figure 2: The amount of recycled materials recovered from electronic equipment

Separation processes are divided into three categories: interphase separation, chemical reaction separation and mass transfer separation in one phase [15, 10]. One of the attractions that have had high efficiency in the gold separation process is Soltani acid [6]. Accordingly, in this study, the effect of Soltani acid on gold recovery in electronic waste has been investigated. Also innovative is the production of gold nanoparticles with the smallest particle size with the help of reducing materials and in a cost-effective way while saving energy.

Experimental design and response surface methodology can be used as a powerful tool to make the most of laboratory or simulation facilities. One of the most important issues in chemistry and chemical engineering today is the design of experiments. In all processes, testing is performed to obtain information about one or more specific processes. In fact, by performing the experiment, the changes made to the desired response variable can be examined by applying changes to the input variables and the control factors affecting the experiment. Since some of the factors identified by the experimenter as effective factors and changes are made on them, have little effect on the desired response variable and only cause additional costs, it is necessary to correctly identify these factors and create appropriate levels. These factors avoided incurring additional costs as much as possible to examine changes to the response variable under study. Experimental design is used to solve this problem [4, 1]

Experimental design using existing software in the field of experimental design is very popular today. Doing experiments always saves a lot of time and money, so performing effective experiments that obtain the most information with the least cost and time is the goal of every researcher. The use of experimental design also helps engineers to develop and evolve production and create processes that are resistant to environmental factors and other sources of change [3].

Among the experimental design methods, the parameters related to the well-estimated procedure-response models can be easily performed. This type of test design is used to fit a procedure to the data. This method requires two experimental factors. Levels of independent factors (control factors) are considered as +1 for high levels, 1- for low levels and 0 for the design center [2]. RSM method is divided into the following three categories according to the type of design space [11]:

- Box-Benken (BB).
- Central Composite (CCD).
- D-Optimal materials and methods.

Materials used in the present study include 37% hydrochloric acid (Kimia Tehran acid), electronic plates (Firmco), Tizab Soltani (Kimia Tehran acid), urea (merc), iron sulfate (merc) and deionized water. All materials used in this study are from reputable brands of laboratory materials, so there was no need for purification for them.

The method of making nanoparticles was top-down. In this method, using specific tools, the material is separated from the bulk body and the body reduces in size and scale to reach nanometer sizes. The method of gold nanoparticle extraction in this research was the use of reducing compounds. First, 5 electronic plates (containing a total of about 1 gram of gold) were boiled in a solution of 200 ml of 1 M hydrochloric acid at 100° C for 15 minutes. After the solution had cooled, the solution was passed through a strainer and the gold foils were separated. The gold foils separated from the first stage were mixed with royal acid and subjected to stirring (ten minutes) at 100° C until complete dissolution of the gold in the acid.

After 10 minutes, the solution was cooled to 70° C and then formed to neutralize $[NO_3]^{-200}$ mL of urea was added to the solution and stirred for 10 min at 100° C. Celsius was continued until the reaction between urea and gold was complete. In the next step, 150 ml of 0.1 M iron sulfate was added to the system and the resulting solution was stirred for 15 minutes. Finally, the solution was purified and washed, and the resulting solid containing gold nanoparticles was dried under ambient conditions for 24 hours. Figures 3 and 4 show a view of the electronic screens used and the extracted solid gold.

In Table 1, you can see the test conditions and the results obtained from the test design in the software.

2 Discussion

For the present study, some assay analyzes have been used to prove the synthesis of gold nanoparticles. These analyzes include XRD, FESEM, EDX and TEM, which are interpreted in the following. X-ray diffraction pattern of gold nanoparticles in Figure 5,in the 40 and 67 ˚ regions, the nanoparticles show sharp peaks, which is a reason for the proper extraction of gold. Structural analysis shows that the nanoparticles have a crystalline structure with Miller indices (111) and (220) in a cubic lattice. Comparing the intensity of the peaks, we find that the peak related to

Figure 3: Extraction of gold from electronic plates

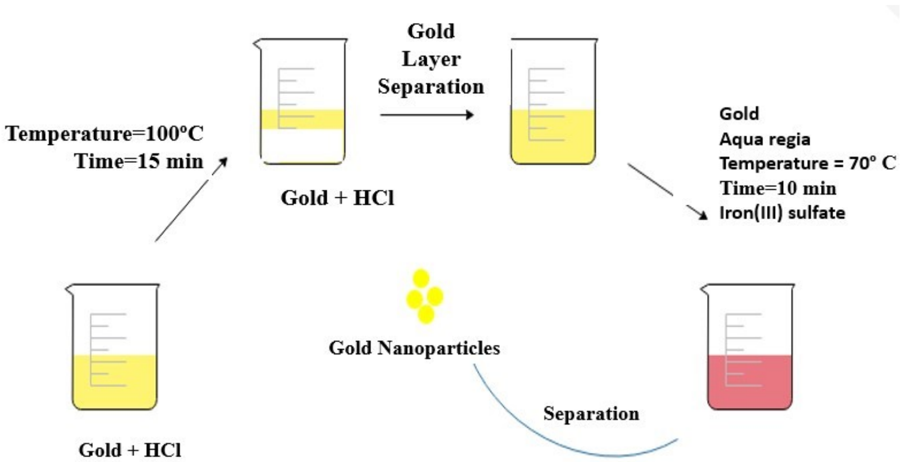

Figure 4: A view of the method of making gold nanoparticles

Table 1: Conditions of experiments and results obtained

Recovery percentage	Aqua regia level	Soluble temperature	pH	Test number
84/5	100	20	8	1
83/3	200	60	4	2
87/86	100	60	8	3
85	100	60	4	4
82/5	100	20	4	5
97/3	200	60	8	6
92/2	200	20	8	7
81/20	200	20	4	8

the structure (220) is more severe than the peak related to the structure (111), as a result, more nanoparticle crystal plates are formed in this direction [12, 9]

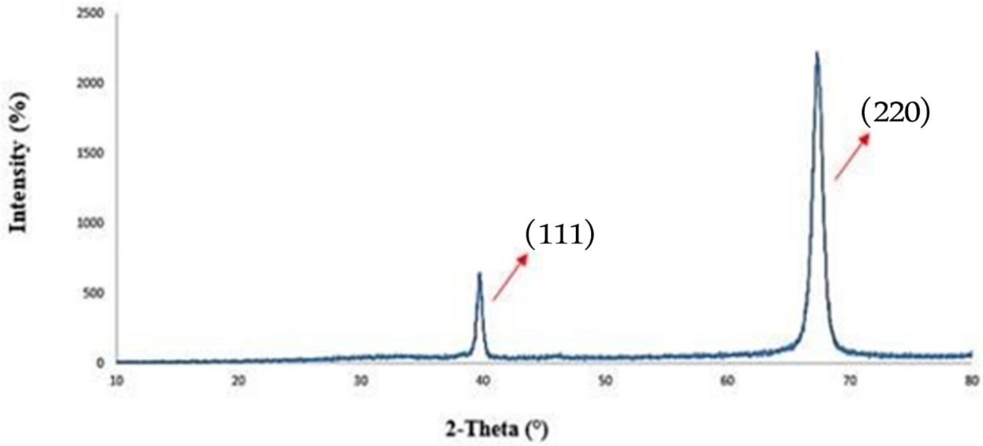

Figure 5: X-ray diffraction (XRD) spectrum of gold nanoparticles extracted from electronic plates

Also, to ensure the presence of gold in the system after extraction from electronic pages, an element check test was performed. The results confirm and indicate the presence of gold and copper in the system after the fabrication of nanoparticles, which indicate its proper synthesis and extraction (Figure 6).

As shown in Figure 6,one of the components of electrical panels is gold metal, which is placed in the form of nanoparticles in electron plates. The presence of copper in the results is also due to its presence in the structure of electronic plates. Silver metal is also one of the materials used in the manufacture of electrical panels; But obviously the peak that appears is related to gold and indicates that gold nanoparticles are used in the construction of electrical panels more than any other metal.

In addition, in Figure 7,SEM images of synthetic nanoparticles can be seen. According to the images of gold nanoparticles hemispherical particles are well synthesized, hemispherical nanoparticles have a homogeneous and smooth surface and uniform particle size distribution and can be seen in some areas of agglomeration which indicates the merging of several particles. Also, by measuring the particle size in Figure 7,the presence of particles at the nanometer scale is well visible (average size in the range of 60 to 120 nm); Therefore, it can be said that the synthesis method has a good quality and homogeneous hemispherical particles have been synthesized.

The results of TEM analysis are shown in Figure 8.In these figures, it can be clearly seen that the gold nanoparticles are spherical in different parts of the mass. Therefore, their sphericity and dispersion are also proved in this analysis.

In examining the effect of Soltani acid level, other influential variables were considered constant and only Soltani acid level changed. Different volumes of Soltani acid were equal to 50, 75, 100, 125, 150 and 200 ml and other variables were constant. The effect of Soltani acid concentration on gold recovery is shown in Figure 9.As shown in the figure, Tizab Soltani has the ability to extract gold. The presence of 50 ml of Soltani acid at the beginning of the recovery process leads to the extraction of about 65% of gold (Area 1). Further increase of the recovery material in the system drastically increases the amount of gold mining to the point that eventually the recovery rate increases to over 90%.

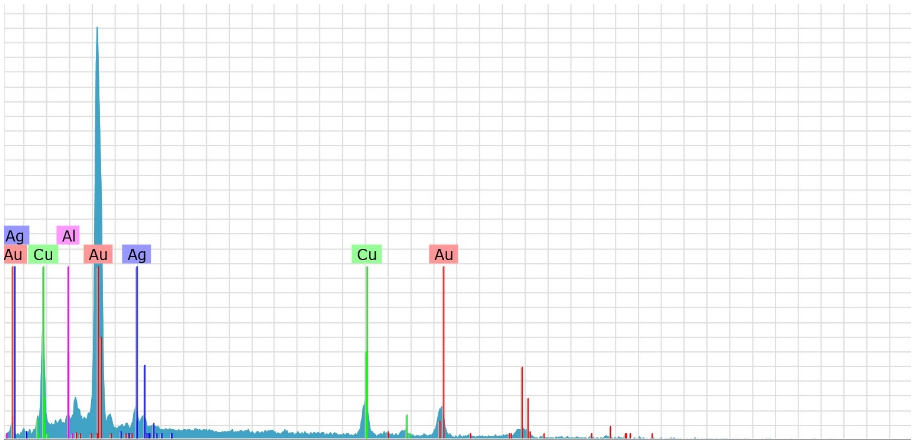

Figure 6: Investigation of the presence of gold in the solid structure synthesized using EDX analysis

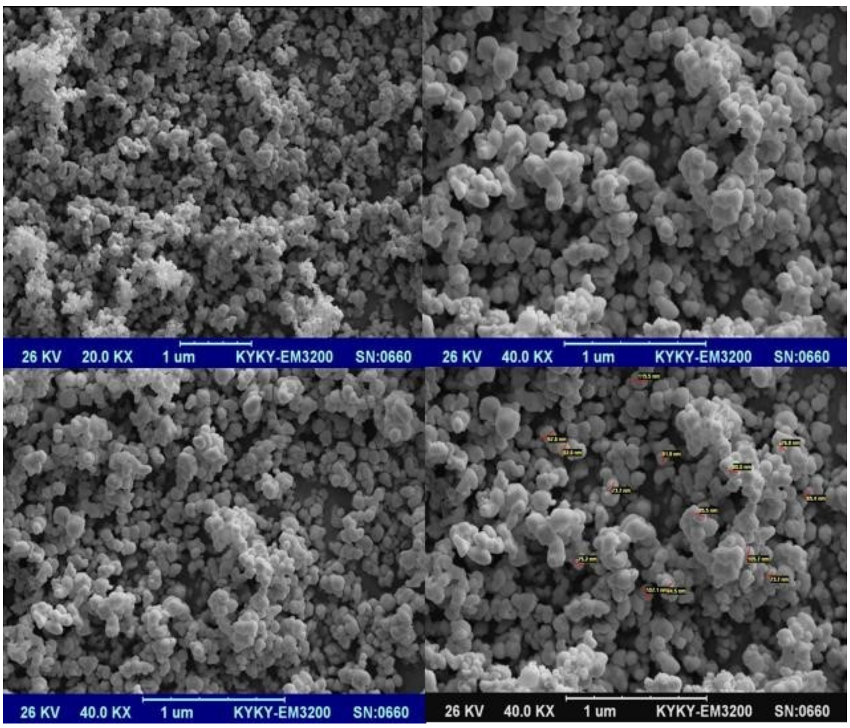

Figure 7: FESEM images of gold nanoparticles extracted from electronic plates

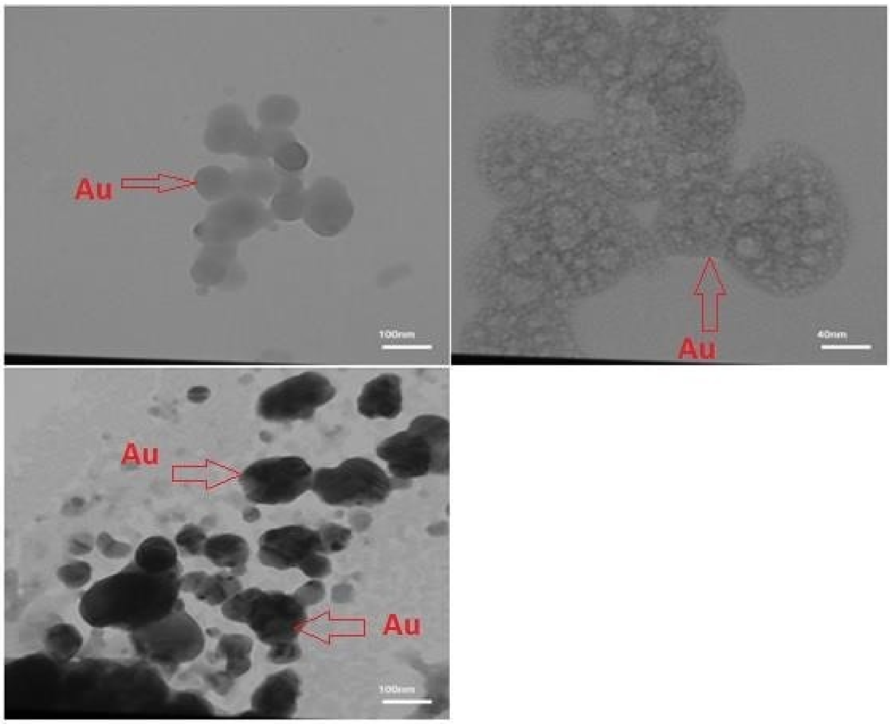

Figure 8: TEM images of synthesized gold nanoparticles extracted from electronic plates

With increasing the concentration of Soltani acid up to 120 ml, the slope of changes in the amount of recovered gold increases significantly (Area 2). But at a rate of 120 to 200 ml of royal jelly, the slope of gold recovery increases slowly (Zone 3). Decreasing the slope of the curve by increasing the concentration of the extractor can indicate the equilibrium of the extraction reaction and reduce the effect of the extractor in these conditions.

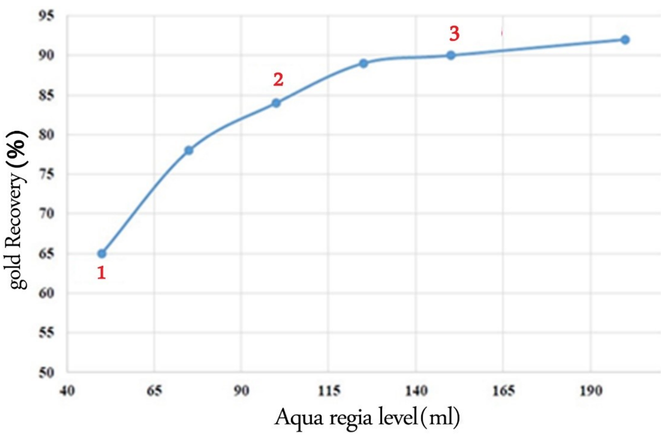

Figure 9: The amount of gold absorption in different amounts of Aqua regia

By studying the diagram, it can be concluded that with increasing the amount of adsorbent, the amount of gold absorption increases, which is due to the increase of absorption sites in Soltani acid, and in fact, with increasing the amount of adsorbent, the capacity of gold increases.

In order to investigate the effect of pH, five solutions with a concentration of 5 ppm and a volume of 20 ml solution with pHs of 3, 5, 7, 9 and 11 were tested using a pH meter and 0.1 M acidic and alkaline solutions. Other influential variables except pH were considered constant.

The results of different pH tests can be seen in Figure 10.

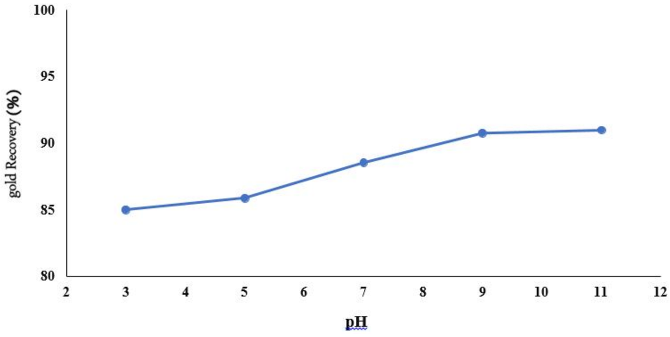

Figure 10: The rate of gold absorption at different pH

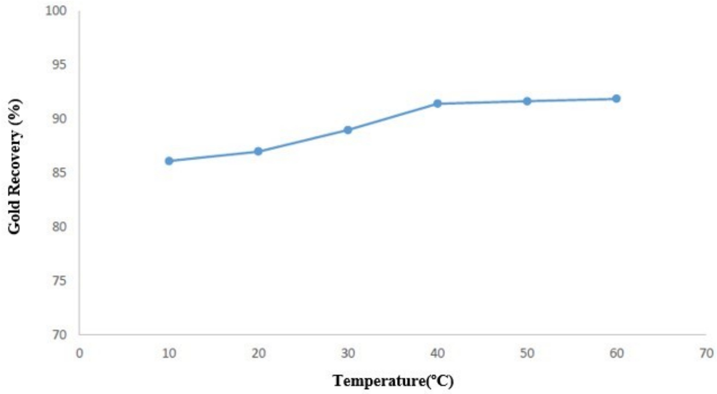

Figure 11: The rate of gold absorption at different temperatures

In Table 2, we can see the results of the analysis of gold recovery with the help of Tizab Soltani. To achieve the experimental model for predicting the rate of gold recovery, a linear relationship was considered between the data obtained from the experiment.

Table 2: Results of linear model analysis of variance for gold recovery

P	T	Standard error coefficient	coefficient	Variable
0/000	32/81	1/95	63/98	Mathematical constants
0/000	18/76	1/47	27/65	pH
0/027	3/43	0/02	0/08	Aqua regia level
0/003	6/50	1/47	-9/55	Soluble temperature

R-Sq=%98/8 R-Sq (pred)= %94/5 R-Sq(adj)= %96/21

The coefficient characteristics of each variable, the standard error of each estimate, the T and P characteristics can be seen in this table. It should be noted that the T component alone does not give the correct answer; But to calculate P, it is necessary to have this component. Based on the P component, can we say whether the desired answer is acceptable or not? If the P value is less than 0.05, it indicates the effect of that phrase on the results. The higher the R and R2 values, the better the model. Based on the results of Table 2, it can be said that the pH variable has the greatest effect on gold recovery. After that, temperature has a significant effect on gold recovery.

3 Conclusion

In this study, gold metal was extracted as gold nanoparticles from electronic plates with the help of a chemical reducing agent made from iron compounds. The results of X-ray diffraction test showed the crystal structure with the presence of peaks (1 1 1) and (0 2 2) related to the type of crystallinity of the material. Also, with the help of ultraviolet spectroscopy, the peak of gold particles in the range of 700-400 nm has been observed. Finally, scanning and transmission electron microscopy tests were performed to ensure the morphology, microstructure and particle size of the synthesized gold. The obtained results showed the formation of homogeneous and uniform gold nanoparticles. In other words, tests such as visible-ultraviolet spectroscopy, scanning electron microscopy, and transmission electron microscopy have confirmed the quality fabrication and proper extraction of gold particles at the nanometer scale from electronic plates.

In this study, optimization and the effect of different variables on the amount of gold nanoparticles were performed using Design Expert software. In this software, the surface response technique and the central composite part were used. In order to determine the effect of each variable and to determine which of the studied variables have the greatest impact on the gold recovery process, statistical methods were used. The variables of pH, contact temperature and concentration of Soltani acid solution had the greatest effect on the recovery of gold nanoparticles. According to statistical calculations, the highest recovery rate of gold nanoparticles in this study was 97.3%, which is a very high and good rate for the extraction of gold nanoparticles (at a temperature of 60 $^\circ$ C, 200 cc of Soltani acid and pH equal to 8).

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