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Research Article APPLICATION OF MULTI CRITERIA DECISION MAKING METHODS TO LEACHING PROCESS OF COPPER FROM MALACHITE ORE

Bilal KURŞUNCU*¹, Ali YARAŞ², Hasan ARSLANOĞLU³

¹Bartin University, Department of Mechanical Engineering, BARTIN; ORCID:0000-0002-2304-2962 ²Bartin University, Dep. of Metallurgical and Materials Eng., BARTIN; ORCID:0000-0003-1725-7788 ³Firat University, Department of Chemical Engineering, ELAZIĞ; ORCID:0000-0002-3132-4468

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ABSTRACT

This study concerns with optimization of the leaching conditions of copper from malachite ore in the presence ammonium nitrate solution. In order to select the effective parameter from available dissolution process, all experiments were performed using L_{25} orthogonal experimental design by Taguchi method and the temperature was found as a more effective parameter. Then multi-criteria decision making (MCDM) method was applied to experimental results and it was found that the process is in concordance with MCDM values. Based on the results, the highest copper extraction value (99.4%) was reached under optimum leaching conditions are as follows: time, 60 min; temperature, 75°C; stirring speed, 450 rpm; concentration of leaching reagent, 4 mol/L and solid/liquid ratio, 8 g/mL. In conclusion, Taguchi and MCDM method can be used effectively for optimization of various hydrometallurgical processes.

Keywords: Leaching, malachite, ammonium nitrate, optimization, Taguchi, TOPSIS, MCDM.

1. INTRODUCTION

In nature, the ores with copper content are available such as malachite, azurite, tenorite, covellite, chalcopyrite and bornite [1]. Metallic copper production from these ores generally can be performed by pyrometallurgical and hydrometallurgical methods at the present time [2]. Hydrometallurgical method has numerous advantages over pyrometallurgical method; less costly, more environmentally friendly and acceptable economically reported to be most appropriate method [2-6]. For these reasons, hydrometallurgical method was preferred in this study.

The strong acids are commonly utilized in the conventional dissolution process as the leaching reagents. However, the acids may cause some problems such as excessive acid consumption because of in presence of gangue minerals and dissolving the impurities in the ore matrix during leaching reaction. Therefore, leaching reagents including ammonia were utilized in leaching process of oxidized copper ores in previous works [2, 3, 5-9]

It is complicated to optimize any leaching process since it is affected simultaneously various conditions such as concentration of leaching agent, temperature, time, stirring speed and solid/liquid ratio. Several experiments are necessary to determine the simultaneous effects with

^{*} Corresponding Author: e-mail: bkursuncu@bartin.edu.tr, tel: (378) 501 16 65

correctly approach of all parameters on leaching, which means also highly cost and time consuming. Therefore, instead of classical experimental design, Taguchi method was used to find optimum leaching conditions. Multi criteria decision making (MCDM) method was also used to verify optimum leaching condition which was validated with experiment. The stages were detailed in Figure 1.

In this work, we were presented an approach related to the optimization leaching parameters of copper from malachite ore in ammonium nitrate solution by proposed methods.

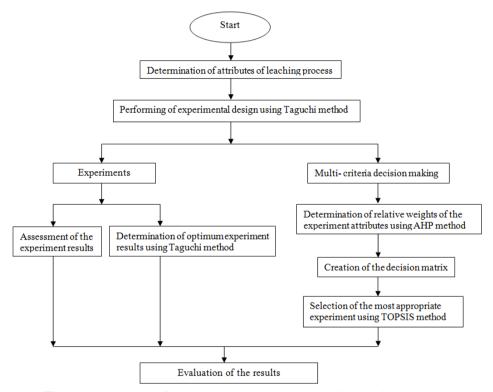


Figure 1. Flow diagram for proposed algorithm using Taguchi and MCDM methods

2. EXPERIMENTAL METHOD

Oxidized copper ore $(CuCO_3Cu(OH)_2)$ utilized in the experiments was sieved using ASTM standard sieves to obtain certain particle fractions, after crushed and ground operations.

All experiments were carried out 1000 mL. a jacketed glass reactor at atmospheric pressure. The reactor contents a mechanical stirrer, a temperature probe and a condenser to prevent the volume reduction of the leaching solution by evaporation. The temperature of ammonium nitrate solution in reactor was adjusted to desired value, sample was added to the solution, and mixtures were stirred for the required reaction time and filtered. Then, amount of copper in the supernatant was measured using an Atomic Absorption Spectrophotometer (Perkin Elmer AAnalyst-400).

3. EXPERIMENTAL DESIGN AND OPTIMIZATION OF PROCESS PARAMETERS

3.1. Taguchi Method

Taguchi method is a powerful design and reduced the number of tests by using orthogonal arrays in engineering analysis. The quality of any experimental design can be described by basic philosophy of Taguchi method. In this way, the experiment time can be shortened and the process cost can be minimized in the any engineering application [21, 22].

Deviation experimental values and desired values are calculated by Taguchi method as a loss function. Thereafter, this loss function is converted into a signal-noise (S/N) ratio. Normally there are three kinds of quality characteristics in analysis of the S/N ratio.

- The lower the better
- The higher the best
- The nominal the better

S/N ratio is calculated based on the S/N analysis for each level of the process parameters. The higher the best signal to noise ratio was preferred since we plan to obtain the maximum copper extraction value in this process.

3.2. TOPSIS (Technique for Order Performance by Similarity to Ideal Solution) Method

The MCDM methods are generally preferred to determine the optimum process parameters for numerous engineering applications at the present time [10]. There are various types of MCDM methods such as TOPSIS (Technique for Order Performance by Similarity to Ideal Solution) [11-VIKOR (VlseKriterijumskaOptimizacijaKompromisnoResenje, means 14]. Multicriteria Optimization and Compromise Solution) [15, 16], ELECTRE (Elimination and Choice Expressing The Reality) [17], PROMETHE (preference ranking organization method for enrichment evaluation) [18], COPRAS (complex proportional assessment) [19], COPRAS-G [20]. Although there are only a few studies for dissolution process in literature, a study regarding to selection the best experiment conditions for a leaching process is not available in the literature [13]. TOPSIS method is used to find a solution which is nearest ideal solution and farthest nonideal solution [14, 23].

Та	Table 1. Chemical analysis of malachite ore used in the experiments.						
Component	SiO ₂	CuO	Al ₂ O ₃	PbO	Fe ₂ O ₃	Ignition loss	Other oxides
Value, %	40.52	22.86	14.53	1.28	1.01	16.80	3.00

3.3. Criteria Weighting

The criteria weights are calculated with analytic hierarchy process (AHP) method which is a compromised weighting method [24]. AHP method was proposed first by Saaty (1977,1980) to model subjective decision making processes based on multiple attributes in a hierarchical system [25]. The method has three main steps to calculate the weights of attributes: First, structure of hierarchical system; second, comparative judgment of the alternatives and the criteria; third, synthesis of the priorities [11, 23]. In order to compare a set of "n" criteria pairwise according to their relative importance weights, pairwise comparison matrix is used and it can be represented as [23] and relative importance of between two criteria is determined using Table 2.

Table 2. Kato Scale in the AFF method [25]						
Intensity	1	3	5	7	9	2,4,6,8
Linguistic	Equal	Moderate	Strong	Demonstrated	Extreme	Intermediate value

Table 2. Ratio Scale in the AHP method [25]

4. RESULTS AND DISCUSSION

4.1. Analysis of Signal to Noise (S/N) Ratio

Time (l_c), temperature (T), stirring speed (A_r), leaching reagent of concentration (C), solid/liquid ratio (SL_r) were considered as leaching process parameters. The leaching process parameters and their levels were given in Table 3. Experimental design for five parameters of leaching process (time, temperature, stirring speed, leaching reagent of concentration, solid/liquid ratio) with five levels organized by Taguchi's L_{25} orthogonal array was shown in Table 4.

	Tuble 5. Let	aening para	meters und	levels		
Symbol	Leaching Parameters	Level 1	Level 2	Level 3	Level 4	Level 5
А	Time (<i>minute</i>)	15	30	60	90	120
В	Temperature ($^{\circ}C$)	35	45	55	65	75
С	Agitation rate (rpm)	150	250	350	450	550
D	Concentration (mol/L)	1	2	3	4	5
E	Solid/Liquid Ratio (g/mL)	1	2	4	6	8

Table 3. Leaching parameters and levels

		e		• •	
Experiment	А	В	Agitation	D	Е
No	(Time)	(Temperature)		(Concentration)	(Solid/liquid)
1	1	1	1	1	1
2	1	2	2	2	2
3	1	3	3	3	3
4	1	4	4	4	4
5	1	5	5	5	5
6	2	1	2	3	4
7	2	2	3	4	5
8	2	3	4	5	1
9	2	4	5	1	2
10	2	5	1	2	3
11	3	1	3	5	2
12	3	2	4	1	3
13	3	3	5	2	4
14	3	4	1	3	5
15	3	5	2	4	1
16	4	1	4	2	5
17	4	2	5	3	1
18	4	3	1	4	2
19	4	4	2	5	3
20	4	5	3	1	4
21	5	1	5	4	3
22	5	2	1	5	4
23	5	3	2	1	5
24	5	4	3	2	1
25	5	5	4	3	2

Table 4. Full factorial design with orthogonal array of Taguchi L25

Optimization of leaching parameters is the most critical step in hydrometallurgical processes as it is considered process costs. Therefore, the influence of each factor level on process conditions was analyzed by signal to noise (S/N) ratio. S/N ratios and level values were calculated with the condition of "the higher the best" criteria. For each level, average S/N ratio is calculated on the basis of the recorded value in Table 5. It demonstrates optimum levels of control factors for the highest percentage extraction values of copper from malachite ore. S/N ratio values and graphics are seen in Figure 3. Optimum leaching parameters of control factors for the maximum percentage recovery values of copper from malachite ore can be easily determined from Figure 2. According to Figure 2, the levels and S/N ratios for the factors giving the best percentage recovery value of copper were determined as a factor A (Level 3, S/N=-0.8207), factor B (Level 5, S/N=-1.2899), factor C (Level 4, S/N= -1.4538), factor D (Level 4, S/N=-1.5851), factor E (Level 5, S/N=-1.4629). In other words, optimum leaching conditions in this situation is A₃B₅C₄D₄E₅. S/N response table for efficiency is seen Table 6. Bold values of control factors of S/N ratios are represented optimum parameters of process.

Experimental Number	Control Factors						S/N Ratios
	A Time (<i>minute</i>)	B Temperatur e (^{O}C)	C Agitation Rate (<i>rpm</i>)	D Concentratio n (<i>mol/L</i>)	E Solid/Liquid Rate (g/mL)	-	
1	15	30	100	0.1	1	0.513	-5.798
2	15	40	150	0.3	1.5	0.625	-4.082
3	15	45	200	0.5	2	0.695	-3.16
4	15	50	250	0.7	3	0.758	-2.407
5	15	55	300	0.9	4	0.789	-2.058
6	30	30	150	0.5	3	0.711	-2.963
7	30	40	200	0.7	4	0.762	-2.361
8	30	45	250	0.9	1	0.752	-2.476
9	30	50	300	0.1	1.5	0.761	-2.372
10	30	55	100	0.3	2	0.709	-2.987
11	60	30	200	0.9	1.5	0.856	-1.351
12	60	40	250	0.1	2	0.894	-0.973
13	60	45	300	0.3	3	0.928	-0.649
14	60	50	100	0.5	4	0.933	-0.602
15	60	55	150	0.7	1	0.941	-0.528
16	90	30	250	0.3	4	0.861	-1.3
17	90	40	300	0.5	1	0.853	-1.381
18	90	45	100	0.7	1.5	0.889	-1.022
19	90	50	150	0.9	2	0.855	-1.361
20	90	55	200	0.1	3	0.916	-0.762
21	120	30	300	0.7	2	0.831	-1.608
22	120	40	100	0.9	3	0.873	-1.18
23	120	45	150	0.1	4	0.892	-0.993
24	120	50	200	0.3	1	0.936	-0.574
25	120	55	250	0.5	1.5	0.987	-0.114

Table 5. The experiment results and S/N ratios

Levels	Control Factors						
			Efficiency				
	А	В	С	D	Е		
Level 1	-3.5011	2.6027	-2.3178	-2.1796	-2.1514		
Level 2	-2.6317	-1.9955	-1.9853	-1.9186	-1.7882		
Level 3	-0.8207	-1.6599	-1.6417	-1.644	-2.0179		
Level 4	-1.1651	-1.4633	-1,4538	-1,5851	-1,592		
Level 5	-0.8937	-1.2899	-1.6138	-1.685	-1.4629		
Delta	2.6804	1,3138	0.8639	0.5945	0.6885		

Table 6. S/N response table for efficiency factor

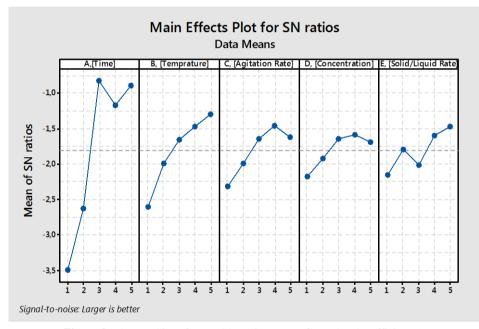


Figure 2. The graphics of mean S/N ratios versus factor levels (efficiency).

4.2. Confirmation Test

Confirmation test was performed with optimum parameters which were obtained by Taguchi method. Consequently, percentage extraction of copper from malachite ore is 0.994% which is higher than 25^{th} experiment. It has been found that Taguchi method is a suitable method to determine the optimum parameters of process.

4.3. Analysis by TOPSIS method

The applicability of TOPSIS method was investigated in order to the most suitable experimental conditions. Criteria weightings were obtained by AHP method which was used in

TOPSIS. Based on Table 2, a pairwise comparison matrix was created in Table 7. Here as criteria the time is considered most important for leaching process. Other criteria are arranged according to their importance respectively; temperature, stirring speed, solid/liquid ratio, concentration. Table 8 shows that criteria weights of leaching parameters were obtained by AHP method.

		1		1		
		Α	В	С	D	Ε
	Α	1	1.286	1.8	9	3
	В	0.778	1	1.4	. 7	2.334
	С	0.556	0.715	1	5	1.667
	D	0.112	0.143	0.2	1	0.334
	Е	0.334	0.429	0.6	3	1
		T	able 8. Cri	iteria wei	ights	
	Time	Temperature	Agitatio	n Rate	Concentration	Solid/Liquid Ratio
wj	0.360	0.280	0.20	00	0.040	0.120

Table 7. The pairwise comparison matrix for criteria

The decision matrix which was needed for TOPSIS method was determined with Taguchi's L_{25} orthogonal array which was used in leaching experiments (Table 9). The normalized decision matrix is seen from Table 10. It was multiplied by the relative importance in Table 8 and obtained the weighted normalized matrix is presented in Table 11. The ideal (V_i^+) and non-ideal solution (

 V_i^-) were obtained (Table 12). The Euclidean Distances and relative closeness to ideal solutions are presented in Table 13. The highest percentage value of extraction copper was assessed to be the 25th experiment by this method and it is also same the experiment results. The comparison of rankings obtained from the experiment and TOPSIS method is seen Figure 3. The test results and MCDM method is good agreement with each other.

Experiment	Time	Temperature	Agitation rate	Concentration	Solid/Liquid ratio		
1	15	35	100	0.1	1		
2	15	45	150	0.3	1.5		
3	15	55	200	0.5	2		
4	15	65	250	0.7	3		
5	15	75	300	0.9	4		
6	30	35	150	0.5	3		
7	30	45	200	0.7	4		
8	30	55	250	0.9	1		
9	30	65	300	0.1	1.5		
10	30	75	100	0.3	2		
11	60	35	200	0.9	1.5		
12	60	45	250	0.1	2		
13	60	55	300	0.3	3		
14	60	65	100	0.5	4		
15	60	75	150	0.7	1		
16	90	35	250	0.3	4		
17	90	45	300	0.5	1		
18	90	55	100	0.7	1.5		
19	90	65	150	0.9	2		
20	90	75	200	0.1	3		
21	120	35	300	0.7	2		
22	120	45	100	0.9	3		
23	120	55	150	0.1	4		
24	120	65	200	0.3	1		
25	120	75	250	0.5	1.5		

 Table 9. The Decision Matrix

Table 10. Normalized Decision Matrix

Experiment	Time	Temperature	Agitation Rate	Concentration	Solid/Liquid ratio
1	0.0407	0.1233	0.0795	0.0603	0.0407
2	0.0407	0.1585	0.1325	0.1206	0.0813
3	0.0407	0.1937	0.1854	0.1809	0.1626
4	0.0407	0.2289	0.2384	0.2412	0.2439
5	0.0407	0.2641	0.2914	0.3015	0.3252
6	0.0813	0.1233	0.1325	0.1809	0.2439
7	0.0813	0.1585	0.1854	0.2412	0.3252
8	0.0813	0.1937	0.2384	0.3015	0.0407
9	0.0813	0.2289	0.2914	0.0603	0.0813
10	0.0813	0.2641	0.0795	0.1206	0.1626
11	0.1626	0.1233	0.1854	0.3015	0.0813
12	0.1626	0.1585	0.2384	0.0603	0.1626
13	0.1626	0.1937	0.2914	0.1206	0.2439
14	0.1626	0.2289	0.0795	0.1809	0.3252
15	0.1626	0.2641	0.1325	0.2412	0.0407
16	0.2439	0.1233	0.2384	0.1206	0.3252
17	0.2439	0.1585	0.2914	0.1809	0.0407
18	0.2439	0.1937	0.0795	0.2412	0.0813
19	0.2439	0.2289	0.1325	0.3015	0.1626
20	0.2439	0.2641	0.1854	0.0603	0.2439
21	0.3252	0.1233	0.2914	0.2412	0.1626
22	0.3252	0.1585	0.0795	0.3015	0.2439
23	0.3252	0.1937	0.1325	0.0603	0.3252
24	0.3252	0.2289	0.1854	0.1206	0.0407
25	0.3252	0.2641	0.2384	0.1809	0.0813

Experiment	Time	Temperature	Agitation	Concentration	Solid/Liquid ratio
			Rate		
1	0.0146	0.0345	0.0159	0.0024	0.0049
2	0.0146	0.0444	0.0265	0.0048	0.0098
3	0.0146	0.0542	0.0371	0.0073	0.0195
4	0.0146	0.0641	0.0477	0.0097	0.0293
5	0.0146	0.0739	0.0583	0.0121	0.039
6	0.0293	0.0345	0.0265	0.0073	0.0293
7	0.0293	0.0444	0.0371	0.0097	0.039
8	0.0293	0.0542	0.0477	0.0121	0.0049
9	0.0293	0.0641	0.0583	0.0024	0.0098
10	0.0293	0.0739	0.0159	0.0048	0.0195
11	0.0585	0.0345	0.0371	0.0121	0.0098
12	0.0585	0.0444	0.0477	0.0024	0.0195
13	0.0585	0.0542	0.0583	0.0048	0.0293
14	0.0585	0.0641	0.0159	0.0073	0.039
15	0.0585	0.0739	0.0265	0.0097	0.0049
16	0.0878	0.0345	0.0477	0.0048	0.039
17	0.0878	0.0444	0.0583	0.0073	0.0049
18	0.0878	0.0542	0.0159	0.0097	0.0098
19	0.0878	0.0641	0.0265	0.0121	0.0195
20	0.0878	0.0739	0.0371	0.0024	0.0293
21	0.1171	0.0345	0.0583	0.0097	0.0195
22	0.1171	0.0444	0.0159	0.0121	0.0293
23	0.1171	0.0542	0.0265	0.0024	0.039
24	0.1171	0.0641	0.0371	0.0048	0.0049
25	0.1171	0.0739	0.0477	0.0073	0.0098

Table 11. Weighted normalized decision matrix

Table 12. The Ideal and non-ideal solutions

	Time	Temperature	Agitation Rate	Concentration	Solid/Liquid ratio
$V^{\scriptscriptstyle +}$	0.1171	0.0739	0.0583	0.0121	0.0390
V^{-}	0.0146	0.0345	0.0159	0.0024	0.0049

S_i^+	S_i^-	C_{i}	Rank
0.0151	0	0	25
0.0133	0.0002	0.0177	24
0.0117	0.0011	0.084	22
0.0108	0.0025	0.19	18
0.0105	0.0046	0.3054	15
0.0104	0.0009	0.0834	23
0.009	0.002	0.1797	19
0.0094	0.0017	0.154	21
0.0088	0.0029	0.2494	17
0.0099	0.002	0.1668	20
0.0063	0.0025	0.284	16
0.0049	0.0032	0.3993	13
0.004	0.0047	0.5433	11
0.0053	0.004	0.4276	12
0.0056	0.0036	0.394	14
0.0026	0.0075	0.7452	7
0.0029	0.0073	0.7133	9
0.0039	0.0058	0.5984	10
0.0023	0.0066	0.7392	8
0.0015	0.008	0.8418	4
0.0019	0.0126	0.866	5
0.0028	0.0113	0.803	6
0.0015	0.0122	0.8906	2
0.0018	0.0118	0.87	3
0.001	0.0131	0.9296	1

Table 13. S_i^+ , S_i^- and C_i

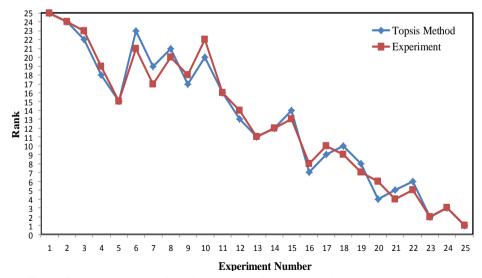


Figure 3. The comparison of rankings obtained from the experiment and TOPSIS method

5. CONCLUSIONS

There are several attributes which influence the optimization of leaching parameters of copper from malachite ore in the presence ammonium nitrate solution. Some of these conditions are time, temperature, stirring speed, concentration of leaching reagent, solid/liquid rate. The various process conditions have been defined, compared and ranked with the aid of Taguchi and MCDM methods, which will maximize the amount of copper passing into solution.

The results obtained from experiments were analyzed with Taguchi method. Taguchi analyses were clearly indicated that the most efficient experiment combination of attributes time, 60 min; temperature, 75°C; stirring speed, 450 rpm; concentration of leaching reagent, 4 mol/L and solid/liquid ratio, 8 g/mL. This result was confirmed with a new experiment in above-mentioned conditions. Consequently, the result of the confirmation experiment was higher compared with performed by Taguchi L_{25} orthogonal array previously. It can be inferred that the higher temperature and solid/liquid ratio were found to be highly ranked with better leaching performance. However, the acid concentration, stirring speed and time also play a significant role in order to achieved better leaching performance.

At the second part of this study, TOPSIS method was used for rank the experiment conditions for better to worst efficiency which was created with Taguchi L₂₅ orthogonal array without doing any experiment. It was clearly reveals that the results are good agreement with obtained TOPSIS method. Consequently, 25th experiment has the best efficiency compared to the obtained results of the experiments and TOPSIS method. It is determined that the highest percentage recovery of copper from malachite ore can be achieved using only experimental design data without performing any experiments. For future works, this process can be also extended with using various MCDM methods.

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