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Improvement of the Metal Extraction and Removal of Harmful Impurities from Sulfide Ores by Polyoxometalate Oxidizers: Design of Experiments and Industrial Modeling

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ABSTRACT

In the present study, a new method has been suggested to solve the problems of the very low solubilityof sulfide ores in acidic solution and also the production of toxic impurities for the first time. In this work, the polyoxometalate (POM) oxidizer was applied for the dissolution of sulfide ores, extraction of metals, and removal of toxic and harmful wastes. In this procedure, POMs were used as strong oxidizers of sulfur compounds to dissolve sulfide ores. Also, acid was applied as a solvent and catalyst to increase the reaction rate. The Taguchi experimental design along with the ProMax simulation software was applied for studying the leaching of sulfide ores by POM oxidizers as a novel plan in experimental to industrial scales. The optimum data achieved by the Taguchi method was used as the input data to the simulation and sensitivity analysis of the process was executed by the ProMax software. The effects of curicital operating parameters such as the concentration of acid (C_A) in the 60-90 g/l range, the reaction temperature (T_R) with the values of 60-90 ºC, the rotation rate (R) with the amounts of 50- 300 rpm, the retention time (τ) in the 0.5-2.0 h range, the concentration of polyoxometalate oxidizer with the values of 0.1- 0.5 g/l, the acid types of H_2SO_4, HNO_3, HCl, H_3PO_4, the grain sizes of sulfide ores (Sparticle) in the 0.5-3.0 mm range and polyoxometalate with the types of $[Mo_6O_{19}]^{2-}$, $[Mo_8O_{26}]^{4-}$, $[V_{10}O_{28}]^{6-}$ and $[H_2W_{12}O_{40}]^{10-}$ on the extraction efficiency of metals and removal of toxic heavy metals from sulfide ores by polyoxometalates were investigated. The optimum conditions to extract maximize metals from the sulfide ores were obtained as the C_A; 80 g/l, T_R; 90 ºC, R; 300 rpm, τ; 1.0 h, m POMs; 0.5g/l, acid type of H_2SO_4, S_{grain};1.0 mm and POMs type of $[H_2W_{12}O_{40}]^{10-}$. Under optimized conditions, the extraction efficiency of zinc, copper, and lead and the removal of toxic heavy metals from sulfide ores were determined as above 85%, 81%, 83%, and 99.9% receptivity.

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1. Introduction

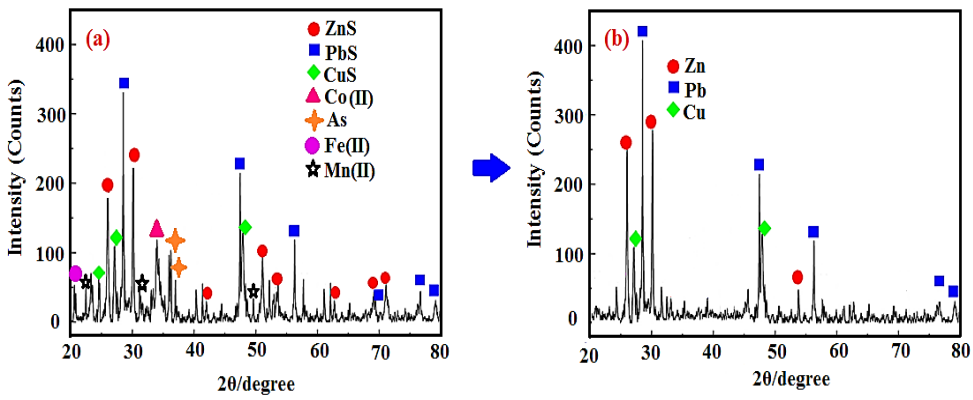
In recent years, due to the depletion of the reserves of oxide ore minerals, the reserves of sulfide ore minerals have received much attention [1, 2]. Sulfide ore minerals contain the largest and widest reserves of precious metals including zinc, copper, and lead throughout the world, but reasons such as environmental problems, high temperatures and pressures (autoclave) required for their dissolution, the production of harmful and toxic by-products, their very low solubility constant in acids ($Ksp<10^{-21}$), especially sulfuric acid compared to oxidized mineral ores ($Ksp = 10^{-16}$) have made it difficult to work with sulfide ores [3, 4]. Nowadays, in industrial plants, three methods are applied to dissolve sulfide ores; 1) roasting operation at high temperatures of between 1000 to 1200 ^{o}C where sulfides ores are heated inside a furnace. This method requires a lot of energy and costs high and the number of impurities (formation of ferrite compounds (MFe_2O_4)) in it is great [5, 6]. 2) pressurized tanks (autoclave); this is a dangerous method and there is a possibility of the explosion of tanks in it. Also, the dissolution of sulfide ores in the sulfuric acid solution at high temperatures and pressures release hazardous and toxic gases which will cause many problems if not removed. If these harmful gases are aggregated more than the permissive limit in the atmosphere, they can be harmful to health [7, 8]. 3) iron sulfate additive; the efficiency and speed of this method are very low [9-11]. Therefore, researchers are trying to find new and safe methods with high-efficiency. A novel and suitable alternative method for the mentioned methods is the hydrometallurgical Polyoxometalates (POMs) oxidizer method. The hydrometallurgical approach by POM strong oxidizers is a novel method for the dissolution of sulfide ores in the acidic solvents. Polyoxometals have attracted the attention of research groups in less than a few decades so they have been extensively studied in various fields of catalysts, pharmaceuticals, photochromism, and magnetic materials. POMs belong to a group of metal-oxygen clusters that have gained great importance, due to their unique properties and structure, in various fields, especially in the field of catalysts. The metals used in these compounds are usually W, Mo, and V, which are in their high oxidation state. This allows these compounds to easily accept electrons and perform their catalytic role as acidic and oxidative catalysts [12]. The dissolution of sulfide ores via the hydrometallurgical method by POM oxidizers has advantages over the above methods, which are including; no production of toxic and harmful gases and wastes, low environmental pollution, high extraction efficiency, the production of recyclable by-products, no need for high temperatures and pressures and avoiding the dangers caused by them, low cost and less energy consumption. The process of the dissolution of sulfide ores by POMs oxidizer can be divided into four key sections: 1) the stage of the leaching of sulfide ores in different acid solutions by POM oxidizers, 2) the removal of toxic heavy metals by POM oxidizers, 3) the section of the negation of the acid solution with $Ca(OH)_2$, 4) the stage of refining that is divided to two sub-sections; a) cobalt purification (removal of cobalt, iron, and manganese) and refining nickel(removal of nickel and cadmium), b) the extraction of desired metals (Zn, Cu, and Pb) . The contemporaneous utilization of the Taguchi procedure and ProMax software is a strong method that can be utilized to predict the optimum situation, simulate the process, and study the performance of the system from the experimental scale (pilot) to industrial scale [13, 14]. Also, the main purpose of this work

can be presented in three parts; (1) to experimentally design and optimize the removal of heavy metals and the condition of extracting them from sulfide ores by POM oxidizers in experimental scales by using the Taguchi procedure (2) to utilize the obtained optimum condition as the input data to the ProMax software for the simulation of the process of the operation in industrial scales, (3) to apply the final simulation results as data required for the design and construction of industrial plants. In industrial processes, modeling is very suitable to evaluate the execution of the system. In this novel work, the sensitivity analysis of operating conditions such as the concentration of acids (C_A) in the range of 60-90 g/l, the reaction temperature (T_R) with the value of 60-90 ºC, the rotation rate (R) with the amount of 50- 300rpm, the retention time (τ) being in the range of 0.5-2.0h, the polyoxometalate values of 0.1- 0.5g/l, the acid type of H_2SO_4, HNO_3, HCl, H_3PO_4, the grain size of sulfide ores ($S_{particle}$) in the range of 0.5-3.0 mm and polyoxometalate with the types of $[Mo_6O_{19}]^{2-}$, $[Mo_8O_{26}]^{4-}$, $[V_{10}O_{28}]^{6-}$, and $[H_2W_{12}O_{40}]^{10-}$ are investigated. The basic goal of the present project is to prospect a novel plan for the dissolution of sulfide ores and extraction of metals with high purities and efficiencies by using POM oxidizers in acid solutions.

2. Experimental, Optimization and Modeling Procedures

2.1 Experimental Procedure

Firstly, the required sulfide ores contain sphalerite (ZnS) ore comprising galena (PbS) and chalcopyrite ($CuFeS_2$) was obtained from Anguran mines in Iran. The dissolution of sulfide ores with a 10 g value by POM oxidizers was experimented with a 1liter of different acid solution. Then provided solutions were stirred on a magnetic stirrer. In each test, 10 g of sulfide ores was mixed with POM oxidizers in the sulfuric acid solution. After the leaching of sulfide ores by POM oxidizers in the sulfuric acid solution , the attained solution was analyzed by Atomic Absorption Spectrometer (AAS) (model of Varian-240Z) and X-ray diffractometer to characterize the values of the extracted metals of zinc, copper, and lead and also the removal percentage of toxic heavy metals. The X-ray diffractograms of the sulfide ores before and after the removal of toxic heavy metals are presented in Fig. 1. Also, the chemical compounds of the sulfide ores were characterized by X-ray fluorescence spectrometer (XRF, model of EA1000VX), and the results are given in Table 1.



(a) (b)

Figure 1. XRD patterns of sulfide ores; (a) before and (b) after the removal of toxic heavy metals

Table 1
Chemical composition and weight percentage of sulfide ores

Elements	Wt.%	Content	Wt.%
Zn	32.786	ZnS	41.472
Pb	5.946	PbS	6.135
Cu	0.753	$CuFeS_2$	1.693
Fe	20.731	Fe_2O_3	5.957
Mn	0.325	Al_2O_3	2.135
Ca	12.604	Cr_2O_3	0.276
Ni	0.045	As_2O_3	0.184
Co	0.026	P_2O_5	0.063
Si	8.531	K_2O	3.175
As	0.089	CaO	3.014
Cd	0.099	MgO	0.671
Sb	0.054	MnO_2	0.299
Sulfur	15.967	Na_2O	2.768
Ge	0.042	SiO_2	21.549
Cr	0.016	FeS_2	7.0543
Tl	0.005	Other	0.949
Other	1.984		

In the next step, the experimentally achieved results were used for the experimental design and process simulations. The extraction percentage of Zn, Cu, and Pb metals was calculated by:

$$\%R = \frac{VM_1}{mM_0} \times 100 \quad (1)$$

where R is the extraction percentage of metals, M_1(g/L) is the concentration of the final solution; V (l) is the volume of the leach solution; M_0(%) is the concentration of the first solution and m (g) is the mass of the sulfide ore used in the experiment. The removal percentage of toxic heavy metals from the sulfide ore is determined by Eq. (2) as follows:

$$\%\,Toxic\ heavy\ metals\ removal = \left(\frac{C_0 - C}{C_0}\right) \times 100 \quad (2)$$

where C_0 and C are the initial and equilibrium concentrations of impurities (mg/L) respectively.

2.2. Factors Optimization by the Taguchi Experimental Design

The Taguchi procedure is one of the impressive designs utilized by engineers and researchers. The Taguchi method compared to factor methods has advantages such as less number of tests and therefore less cost and time of testing, the ability to study the interactions and perform experiments in parallel, and ultimately predict the optimal response [16]. The stages of the Taguchi experimental design are as follows: (1) to determine the variables and their levels, (2) to determine the number of main and key tests, (3) to investigate the analysis of variance (ANOVA) data, and (4) to predict the optimum situations for the performance, cost and quality of the process [17, 18]. In the present research, 8 parameters with 4 levels were chosen for the test design. Parameters and their levels in twenty tests are reported in Table 2. The list of parameters is considered based on the reducing order of their effects on the presented test response as follows: (the concentration of the acid (C_A) (P_1) with the values of 60, 70, 80, and 90 g/l was considered as the first parameter affecting the extraction of the metals by POM oxidizers, the reaction temperature (T_R (P_2)) with the values of 60, 70, 80, and 90 °C was chosen as the second effective parameter. The other affecting parameters were the rotation rate (R (P_3)) with the amounts of 50, 100, 200, and 300 rpm, the reaction time (τ (P_4)) with the amounts of 0.5, 1.0, 1.5, and 2.0 h, the amount of POM oxidizers (m_{POMs} (P_5)) with the values of 0.1, 0.2, 0.3, and 0.5 g/l and the acid types of H_2SO_4, HNO_3, HCl, and H_3PO_4 (P_6)) are the third, fourth, fifth and sixth factors respectively. Finally, the grain size (S_{grain} (P_7)) with the values of 0.5,1.0, 2.0, and 3.0 mm and the POMs (tp (P_8)) with the types of $[Mo_6O_{19}]^{2-}$, $[Mo_8O_{26}]^{4-}$, $[V_{10}O_{28}]^{6-}$, and $[H_2W_{12}O_{40}]^{10-}$ were considered as the seventh and eighth parameters (Table 2).

Table 2
Parameters and their levels

Parameters	Levels			
	1	2	3	4
P_1. Acid concentration (g/l)	60	70	80	90
P_2. T_R (°C)	60	70	80	90
P_3. R (rpm)	50	100	200	300
P_4. τ (min)	0.5	1.0	1.5	2.0
P_5. m_{POMs} (g/l)	1	2	3	5
P_6. Acid type	H_2SO_4	HNO_3	HCl	H_3PO_4
P_7. S grain (mm)	0.5	1.0	2.0	3.0
P_8. POMs type	$[Mo_6O_{19}]^{2-}$	$[Mo_8O_{26}]^{4-}$	$[V_{10}O_{28}]^{6-}$	$[H_2W_{12}O_{40}]^{10-}$

The effect of factors was optimized by the Taguchi method for enhancing the extraction of metals and removal of toxic heavy metals by leaching of sulfide ores using POM oxidizers. According to the number of factors and their levels, the L_{32} array (thirty-two tests) designed by the Taguchi method was suggested for this work (Table 3).

Table 3
L_{32} orthogonal array.

Exp. No.	Factors and their levels											
	P_1	P_2	P_3	P_4	P_5	P_6	P_7	P_8	Y_1	Y_2	Average	S/N
Test1	1	1	1	1	1	1	1	1	44.64	46.78	45.71	53.53
Test 2	1	1	2	2	2	2	2	2	54.37	51.08	52.72	57.96
Test 3	1	1	3	3	3	3	3	3	44.54	42.31	43.42	52.56
Test 4	1	1	4	4	4	4	4	4	57.64	59.03	58.33	58.96
Test 5	1	2	1	2	3	4	1	2	73.56	74.13	73.84	65.12
Test 6	1	2	2	3	4	1	2	3	53.32	54.17	53.88	55.18
Test 7	1	2	3	4	1	2	3	4	67.62	65.87	66.74	62.54
Test 8	1	2	4	1	2	3	4	1	57.68	58.53	58.10	58.67
Test 9	1	3	1	3	2	4	3	1	74.68	73.45	74.06	66.98
Test 10	1	3	2	4	3	1	4	2	44.15	44.27	44.21	53.21
Test 11	1	3	3	1	4	2	1	3	62.53	63.64	63.08	61.45
Test 12	1	3	4	2	1	3	2	4	54.51	55.67	56.58	57.73
Test 13	1	4	1	4	2	3	3	1	48.07	47.89	47.98	54.16
Test 14	1	4	2	1	3	4	2	4	56.78	54.63	55.70	56.49
Test 15	1	4	3	2	4	1	3	1	76.57	75.34	75.95	67.19
Test 16	1	1	4	3	1	2	4	2	59.67	60.89	60.28	60.06
Test 17	2	1	1	2	4	2	4	3	51.98	49.75	50.86	51.85
Test 18	2	1	2	3	1	3	1	4	66.82	65.83	66.32	62.45
Test 19	2	1	3	4	2	4	2	1	75.94	77.61	76.77	67.72
Test 20	2	1	4	1	3	2	3	2	54.24	55.69	54.96	55.33
Test 21	2	2	1	3	4	3	2	2	43.78	41.72	42.75	51.79
Test 22	2	2	2	4	1	4	3	3	66.21	63.75	64.98	62.37
Test 23	2	2	3	1	2	1	4	4	59.73	60.25	59.99	59.42
Test 24	2	2	4	2	3	2	1	1	65.47	66.84	66.15	62.13
Test 25	2	3	1	4	3	2	2	1	67.11	69.24	68.17	62.96
Test 26	2	3	2	1	4	3	3	2	60.17	59.53	59.85	58.64
Test 27	2	3	3	2	1	4	4	3	53.54	54.93	54.23	56.86
Test 28	2	3	4	3	2	1	1	4	40.53	42.13	41.33	51.25
Test 29	2	4	1	1	1	2	2	3	66.41	65.78	66.09	63.62
Test 30	2	4	2	2	2	3	3	4	61.34	62.74	62.04	62.56
Test 31	2	4	3	3	3	4	4	1	56.89	58.62	57.75	58.23
Test 32	2	4	4	4	4	1	1	2	45.32	44.67	44.99	52.90

Table 3 exhibits the number of duplicated tests based on the Taguchi approach. The S/N ratio refers to improving the quality by reducing deflections in the lowest tests. To investigate the effect of noise sources on the dissolution process of sulfide ores, each test was carried out three times under a similar condition. In the Taguchi procedure, if the tests are duplicated more than once, the Signal/Noise (S/N) ratio is the best approach for evaluating the impact of the total perversion of the mean value from the purpose amount. The application of signal-to-noise (S/N) ratios in the Taguchi approach was recommended for the optimization of the parameters of the process [19]. The extraction of metals from sulfide ores by POM oxidizers was selected as the test responds for each test as greater Signal/Noise (SNL). Since the basic focus of the current work is to enhance the extraction of metals (S/N ratios for "greater is better") was considered based on response amounts. To study the effects of parameters on the extraction of metals, the analysis of variance (ANOVA) was performed and the achieved data are given in Table 4. The ANOVA table represents the contribution of each parameter to the response.

Table 4
Results of the ANOVA

Parameters	**Optimum operation condition**			
	Degree of freedom (f)	Sum of square (S)	Variance (v)	F- ratio
P_1. Acid concentration (g/l)	3	134.51	207.34	174.64
P_2. T_R (°C)	3	123.45	113.26	69.85
P_3. R (rpm)	3	43.54	29.53	19.75
P_4. τ (min)	3	27.69	21.91	35.75
P_5. m_{POMs} (g/l)	3	778.58	671.69	316.43
P_6. Acid type	3	238.85	232.17	153.25
P_7. S grain (mm)	3	228.13	214.35	97.89
P_8. POMs type	3	73.64	55.63	49.78

The essential situation for each parameter to optimize the extraction of metals was presented in Table 5. The optimal situations to maximize the extraction efficiency of metals (Zn, Cu, and Pb) and the removal of toxic heavy metals were chosen at the concentration of the acid (C_A) in the range of 60-90 g/l, the reaction temperature (T_R) with the value of 60-90 ºC, the rotation rate (R) with the amount of 50- 300 rpm, the retention time (τ) in the range of 0.5-2.0 h, the amount of POMs with the values of 1-5g/l, the acid types of H_2SO_4, HNO_3, HCl, and H_3PO_4, the grain size of sulfide ores ($S_{particle}$) in the range of 0.5-3.0 mm and POM oxidizers with the types of $[Mo_6O_{19}]^{2-}$, $[Mo_8O_{26}]^{4-}$, $[V_{10}O_{28}]^{6-}$, and $[H_2W_{12}O_{40}]^{10-}$. According to the results presented in Table 5, the concentration of acids, the reaction temperature, the amount of POM oxidizers, and the grain size of sulfide ore have the highest impact on the extraction of metals, while the type of POMs, rotation rate, and retention time have no considerable impact within our working range. Also, the amount of POM oxidizers had the highes effect on the removal of toxic heavy metals.

Table 5
The optimized results

Parameters	Optimum operation condition		
	Level description	Level	Contribution (%)
P_1. Acid concentration (g/l)	80	3	10.65
P_2. T_R (°C)	90	4	8.46
P_3. R (rpm)	300	4	1.34
P_4. τ (min)	1.0	2	3.52
P_5. m_{POMs} (g/l)	0.5	4	34.89
P_6. Acid type	H_2SO_4	1	25.78
P_7. S grain (mm)	1.0	1	11.20
P_8. POMs type	$[H_2W_{12}O_{40}]^{10-}$	4	4.13
Total			99.97
Error	—	—	0.03

2.3. Process Modeling and formulation

The ProMax software is one of the most powerful chemical engineering software. This software is widely utilized for the simulation of processes in pilot to industrial scales. The basic sections in the simulation with the ProMax software are:1) to choose a list of materials 2) to select the fluid package 3) to choose flowsheet 4) to simulate operational equipment such as tanks, pumps, boilers, compressors 5) to provide results [25]. Fig. 2 shows the modeled flowsheet of the industrial process of the extraction of zinc, copper, and lead from sulfide ores by the ProMax software. The achieved data from the Taguchi method which are given in Table 6, are applied as initial data for the process simulation by the ProMax software. In the simulated flowsheet by the ProMax software, the extraction of metals and removal of toxic heavy metals from sulfide ores were considered. In this process, sulfide ores were dissolved in different solutions of acids (as reaction catalysts and solvents) by POM oxidizers. In the presented plan, the POM oxidizer compounds were applied as oxidizing agents to oxidize the sulfide impurities of ores. Key information about modeling factors and process specifications are illustrated in Fig. 2 and Table 6. Also, a schematic flowsheet for a novel plan for the extraction of metals and removal of toxic heavy metals from sulfide ores by POM oxidizers in the present of acid catalysts is represented in Fig. 3.

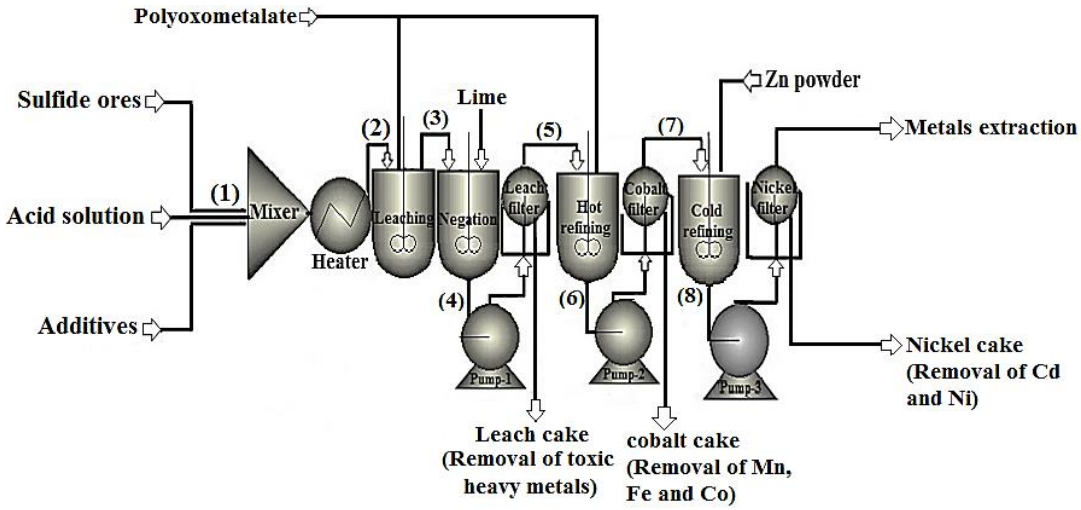


Figure 2. Modeling flowsheet for the extraction of metals and removal of toxic heavy metals from sulfide ores by the Polyoxometalate oxidizer using the ProMax software.

Table 6
Optimum conditions attained from the Taguchi procedure for the **extraction of** metals from sulfide ores by the Polyoxometalate oxidizer

Blocks				**Streams**			
CSTR-1		**CSTR-2**		**Feed stream**		**H_2SO_4 stream**	
T_R /°C	90	T_R /°C	90	$F_{Sulfide\ ores}$/kmol/h	4.022	F/(kmol/h)	18.743
R/ rpm	300	R/rpm	300	$_{POMs}$/kmol/h	0.00542	T_R/°C	25
τ/ h	1.5	τ/h	1.5	T_R /°C	25	P/bar	1.0
C_A/g/l	80	m_{POMs}/(g/l)	0.5	P/bar	1.0	pH	1.5
m_{POMs}/(g/l)	0.5	C_A/g/l	80				
Acid type	H_2SO_4	pH	4.5-7				
pH	1.5						
Pumps		**Filters**		**$Ca(OH)_2$ stream**			
T_R/°C	90	pH filter-1	4.5-4.8	F/(kmol/h)	6.748		
P/bar	170	T (°C)	70	T_R /°C	25		
		pH filter-2	4.8-5.2	P/bar	1.0		
Heater		T(°C)	70	pH	12		
T/(°C) H-1	90	pH filter-3	4.8-5.2				
P/(bar) H-1	1.7	T (°C)	70				
T/(°C) H-2	90						
P/(bar) H-2	1.0						

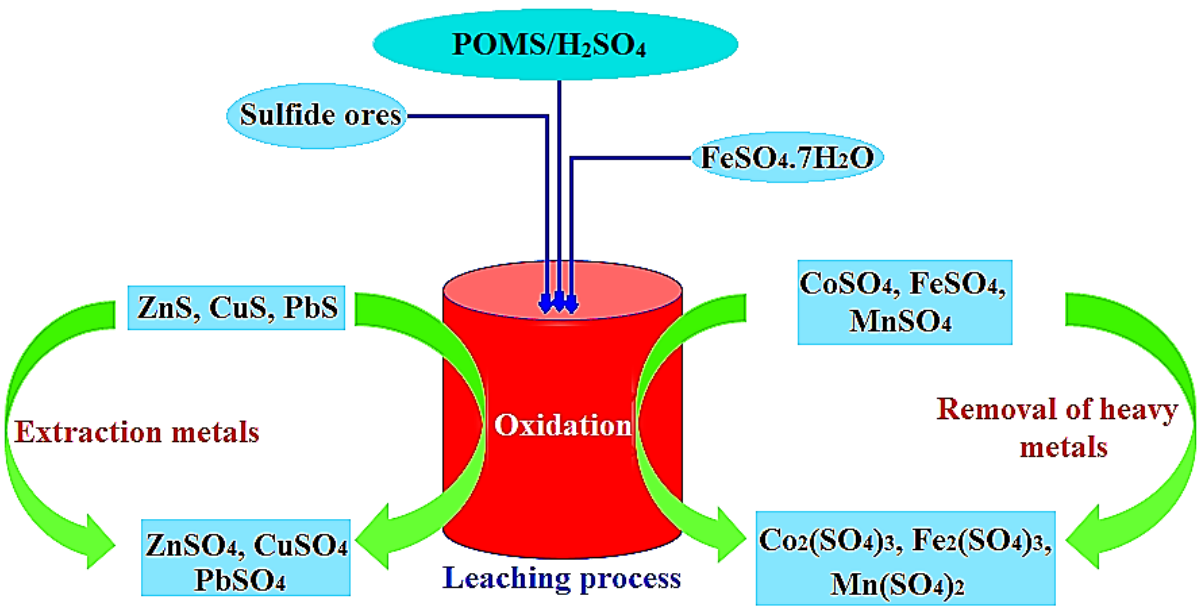


Figure 3. Schematic diagram of the suggested configuration for the extraction of metals and removal of toxic heavy metals from sulfide ores by using the Polyoxometalate oxidizer.

At the beginning of the modeling, the sulfide ores were dissolved by POM oxidizers in an acid solution at the leaching reactor (first reactor). The equations of the reactions of sulfide ores by POM oxidizers in this stage that were provided by the ProMax software are as follows;

$$ZnS\ (s) + POMs\ oxidizer + acid \rightarrow ZnSO_4\ (aq) \quad \text{(R-1)}$$

$$CuS\ (s) + POMs\ oxidizer + acid \rightarrow CuSO_4\ (aq) \quad \text{(R-2)}$$

$$PbS\ (s) + POMs\ oxidizer + acid \rightarrow PbSO_4\ (aq) \quad \text{(R-3)}$$

The acid dissolution of sulfide ores is performed in the leaching reactor which can include one or more acidic tanks. Generally, the reaction rate equation (conversion fraction(x)) of an ore dissolution in an acid solution can be expressed by the following equation:

$$M(M=Zn,\ Cu\ and\ pb)X\ (X=O,\ S,\ \ldots)\ (s) + Acid\ (l) \rightarrow M^{n+}(aq) + H_2X\ (l\ or\ g) \quad \text{(R-4)}$$

$$\text{Reaction rate} = -\,dC/dt = k \times C_{MX} \times C_{Acid}$$
$$-\int_{C_0}^{C} dC/C_{MX} = k \times C_{acid} \times \int_0^t dt \rightarrow -LnC/C_0 = k \times C_{acid} \times t \rightarrow$$
$$C = C_0(1-x_{MX})\ \&\ C_{acid} = M_{acid}/n \times 10^{-pH} \rightarrow$$
$$x_{MX} = (1-\exp(-k/n.t.10^{-pH})) \times 100,\ \ k = Zp\ \exp(-E_a/RT) \quad \text{(Eq-3)}$$

where x_{MX}, pH, k and t are the conversion fraction of ore, solution pH, rate constant, and reaction time. By raising the number of acid tanks, the acid retention time and dissolution of sulfide ores increase. As a result, the acidic efficiency increases. The acid retention time can be determined by the following relation:

$$\tau_A = \frac{(24hr) \times n_A \times V_A \times \rho_A \times \Delta C}{W_{ore} \times R_{ore} \times P_{ore} \times (1-W_{H2O})_{ore}} = \frac{(24hr) \times n_A \times V_A \times C}{W_{ore}} \quad \text{(Eq-4)}$$

where τ_A (hr) is the acid retention time, V_A (m^3) is the volume of acid tanks, n_A is the number of the acid tanks, ρ_A is the density of the leaching solution, m_{ore} (kg) is the amount of sulfide ores, R_{ore} (%) is the dissolution efficiency sulfide ores, W_{H2O} (%) is the wet percentage of sulfide ores, P_{ore} is the grade of utilized sulfide ores (%), ($\Delta C = C_{make\ up} - C_{spent}$); is the difference of the fresh solution and spent concentration (kg/m^3). The amounts of sulfide ores required to extract m (tons) of pure metals are calculated by the following equation:

$$W_{ore} = \frac{m}{R_{ore} \times P_{ore} \times (1-W_{H2O})_{ore}} = \frac{N.\ V.\ C}{R_{ore} \times P_{ore} \times (1-W_{H2O})_{ore}} \quad \text{(Eq-5)}$$

$$R_{ore}\ (\%) = \left(1 - \frac{n \times p_{waste} \times (1-W_{H2O})_{waste}}{p_{ore} \times (1-W_{H2O})_{ore}}\right) \times 100 \quad \text{(Eq-6)}$$

where W_{ore}, m, R_{ore}, P_{ore}, $(W_{H2O})_{ore}$, $(W_{H2O})_w$, n, N, V, and C are the mass of raw sulfide ores (ton), the mass of extracted pure metals (ton), the dissolution efficiency of sulfide ores by

POM oxidizers in H_2SO_4 (%), the grade of sulfide ores (%), the wet percentage of sulfide ores (%), the wet percentage of the produced waste of the process (%), the mass ratio of the output waste to the mass of raw sulfide ores and the grade of the specified metal in the output waste, the number of dissolution tanks, the volume of the unit tank (m^3) and the concentration of the solution respectively. The value of the required ore to change the concentration of the desired metal in the solution from C_0 to C, the amount of the acid required to change the concentration of the acid of a solution from pH_0 to pH, and also the amount of the lime required to neutralize the solution are used from following relationships:

$$[M^{2+}] - [M^{2+}]_0 = \frac{W_{ore}}{N \times V \times R_{ore} \times P_{ore} \times (1 - W_{H2O})} \quad \text{(Eq-7)}$$

$$[H^+] - [H^+]_0 = \frac{m_{H2SO4}}{2 \times N \times V \times P_{H2SO4}} \quad \text{(Eq-8)}$$

$$[OH^-] - [OH^-]_0 = \frac{m_{Ca(OH)2}}{2 \times N \times V \times P_{Ca(OH)2}} \quad \text{(Eq-9)}$$

where N, V, P and m are the number of tanks, the volume of tank (m^3), the purity percentage of the considered substance (%), and the mass value of the considered substance (kg). To calculate the amount of spent required to dilute the high-concentration content of the related tanks and bring their concentration to a balanced amount using the following equation:

$$(C - C_0)_{dissolution} \times V_{dissolution} = C_{spent} \times V_{spent} \quad \text{(Eq-10)}$$

where C, C_0, $V_{dissolution}$, C_{spent}, and V_{spent} are the final concentration of dissolution solution, the initial concentration of dissolution solution, the concentration of the spent, and the volume of the spent.

One of the important factors in designing an industrial unit is knowing the number of tanks and the number of filters required for the process. The following equations can be used to calculate the number of acid leach tanks and the number of dissolution filters required to produce m tons of zinc, copper, and lead:

$$N_{tank} = \frac{m_{Metal}}{V_{tank} \times C_{Metal}} \quad \text{(Eq-11)}$$

$$N_{filter} = \frac{Wore}{m_{cake} \times N_{Filter\ plate} \times N_{Filter}} \quad \text{(Eq-12)}$$

where N_{tank} and N_{Filter} are the indicated number of required tanks for the acid leaching stage (or cake washing stage) and the number of required filters to filter the solution resulted from the dissolution of sulfide ore and cake wash solution. In the above equations, m_{Metal}, V_{tank}, C_{Metal}, W_{ore}, m_{cake}, $N_{filter\ plate}$, and N_{filter} are the amount of metal contained in the ore or cake, the volume of a considered tank, the concentration of the considered solution, the amount of consumed ore or cake, the weight of the cake produced by a filter plate, the number of filter plates of a filter and the number of filters respectively. Also, to obtain the volume of the required water to prepare a leaching solution or the volume of the solution produced with a certain concentration can be used from the following equation;

$$V\ (m^3) = \frac{W \times R \times P \times (1 - W_{H2O})}{C} \quad \text{(Eq-13)}$$

where V is the volume of water required, W is the weight of the sulfide ore or cake, R (%) is the dissolution efficiency of the sulfide ore or cake, W_{H2O} (%) is the wet percentage of the ore or cake, P is the grade of the utilized ore or cake (%), and C is the leaching solution with a specified concentration. This relation states how much water is needed to prepare a leach slurry solution from sulfide ores. Another

important issue in the leaching of sulfide ores is the dissolution efficiency of the acid. The concentration of the leaching solution after the dissolution of sulfide ores by POM oxidizers in the H_2SO_4 solution can be obtained from the following equation:

$$[M^{2+}]_A\,(M) = \frac{W_{ore} \times R_{ore} \times (1 - W_{H2O}) \times p_{ore}}{M_{metal} \times V_A \times N_{tank}} \quad \text{(Eq-14)}$$

In dissolving all types of mineral ores, especially sulfide ores in acidic solutions, the efficiency of the dissolution of acids is very important and vital. Because sulfide ores have low solubility and do not dissolve in sulfuric acid under normal conditions. The dissolution efficiency of acids can be calculated by the following equation:

$$R_A\,(\%) = \frac{V_A \times C_A - V_{Spent} \times C_{Spent}}{W_{ore} \times p_{ore} \times (1 - W_{H2O})_{ore}} \times 100 \quad \text{(Eq-15)}$$

where R_A, V_A, C_A V_{spent}, C_{spent}, m_{ore}, p_{ore}, w_{ore} are the acidic efficiency of the dissolution of sulfide ores in the H_2SO_4 solution (%), all volume of the acid leaching solution (m^3), the concentration of the acid leaching solution (kg/m^3), the volume of the produced electrolysis spent solution (m^3), the concentration of the spent solution (kg/m^3), the weight of applied raw sulfide ores (kg), thr grade of sulfide ores (%), the moisture content of sulfide ores (%) respectively. According to the sensitivity analysis results of the ProMax software, more than 70% of metals were extracted from sulfide ores by POM oxidizers. Also, the extraction values of metals from sulfide ores can be determined by the following equation;

$$R_{extraction}\,(\%) = \frac{F_{ore} - F_{cake}}{F_{ore}} \times 100 \quad \text{(Eq-16)}$$

The molar flow rates of sulfide ores and cake are expressed by:

$$F_{ore}(\text{kmol/h}) = \frac{W_{ore} \times (1 - w_{H_2O}) \times p_{ore}}{M_{ore} \times t} \quad \text{(Eq-17)}$$

$$F_{cake}(\text{kmol/h}) = \frac{W_{cake} \times (1 - W_{H_2O}) \times p_{cake}}{M_{cake} \times t} \quad \text{(Eq-18)}$$

where η_{ore} (kmol/h) and η_{cake}(kmol/h) are the molar flow rates of the sulfide ore and cake, m_{ore} and m_{cake} are the mass of the sulfide ore and cake (ton); W_{H2O} (%) is the wet percentage of the sulfide ore and cake; p_{ore} and p_{cake} (%wt./wt.) are the grades of the sulfide ore and waste; M_{ore} and M_{cake} (g/mol) are the molecular mass of the sulfide ore and cake respectively and τ (h) is the retention time. In the second reactor (that can include one or more neutralization tanks), the negation process of the dissolution of the acid solution with $Ca(OH)_2$ is performed to raise the solution pH to the values in the range of the 4.5- 7.0. One of the important products that are formed in the neutralization stage is $MSO_4.3M(OH)_3.4H_2O$ (B.M.S) (M: Zn, Cu, Pb, Fe, Co, Cd, Ni, and …) compound. In fact, at pH values of above 4, divalent metals are precipitated and concentrated in the form of the $MSO_4.3M(OH)_3.4H_2O$ (B.M.S) compound. The concentration of the negation

solution after the dissolution of B.M.S in the acid solution can be attained from the following equation:

$$[M^{2+}]_N\,(M) = \frac{W_{B.M.S} \times R_{B.M.S} \times (1-W_{H2O}) \times p_{B.M.S}}{V_N \times N_{tank}} \quad \text{(Eq-19)}$$

Also, the amounts of B.M.S required to extract m (tons) pure metal is determined by the following relation:

$$W_{B.M.S} = \frac{m_{metal\ content}}{R_{B.M.S} \times P_{B.M.S} \times (1-WH2O)B.M.S} = \frac{R_{cake} \times P_{cake} \times (1-WH2O)cake \times W_{cake}}{R_{B.M.S} \times P_{B.M.S} \times (1-WH2O)B.M.S} \quad \text{(Eq-20)}$$

where R_{cake}, P_{cake}, $(1-WH2O)cake$, W_{cake}, $R_{B.Z.S}$, $P_{B.Z.S,}$ and $(1-WH2O)_{B.Z.S}$ are the washing efficiency of the cake (%), the grade of the washed cake (%), the cake moisture (%), the cake mass (Kg), the B.M.S efficiency (%), the grade of the produced B.M.S (%) and the moisture of the produced B.M.S (%), respectively. One of the key factors in the neutralization stage is the neutralization retention time. The neutralization retention time can be calculated by the following equation:

$$\tau_N = \frac{(24\ hr) \times W_{B.M.S}}{R_{B.M.S} \times (1-W_{H2O})_{B.M.S} \times p_{B.M.S} \times N \times V_N \times \rho_{B.M.S}} \quad \text{(Eq-21)}$$

where t_N (hr) is the neutralization retention time, V (m^3) is the volume of neutralization tanks, N is the number of neutralization tanks, $\rho_{B.Z.S}$ is the density of the B.M.S slurry, m (kg) is the weight of the pure metals in the B.M.S slurry, $R_{B.M.S}$ (%) is the efficiency of the produced B.M.S, W_{H2O} (%) is the wet percentage of B.M.S, $P_{B.M.S}$ is the grade percentage of the utilized B.M.S. Then, the frustrated solution is reserved for the leaching filters for extracting the liquid phase from the solid phase. The reactions performed in the neutralization section provided by the ProMax software are;

$$Ca(OH)_2 + H_2SO_4 + 2H_2O \rightarrow CaSO_4.2H_2O\ (s) \downarrow + 2H_2O \quad \text{(R-5)}$$

$$Fe_2(SO_4)_3\ (s) + 3Ca(OH)_2\ (s) + 6H_2O \rightarrow 2Fe(OH)_3\ (s) \downarrow + 3CaSO_4.2H_2O\ (s)\downarrow \quad \text{(R-6)}$$

$$4ZnSO_4 + 3Ca(OH)_2 + 6H_2O \rightarrow ZnSO_4.3Zn(OH)_2.4H_2O\ (s)\ (B.Z.S) + 3CaSO_4.2H_2O\ (s) \quad \text{(R-7)}$$

$$4PbSO_4 + 3Ca(OH)_2 + 6H_2O \rightarrow PbSO_4.3Pb(OH)_2.4H_2O\ (s)\ (B.P.S) + 3CaSO_4.2H_2O\ (s) \quad \text{(R-8)}$$

$$4CuSO_4 + 3Ca(OH)_2 + 6H_2O \rightarrow CuSO_4.3Cu(OH)_2.4H_2O\ (s)\ (B.C.S) + 3CaSO_4.2H_2O\ (s) \quad \text{(R-9)}$$

Another important issue in the neutralization stage is the grade of the produced B.M.S which can be determined by the reaction equation and follow relation;

$$4MSO_4 + 3Ca(OH)_2 + 10H_2O \rightarrow MSO_4.3M(OH)_2.4H_2O + 3CaSO_4.2H_2O \quad \text{(R-10)}$$

$$\%B.M.S = \frac{4M^{2+}}{4M^{2+} \times 3M_{(OH)2} \times 6M_{H2O} \times M_{MSO4} \times 3M_{CaSO4}} \times 100 \quad \text{(Eq-22)}$$

Also, the efficiency of the neutralization stage can be calculated by the following equation:

$$R_N\,(\%) = \frac{V_N \times C_N - V_{Spent} \times C_{Spent}}{W_{B.M.S} \times p_{B.M.S} \times (1 - W_{H2O})_{B.M.S}} \times 100 \qquad \text{(Eq-23)}$$

where R_N, V_N, C_N, V_{spent}, C_{spent}, $m_{B.M.S}$, $p_{B.M.S}$, $(W_{H2O})_{B.M.S}$ are the efficiency of neutralization stage (%), the volume of the neutralization solution (m^3), the concentration of the neutralization solution (kg/m^3), the volume of the produced electrolysis spent solution (m^3), the concentration of the spent solution (kg/m^3), the weight of B.M.S (kg), the grade of B.M.S (%), and the moisture content of B.M.S (%) respectively. The needed number of reservoirs for increasing the reaction time in the acidic dissolution and neutral steps are calculated by the following relation:

$$N_{tank} = \frac{\rho_{slurry} \times V_{make\,up}}{V_{tank} \times \left(\frac{24}{t}\right)} = \frac{\rho_{slurry} \times W_{ore} \times R_{ore} \times (1 - W_{H2O}) \times p_{ore}}{V_{tank} \times \left(\frac{24}{t}\right) \times \Delta C} \qquad \text{(Eq-24)}$$

where N_{tank} is the number of required reservoirs, ρslurry is the density of the slurry, V_{tank} is the volume of the reservoir, $V_{make\text{-}up}$ is the volume of the extracted fresh solution (makeup), m_{ore} (kg) is the value of the used sulfide ores, p_{ore} (%) is the efficiency of the dissolution of sulfide ores, W_{H2O} (%) is the wet percentage of sulfide ores, p_{ore} is the grade of utilized sulfide ores (%), ΔC is the difference of the concentrations of the fresh solution and the spent (kg/m^3) and t is the reaction time in each acidic dissolution and neutral sections. In dissolution filters, the solution phase is extracted from the solid waste. Then, firstly extracted solution phase comprising Zn^{2+}, Cu^{2+}, and Pb^{2+} ions are reserved for the cobalt treatment stage. In this section, the important pollutions including Co^{2+}, Fe^{2+}, and Mn^{2+} are oxidized and removed in the form of iron trioxide, cobalt trioxide, and manganese ions by adding POM oxidizers and calcium hydroxide. Generally, the reactions of the cobalt purification are as follows;

$$nFeSO_4 + \text{POMs oxidizer/ acid catalyst} \rightarrow nFe_2(SO_4)_3 \qquad \text{(R-11)}$$
$$Fe_2(SO_4)_3 + 3Ca(OH)_2 + 6H_2O \rightarrow 2Fe(OH)_3 + 3CaSO_4.2H_2O \qquad \text{(R-12)}$$
$$nMnSO_4\,(aq) + \text{POMs oxidizer/ acid catalyst} \rightarrow nMnO_2\,(s) \qquad \text{(R-13)}$$
$$nCoSO_4\,(aq) + \text{POMs oxidizer/ acid catalyst} \rightarrow nCo_2(SO_4)_3\,(aq)$$
$$3Co_2(SO_4)_3\,(aq) + 9Ca(OH)_2\,(s) \rightarrow 6Co(OH)_3\,(s) + 9CaSO_4.2H_2O\,(s) \qquad \text{(R-14)}$$

Generally, the consumption amounts of additives such as POM oxidizers for the reaction with sulfide ores during the modeling process can be determined by the following relation:

$$W(kg) = C_i \times V_i \times \frac{n_{additive} \times Madditive}{ni \times Mi} \times P_{additive} \qquad \text{(Eq-25)}$$

where W (kg) is the value of the required additive, C_i (kg/m^3) is the concentration of the impurity in the solution, V_i (m^3) is the volume of the solution, n_i and $n_{additive}$ are stoichiometric coefficients of the impurity and consumables additive in the equilibrium

locations, M_i and $M_{additive}$ are the molecular weights of the impurity and consumables additive respectively and $P_{additive}$ (%) is the purity percentage of the additive. According to this relation, the amounts of needed additives including the amount of sulfuric acid in the acid dissolution step, the amount of $Ca(OH)_2$ in the negation section, the amounts of POM oxidizers, and zinc flour at the purification step can be calculated at each step of the process. In the next step, the solution phase extracted from the cobalt purification filter is reserved for the nickel treatment section and in this part, two main pollutions including Ni^{2+} and Cd^{2+} are removed by zinc flour. The reaction time, the amount of zinc powder and the grains surface of zinc flour are the three key parameters for eliminating Ni^{2+} and Cd^{2+} pollutions in the nickel purification section. The reactions of the nickel purification are as follows;

$$Zn^0\ (powder) + CdSO_4 \rightarrow Cd^0(s) + ZnSO_4\ (aq) \qquad \text{(R-15)}$$

$$Zn^0\ (powder) + NiSO_4 \rightarrow Ni^0\ (s) + ZnSO_4\ (aq) \qquad \text{(R-16)}$$

Accordingly, the values of the elimination of Ni^{2+} and Cd^{2+} pollutions at the nickel purification step by zinc flour can be calculated by:

$$\frac{\frac{dc}{dt}}{A\times W_{powder}} = k\times c \rightarrow \int_{c_0}^{c}\frac{dc}{c} = k.A.W_{powder}\int_{t0}^{t}d\tau \rightarrow Ln\frac{C}{C0} = A.W_{powder}.\Delta\tau.k_0e^{\frac{-Ea}{RT}} \rightarrow \frac{C}{C_0} = e^{k.e^{\frac{-Ea}{RT}}.\ A.\ W_{powder}.\ \Delta\tau} \qquad \text{(Eq-26)}$$

where C_0 and C are the initial and urgent concentrations of the Ni^{2+} and Cd^{2+} pollution (kg/m^3) respectively, k is the equation constant, A is the grains surface of zinc flour, m_{flour} is the amount of zinc flour consumed and Δt is the retention time in the nickel purification step. In the nickel treatment, nickel and cadmium impurities react with zinc powder by various mechanisms. Nickel impurities are removed by the cementation mechanism. In this method, nickel impurities are adhesive to the surface of the zinc powder and are reduced on its surface. Cadmium impurities are reduced by electrochemical or electron exchange methods with zinc powder.

In the final step, the filtered solution phase in the nickel purification section is reserved as a fresh solution (makeup) to make up storage tanks. The value of the extracted fresh solution and the number of hot refining can be determined using the following relations:

$$V_{make\ up}\ (m^3) = \frac{W_{ore}\times\gamma_{ore}\times(1-W_{H2O})\times p_{ore}}{\Delta C} = \frac{n\times V\times C}{\Delta C} \qquad \text{(Eq-27)}$$

where V (m^3) is the volume of the fresh solution, m (kg) is the value of the used sulfide ores, R_{ore} (%) is the efficiency of the dissolution of sulfide ores , W_{H2O} (%) is the wet percentage of sulfide ores, P_{ore} is the grade of sulfide ores (%), ΔC is difference between the concentrations of the fresh solution and spent (kg/m^3). After reserving the fresh solution (makeup) to the electrolysis section, its metal content can be separated by the electric energy consumption (electric current). Electrolysis reactions can be written as follows:

$$\text{Anodic reaction: } H_2O \rightarrow 1/2O_2 + 2H^+ + 2e^- \qquad \text{(R-17)}$$

Cathodic reaction: M^{2+} (Zn^{2+}, Cu^{2+} and Pb^{2+}) $+2e^- \rightarrow M(s)$ (R-18)

The weight of the extracted metals and energy consumption to extracted metals in direct current mode (Eq-21) and in alternating current mode (Eq-22) can be determined by the following equations:

$$m\ (kg) = \frac{M_{metals} \times I_{DC} \times t \times R_I \times N_{cathod} \times N_{cell}}{n_e F} \quad \text{(Eq-28)}$$

$$W_{DC}\ (kw.h/kg) = \frac{I_{DC} \times V_{DC} \times t \times N_{cathod} \times N_{cell}}{m} \quad \text{(Eq-29)}$$

$$W_{AC}\ (kw.h/kg) = \frac{1.73 \times I_{AC} \times V_{AC} \times t \times cos\varphi}{m} \quad \text{(Eq-30)}$$

The maximum amount of the electrical current energy of an industrial unit is consumed in its electrolysis section. Therefore, high electrical current efficiency is very effective in reducing the cost and amount of energy consumed in industrial plants. Electrolysis current efficiency can be calculated by the following equation:

$$R_I\ (\%) = \frac{Vmake\ up \times \Delta C \times n_e F}{I_{DC} \times \tau \times N_{cathod} \times N_{cell} \times M_{metals}} \times 100 \quad \text{(Eq-31)}$$

where m, I_{DC}, t, R_I, N_{cathod}, N_{cell}, V_{DC}, n_e, M_{metals}, F and W have the extracted metals, direct electric current, retention time (sec), current efficiency (%), number of cathodes in each cell, number of electrolysis cells, voltage of electrolysis (v), number of exchanged electrons, molecular weight (g/mol) of metals, faraday constant (c/mol) and electric energy consumption (kw.h/kg) respectively. Another important factor in the electrolysis section is the amount of solution circulated in the electrolysis cells. The circulation rate of the spent solution in the electrolysis section has an important effect on reducing the concentration polarization and reducing the negative effects of impurities. To find out the required volume (how many liters) of the electrolysis spent solution to achieve a certain amount of the production must be rotated per electrolysis cell per minute can use the following equation:

$$\upsilon\ (liter/min) = \frac{m}{\Delta C_{spent} \times t \times n_{cathod} \times n_{cell}} \quad \text{(Eq-32)}$$

where ν is the volumetric flow of the electrolysis solution (liter/min), m is produced metals (kg), t is the passage time of the electrolysis spent solution (min) and ΔC_{spent} is the difference between the concentrations of the input spent solution and the output spent solution (kg/m^3).

The relationship between increasing the concentration of the electrolysis solution from the initial concentration(C_0) to the final concentration (C), the relation of the efficiency changes by the concentration changes of the electrolysis solution, and also the relationship between the amount of the metal adsorption in electrolysis can be expressed as:

$$(C - C_0)_{spent} \times V_{spent} = C_{makeup} \times V_{makeup} \quad \text{(Eq-33)}$$

$$\frac{E}{E_0} = \frac{C}{C_0} = \frac{\left(\frac{ItM}{nFV}\right)}{\left(\frac{ItM}{nFV}\right)_0} = \frac{I}{I_0} = \frac{\frac{m}{t}}{\frac{m_0}{t_0}} \quad \text{(Eq-34)}$$

$$r_A = \frac{C_{theory} - C_{experimental}}{t} = \frac{\frac{\text{C makeup} \times \text{V makeup} - C0 \times \text{V spent}}{\text{V spent}} - C_{experimental}}{t} \quad \text{(Eq-35)}$$

Where C, C_0, V_{spent}, C_{makeup}, V_{makeup}, E, E_0, $r_{absorption}$, and t are the final concentration of the spent, initial concentration of the spent, concentration of the fresh-solution, volume of the fresh-solution, efficiency of the electrolysis in the final concentration, electrolysis efficiency in the initial concentration, absorption rate in the electrolysis and absorption time respectively. The needed kinetic and thermodynamic data of reactions for modeling the process are summarized in Table 7. Also, the obtained modeling results are reported in Table 8.

Table 7
Required thermodynamic and kinetic data for the simulation of designed processes using the ProMax software

Reactions	T (°C)	pH	k (min^{-1})	k_0 (min^{-1})	Ea (kcal/mol)	ΔG° (kcal/mol)	ΔH° (kcal/mol)	ΔS° (kcal/°C. mol)
(1)	90	1.5	0.142	0.139	256.782	-104.899	-128.135	26.634
(2)	90	1.5	0.131	0.128	334.096	-98.273	-156.631	28.527
(3)	90	1.5	0.157	0.143	523.208	-93.842	-143.643	29.381
(4)	90	3.5-4.5	0.476	0.412	12.080	-24.533	-48.757	12.208
(5)	90	4.0-4.5	0.789	0.797	9.051	-535.651	-56.234	13.523
(6)	90	4.0-4.5	0.961	0.944	7.251	-675.951	-63.356	16.853
(7)	90	4.0-4.5	0.975	0.899	5.753	-589.534	-75.952	15.128
(8)	75	4.5-5.2	0.075	0.071	13.136	-48.780	-97.240	5.068
(9)	75	4.5-5.2	0.666	0.713	8.359	-78.638	-57.053	0.807
(10)	75	4.5-5.2	0.045	0.043	10.341	-63.417	-74.652	3.416

Reactions	(1)	- ZnS (s) + POMs + $H_2SO_4 \rightarrow ZnSO_4$ (aq)
	(2)	- CuS (s) + POMs + $H_2SO_4 \rightarrow CuSO_4$ (aq)
	(3)	- PbS (s) + POMs + $H_2SO_4 \rightarrow PbSO_4$ (aq)
	(4)	- $Ca(OH)_2 + H_2SO_4 \rightarrow CaSO_4 + H_2O$
	(5)	- Fe^{2+} + POMs + $Ca(OH)_2 \rightarrow Fe_3O_4$ (s) ↓
	(6)	- Mn^{2+} + POMs + $Ca(OH)_2 \rightarrow MnO_2$ (s) ↓
	(7)	- Co^{2+} + POMs + $Ca(OH)_2 \rightarrow Co_3O_4$ (s) ↓
	(8)	- Zn (s) + Cd^{2+} (aq) → Cd (s) + Zn^{2+} (aq)
	(9)	- Zn (s) + Ni^{2+} (aq) → Ni (s) + Zn^{2+}(aq)
	(10)	- Zn (s) + Pb^{2+}(aq) → Pb (s) + Zn^{2+} (aq)

Table 8
Final results predicted by the ProMax software

Results summary of streams materials																
Parameters	**unit**	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**POMs**	**Leach cake**	**Sulfide ore**	**Sulfuric acid**	**Cobalt cake**	**Nickel cake**	**Metals extraction**
Temperature	°C	25	90	90	90	90	90	90	90	90	70	25	25	85	75	50
Pressure	bar	1.0	1.0	1.0	170	1.0	170	1.0	170	-	-	-	-	-	-	-
Vapor Frac	-	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Mole Flow	kmol/hr	83.23	83.23	83.23	89.98	89.98	89.98	89.98	87.28	2.69946	6.748	0.754	18.734	33.746	33.746	87.283
Mass Flow	kg/hr	3345.7	3345.7	3345.8	3845.7	3845.7	3845.7	3845.7	3730.3	115.4	499.9	59.977	1837.421	907.860	907.864	3730.330
Volume Flow	cum/hr	83.23	83.23	2344	89.98	89.98	89.98	89.98	87.28	2.69	6.748	0.754	18.734	33.746	33.746	87.283
Enthalpy	MMkcal/hr	-8.359	-8.276	-7.37	-9.210	-9.141	-9.210	-9.141	-8.867	-0.274	-1.031	-0.005	-3.550	-2.757	-2.757	-8.934
ZnS	kmol/hr	0.754	0.754	8.4×10^{-7}	8.4×10^{-7}	8.4×10^{-7}	8.4×10^{-7}	8.4×10^{-7}	8.1×10^{-7}	0	0	0.754	0	0	0	0
PbS	kmol/hr	0.043	0.043	7.5×10^{-7}	7.5×10^{-7}	7.5×10^{-7}	7.5×10^{-7}	7.5×10^{-7}	7.3×10^{-7}	0	0	0.043	0	0	0	0
$CuFeS_2$	kmol/hr	0.003	0.003	2.8×10^{-7}	2.8×10^{-7}	2.8×10^{-7}	2.8×10^{-7}	2.8×10^{-7}	1.3×10^{-7}	0	0	0.003	0	0	0	0
H_2SO_4	kmol/hr	22.48	22.48	5.503	10.56	10.56	10.56	10.56	10.56	0	1.258	0	18.734	3.746	3.746	21.074
$Ca(OH)_2$	kmol /hr	0	0	0	6.748	6.748	6.748	6.748	6.546	0	0.875	0	0	0	0	6.546
$CaSO_4$	kmol/hr	0	0	0	7.4×10^{-5}	2.8×10^{-8}	2.8×10^{-8}	2.8×10^{-8}	2.7×10^{-8}	0	3.7×10^{-4}	0	0	0	0	2.67×10^{-8}
$ZnSO_4$	kmol/hr	0	0	0.754	0.754	0.754	0.754	0.716	0.716	0	2.262	0	0	0.003021	0.00247	0.7160
$PbSO_4$	kmol/hr	0	0	0.043	0.043	0.043	0.043	0.039	0.039	0	0.00129	0	0	0.001923	2.6×10^{-3}	0.0360
$CuSO_4$	kmol/hr	0	0	0.024	0.024	0.024	0.024	0.022	0.022	0	0.00072	0	0	0.001741	4.3×10^{-3}	0.0159
Fe^{2+}	kmol/hr	0	0	0.243851	0.243851	0.243851	0.243851	0.0231	0.0231	0	0.241362	0	0	0.005447	6.1×10^{-4}	1.2×10^{-6}
Co^{2+}	kmol/hr	0	0	0.00023	0.00023	0.000012	0.0000118	0.0000003	2.42×10^{-8}	0	0.00011	0.00023	0	0.0000118	5.63×10^{-8}	2.7×10^{-8}
Ni^{2+}	kmol/hr	0	0	0.000081	0.000081	0.000081	0.000081	0.000001	0.000001	0	0.000080	0	0	0	0.000080	1.1×10^{-6}
Cd^{2+}	kmol/hr	0	0	0.000056	0.000056	0.000056	0.000056	4×10^{-7}	4×10^{-7}	0	0.000055	0	0	0	0.000055	3.0×10^{-8}
Mn^{2+}	kmol/hr	0	0	0.010233	0.014233	0.014233	0.014233	0.000843	0.000843	0	0.01013	0	0.009431	5.4×10^{-4}	4.0×10^{-5}	0.002692
Sulfur	kmol/hr	0	0	0.000002	0.000002	0	0	0	0	0	0	0.8	0	0	0	0
As^{3+}	kmol/hr	0	0	0.000052	0.000052	6.8×10^{-8}	5.193×10^{-5}	5.193×10^{-5}	5.193×10^{-5}	0	0.00005199	0.000052	0	5.193×10^{-5}	5.193×10^{-5}	5.193×10^{-5}
POMs	kmol/hr	0	0	0.00542	0.0000145	0.0000025	0.0000623	0.0000002	2.0×10^{-8}	0.00542	7.0×10^{-5}	0	0	3.0×10^{-4}	$1.5.0\times10^{-8}$	1.0×10^{-8}
Zinc powder	kmol/hr	0	0	0	0	0	0	0	0.8333	0	0	0	0	0	0.0798	0

3. Results and Discussion

3.1. ProMax Simulation

By the simulation of the operation of the metals extraction with the ProMax software, some key factors including; the temperature and pressure changes, mole flow rates changes of streams during the modeling process, investigation of kinetic and thermodynamic changes of reactions, mole flow changes of materials in streams and blocks and metals extraction efficiency are investigated. The predicted values of the Zn, Cu, and Pb metals extraction from sulfide ores and the removal of toxic heavy metals from the leaching solution were calculated at about 85%, 81%, 83%, and 99.9% receptivity. The attained optimum conditions from the Taguchi procedure and the simulation results by the ProMax software can be utilized to popularize the experimental gauge to the industrial gauge.

3.2 Impact of Operating Factors

3.2.1 Impact of the Concentration of Acid on the Dissolution Rate of sulfide ores by the POMs Oxidizer

The impact of the concentration of acid on the process of the sulfide ores dissolution (metals extraction) in the acid solution by POM oxidizers and also the removal of toxic heavy metals was investigated in different concentrations of 60, 70, 80, and 90g/l of the acid. The achieved results are given in Fig 4. As indicated in this figure, it is obvious that a concentration of 80 g/l of the acid exhibits the utmost impact (greatest S/N ratio) on the extraction of the metal, whereas the maximum removal of toxic heavy metals occurs at a concentration of equal to 60 g/l of the acid. It seems that raising the concentration of the acid to more than 80 g/l only increases the free acidity of the leach liquor and the cost of the consumable acid. Under the optimized concentrations of 80 and 60 of the acid, the highest extraction efficiency of Zn, Cu, and Pb metals was achieved and the maximum removal of toxic heavy metals with the dissolution of sulfide ores by POM oxidizers were determined about 82.47% and 94.8% respectively.

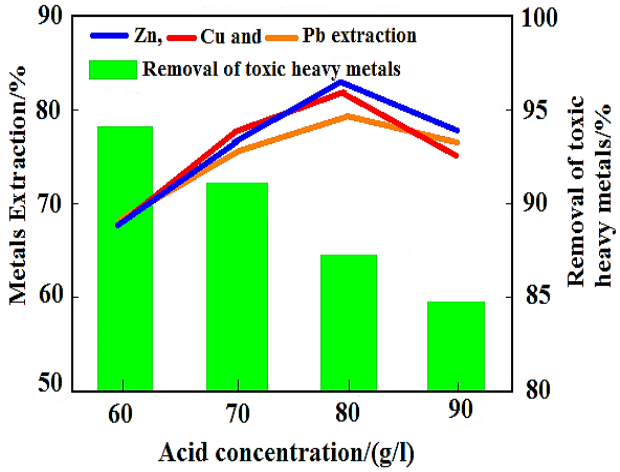


Figure 4. Impact of the concentration acid on the extraction of metals and ther removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the reaction temperature of 90 ºC, rotation speed of 300 rpm, retention time of 1.0h, Polyoxometalate value of 0.5 g/l, acid type of H_2SO_4, grain size of 1 mm and Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

3.2.2 Impact of the Reaction Temperature on the Dissolution Rate of Sulfide ores by POM Oxidizers

The operation temperature is a main and key factor in the dissolution rate of sulfide ores. The impact of the different temperatures of 60, 70, 80, and 90 °C on the extraction of the metal and the removal of toxic heavy metals with the dissolution of sulfide ores by POM oxidizers was evaluated in Fig. 5. As it can be observed in Fig. 5, the leaching of sulfide ore in the acid solution by POM oxidizers is highly temperature dependent. Among variant temperatures, a temperature of 90 °C exhibited the most impressive effect (highest S/N ratio) on the extraction of metals. Also, a temperature of 60 °C illustrated the highest impact (highest S/N ratio) on the removal of

toxic heavy metals. Under an optimized temperature of 90 °C, the greatest extraction efficiency of Zn, Cu, and Pb metals and the maximum removal of toxic heavy metals were obtained at about 73.4%, 71%, 72.5%, and 91.36% respectively.

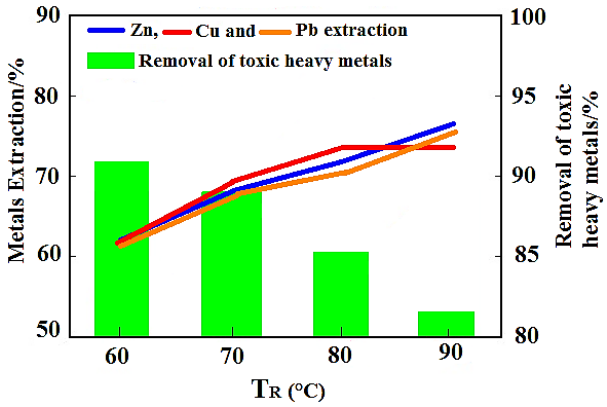


Figure 5. Influence of the operation temperature on the extraction of metals and the removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80 g/l of acids, the rotation speed of 300 rpm, the retention time of 1.0h, the value of 0.5 g/l of POMs, the acid type of H_2SO_4, the grain size of 1 mm and the POMs type of $[H_2W_{12}O_{40}]^{10-}$.

3.2.3 Impact of the Rotation Speed on the Dissolution Rate of Sulfide ores by POM Oxidizers

The dissolution rate of sulfide ores in present of the acid catalyst solution by POM oxidizers is evaluated as a function of rotation rates. The curves of the extraction efficiency of metals and the removal of heavy metals at the variant rotation rates of from 50 to 300 rpm are presented in Fig. 6. Generally, a high rotation rate of the leaching solution has increased the solubility of sulfide ores in the acid solution by explaining the mechanisms of decreasing the film thickness of the diffusion layer. The presented results in Fig. 6 exhibited that the rotation speed of 300 rpm was the most effective (highest S/N ratio) in the dissolution of sulfide ores in the acid solution by POM oxidizers. Under the optimal conditions of 300 rpm, the highest extraction efficiencies of Zn, Cu, and Pb metals with the dissolution of sulfide ores by POM oxidizers were presented at about 73%, 70.23%, and 72.1%. Also, the maximum removal of heavy metals occurred at a rotation speed of 100 rpm, at which the removal amount was attained at about 91.7%.

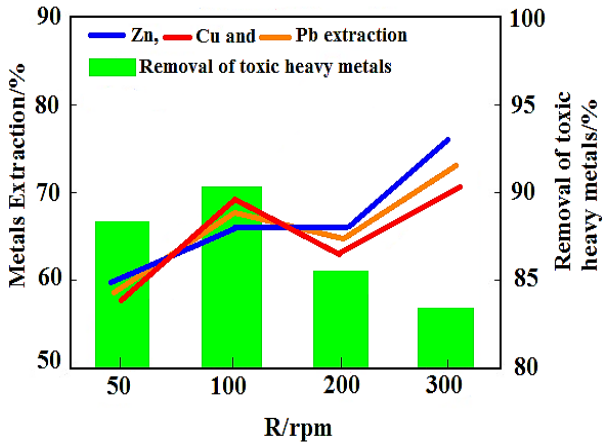


Figure 6. Effect of the rotation rate on the extraction of metals and the removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80 g/l of acids, the reaction temperature of 90 ºC, the retention time of 1.0h, the value of 0.5 g/l of Polyoxometalate, the acid type of H_2SO_4, the grain size of 1 mm and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

3.2.4 Impact of the Retention Time on the Dissolution Rate of Sulfide ores by POM Oxidizers

The impact of the retention time on the dissolution rate of sulfide ores by POM oxidizers in the present of acid catalysts was investigated at a concentration of 60-90g/l of the acid, the reaction temperate of 60- 90°C, the rotation rate of 50-300 rpm, the amount of 1-5g/liter of POMs, the acid type of H_2SO_4, HNO_3, HCl and H_3PO_4, the grain size of 0.5-3.0 mm, and the POMs type of; $[Mo_6O_{19}]^{2-}$, $[Mo_8O_{26}]^{4-}$, $[V_{10}O_{28}]^{6-}$, and $[H_2W_{12}O_{40}]^{10-}$. Fig. 7 illustrates that by raising the reaction

time to 1hr the extraction efficiency of metals increases. Therefore, a reaction time of 1.5h was selected as the optimum reaction time for the removal of toxic heavy metals in the leaching process. As a result, the highest extraction efficiencies of Zn, Cu, and Pb metals and the highest removal of toxic heavy metals with the dissolution of sulfide ores under the optimal condition were attained as about 74%, 78%, 79.8% and 95.3% respectively.

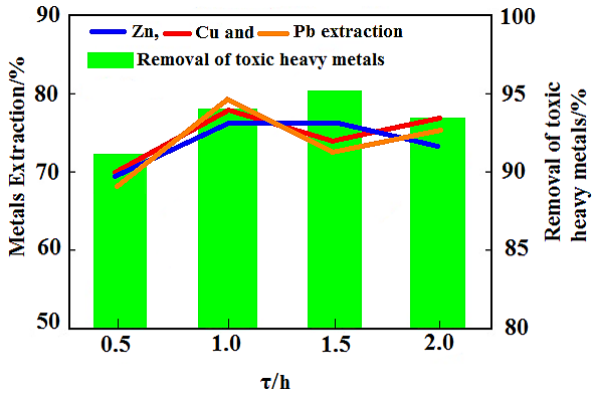


Figure 7. Impact of the reaction time on the extraction of metals and the removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80 g/l of acids, the reaction temperature of 90 ºC, the rotation speed of 300 rpm, the value of 0.5 g/l of Polyoxometalate, the acid type of H_2SO_4, the grain size of 1 mm and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

3.2.5 Impact of the Value of Polyoxometalate on the Dissolution Rate of Sulfide ores by POM Oxidizers

The amount of POM oxidizers is a main factor in increasing the dissolution rate of sulfide ores, extraction of metals, and removal of toxic heavy metals. The impact of the value of POM oxidizers on the extraction metals and the removal of toxic heavy metals is illustrated in Fig. 8. According to Fig. 8, it is obvious that 0.5 g/l of POM oxidizers had the most effective extraction of metals and removal of toxic heavy metals. Under this condition, the maximum extraction efficiency of Zn, Cu, and Pb metals and removal of toxic heavy metals from sulfide ores were calculated as about 81%, 79.85%, 82.5%, and 97% respectively.

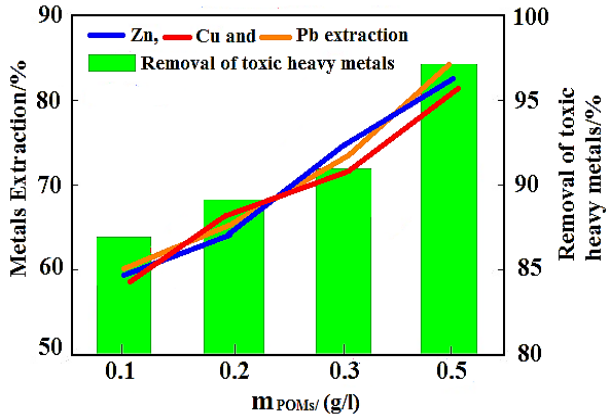


Figure 8. Effective values of Polyoxometalate oxidizers on the extraction of metals and the removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80g/l of acids, the reaction temperature of 90 ºC, the rotation speed of 300 rpm, the retention time of 1.0h, the acid type of H_2SO_4, the grain size of 1 mm and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

3.2.6 Impact of the Acid Type on the Dissolution Rate of Sulfide ores by POM Oxidizers

The impact of the acid type on the extraction of metals from sulfide ores and also the removal of toxic heavy metals by POM oxidizers was studied in the different acid types of H_2SO_4, HNO_3, HCl, and H_3PO_4. As a result, according to the curves of Fig. 9, the acid type of H_2SO_4 was indicated the most impressive on the extraction Zn, Cu, and Pb metals with the values of 82%, 81.5%, and 81.8%. Also, the maximum removal of toxic heavy metals from sulfide ores in the presence of an HNO_3 acid catalyst was determined at about 94%.

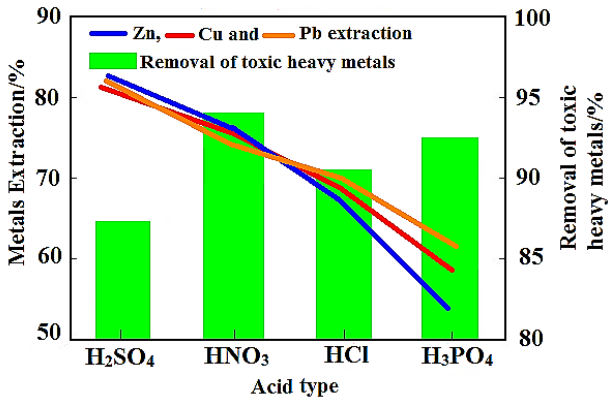


Figure 9. Influence of the acid type on the extraction of metals and the removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80g/l of acids, the reaction temperature of 90 ºC, the rotation speed of 300 rpm, the retention time of 1.0h, the Polyoxometalate value of 0.5 g/l, the grain size of 1 mm and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

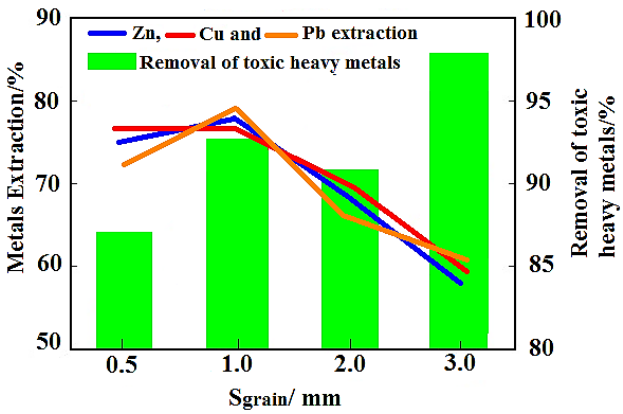


Figure 10. Impact of the grain size of sulfide ores on the extraction metals and removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimized conditions of; the concentration of 80 g/l of acids, the reaction temperature of 90 ºC, the rotation rate of 300 rpm, the retention time of 1.0h, the value of 0.5 g/l of Polyoxometalate, the acid type of H_2SO_4 and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$.

3.2. 7 Impact of the Grain Size on the Dissolution Rate of Sulfide ores by POM Oxidizers

The effect of the grain size of sulfide ores on the dissolution rate of sulfide ores and the extraction of metals is indicated in Fig. 7. The grain size of sulfide ore minerals is one of the main factors in increasing the rate of the dissolution of ores and reactions. The grains size of sulfide soils should be optimized due to two important factors: 1) by over-reducing the the grain size of sulfide ores, although the dissolution rate of sulfide ores increases, the dissolution and release rates of sulfide ore impurities (toxic heavy metals) increase simultaneously. 2) the cost of crushing the ore increases. As shown in Fig. 10, the highest extraction of metals was attained at a grain size of 1.0 mm. Also, the maximum extraction efficiency of Zn, Cu, and Pb metals and removal of toxic heavy metals from sulfide ores at the grain sizes of 1.0 mm and 3.0 mm were obtained at about 78.5%, 77.35%, 80%, and 98% respectively.

3.2.8 Impact of the POM Type on the Dissolution Rate of Sulfide ores by POM Oxidizers

The impact of the POM type on the oxidizing rate of the sulfide compounds of sulfide ores and the removal of toxic heavy metals in the leaching process was evaluated in Fig. 11. According to the extraction curve of metals in Fig. 11, the POM type of $[H_2W_{12}O_{40}]^{10-}$ was the most effective in the extraction of zinc, copper, and lead from sulfide ores. The maximum extraction efficiencies of Zn, Cu, and Pb metals from sulfide ores and the removal of toxic heavy metals at the present of the $[H_2W_{12}O_{40}]^{10-}$ oxidizer were achieved as about 85%, 83%, 81%, and 97.5% respectively.

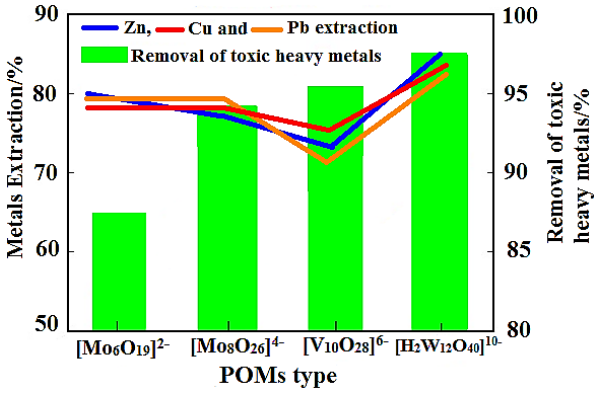


Figure 11. Impact of the Polyoxometalate type on the extraction of metals and removal of toxic heavy metals from sulfide ores by Polyoxometalate oxidizers under the optimum conditions of; the concentration of 80 g/l of acids, the reaction temperature of 90 ºC, the rotation speed of 300 rpm, the retention time of 1.0h, the Polyoxometalate value of 0.5g/l, the acid type of H_2SO_4 and the grain size of 1 mm.

4. Conclusion

In the current work, the metal extraction operation on the sulfide ores by Polyoxometalates compounds was optimized and modeled by Taguchi and ProMax methods. In this proposed plan, POM oxidizer agents were utilized as contributing factors in the dissolution of sulfide ores, extraction of Zn, Cu, and Pb metals and removal of toxic heavy metals. This new method is novel and has advantages over other methods, which include: 1) no production of toxic and harmful wastes, 2) no hazards of working with high temperatures and pressurized tanks, and 3) a low-cost and low-risk method for the environment. The achieved optimum results by the Taguchi method were applied as the input data for the simulation of the process by the ProMax software. The ProMax software is utilized to simulate the designed process and calculate the extraction efficiency of Zn, Cu, and Pb metals from sulfide ores and also the removal of toxic heavy metals by POM oxidizers. Optimized conditions for the extraction of metals from sulfide ores by POM oxidizers are obtained as follows: the concentration of 80g/l of acids, the reaction temperature of 90 ºC, the rotation rate of 100 rpm, the retention time of 1.0 h, the value of 0.5 g/l of Polyoxometalate, the acid type of H_2SO_4, the grain size of 1.0 mm and the Polyoxometalate type of $[H_2W_{12}O_{40}]^{10-}$. In optimum situations, the maximum extractions of metals and removal of toxic heavy metals were calculated as about 85%, 81%, 83%, and 99.9% for the extraction of Zn, Cu, and Pb metals and removal of toxic heavy metals respectively. Finally, the results of the process simulation by the ProMax software can be applied on industrial scales

Nomenclatures

S	Special surface of zinc powder (m^2)
n	Weight ratio of the produced waste to the used ore
C_S	Solution concentration (g/l)
C_{Spent}	Spent electrolysis solution concentration (g/l)
C_0	Initial solution concentration (g/l)
C_1	Final solution concentration (g/l)
R_I	Current efficiency (%)
E_a	Activation energy of reactions (kcal/mol)
F	Faraday constant (c/mol)
I_{DC}	Electric current (Am)
M_{metals}	Molecular weight of metals (g/mol)
m_{ore}	Weight of sulfide ores (ton)
m_{zn}	Weight of used zinc powder (kg)
n_e	Number of exchanges electrons
n_i	Stoichiometry coefficient of impurities in chemical reaction
n_j	Stoichiometry coefficient of additive in chemical reaction
$N_{cathode}$	Number of cell cathodes
N_{cell}	Number of electrolysis cells
P_{ore}	Grade of sulfide ores (%)
P_{waste}	Grade of metals in waste (%)
P	pressure (bar)
f_{ore}	Molar flow rate of sulfide ores (kmol/h)
f_{waste}	Molar flow rate of waste (kmol/h)
R	Universal gas constant (Kcal/°C. mol)
γ_{ore}	Efficiency of sulfide ores (%)
γ_A	Acid dissolution efficiency (%)

γ_N	Negation stage efficiency (%)
$\gamma_{metals\ extraction}$	Extraction efficiency (%)
R	Rotation rate (rpm)
T_R	Temperature (°C)
t_A	Acid retention time (h)
t_N	Negation retention time (h)
τ	Retention time (min)
V_{DC}	Voltage (v)
V_A	Volume of acid leaching solution (m^3)
V_N	Volume of negation solution (m^3)
C_A	Concentration of acid leaching solution (g/lit)
C_N	Concentration of negation solution (g/lit)
V_{tank}	Volume of tank (m^3)
N	Number of tanks
m_{POMs}	Value of consumed POMs (kg/ton)
$(\zeta_{H2O})_{ore}$	Moisture of sulfide ores (%)
$(\zeta_{H2O})_{waste}$	Moisture of waste (%)
$m_{sulfide\ ores}$	weight of sulfide ores (kg or ton)
$W_{metals\ extracted}$	Weight of metals extracted (kg or ton)

Symbols

n	Total number of replications of each test
S_{size}	Grain size (mm)
S/N	Signal to Noise
SNL	Signal to Noise Larger
Y_i	Response in the repeated experiment of 'i'

Greek letters

$\Delta G°$	Gibbs free energy of reactions/(kcal/mol)
$\Delta H°$	Enthalpy of reactions/(kcal/mol)
$\Delta S°$	Entropy of reactions/(kcal/°C. mol)

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