Establishing the Optimal Parameters for the Aqueous Oxidation of Sulphurous Concentrates with Lead Content

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The paper presents the results of the mathematical modelling for the oxidation process (in autoclave) of sulphurous concentrates with lead (PbS containing), in weak alkaline aqueous medium. Using an active program of experiments (an order two orthogonal program, PO2), the optimal conditions were established in order to run the process in a manner that will lead to obtaining a maximum oxidation yield of sulphides from lead concentrates. Preliminary research and studies indicated four parameters (temperature, process duration, oxygen partial pressure and L:S ratio) that have a significant influence on the performance of the oxidation process, which we define as the oxidation yield of the sulphides in concentrate. The optimal values of the oxidation process parameters are: T = 138°C, t = 2.53h, p_{O2} = 4.78 atm and L:S ratio = 15:1.

Keywords: mathematical modelling, oxidation process, parameter optimization, sulphurous concentrates

Metallic lead is currently obtained from sulphurous concentrates, using exclusively pyrometallurgical processes – sintering, smelting (classical processes like Watter Jacket and ISP) and direct melting (as in the recently developed processes QSL, Isamelt and Ausmelt) – which generates gases with SO_2 and volatile dust, thus presenting a high pollution risk for the environment [1, 2].

Recently, most of the research work has been focused on the elaboration of new technological concepts of lead extraction from sulphurous concentrates through hydrometallurgical processes, instead of pyrometallurgical processes, because the technological operations are friendlier to the environment and preferable from an economic point of view. The minimization of emissions is guided by the slogan proposed by the international community: "zero emissions, zero impact production".

Different methods of oxidation and leaching of lead from concentrates have been studied worldwide in different environments with ferric sulphate [3], hexafluosilicic acid [4], chlorides [5], nitric acid [6], oxidation in presence of bacteria [7], autoclave processing under high pressure and temperature, in the presence of oxygen or at atmospheric pressure [8].

In this context, a new technological concept was developed for the sulphurous concentrates oxidation in a weak alkaline medium (sodium carbonate solutions) under air in autoclave, followed by lead leaching in alkaline solutions (sodium hydroxide solutions) and subsequently metallic lead electro-winning. (fig. 1) [9]

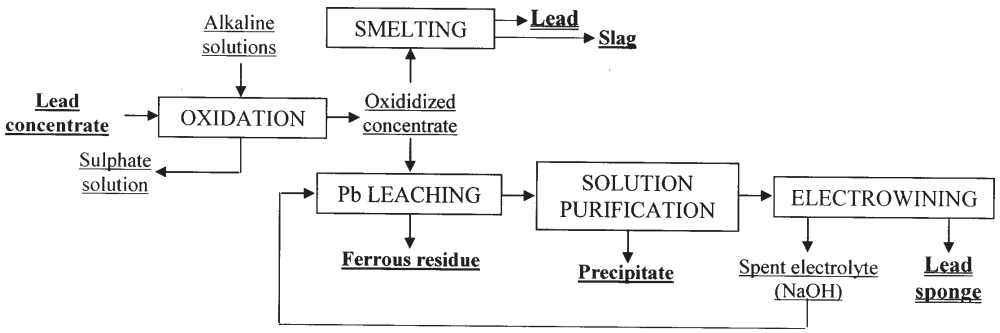

Fig. 1. Technological flow – lead obtaining from sulphurous concentrates

This paper studies the mathematical modelling for the oxidation process of the lead concentrate in aqueous medium, in order to establish the optimal process parameters. The modelling was performed using an active research program: an order two orthogonal program, PO2 [10, 11].

Experimental part

Mineralogical and chemical composition of lead concentrate

Lead concentrate from Flotatia Centrala Baia Mare was used for experiments regarding the oxidation of metallic sulphides in aqueous medium, with the chemical composition: 55.16 % Pb; 2.30 % Cu; 1.95 % Zn; 9.80 % Fe; 0.32 % Sb; 0.052 % As; 0.50 % Si; 0.052 % Ca; 21.4 % S.

The mineralogical and microscopic study was carried out through X-ray diffraction (XRD) and microscopy showed the presence of lead as galena – PbS, copper as chalcopyrite – $CuFeS_2$, zinc as sphalerite – ZnS and iron as pyrite – FeS_2.

Oxidation experiments in aqueous medium – sodium carbonate solutions

The oxidation experiments of metallic sulphides from lead concentrate in aqueous medium (sodium carbonate solutions) were carried out in an autoclave with an impeller stirring system (750 rpm), at temperatures between 110 – 140 °C, processing time from 1/2 h up to 3 h, oxygen partial pressures of 3.5 – 6.5 atm and L/S ratio between 10:1 – 16:1. Air was used as an oxidation agent. While running

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Exp.	Input materials			Operating parameters				Output materials			Sulfur removal efficiency (%)
	Water (ml)	Na_2CO_3 (g)	Lead concentrates (g)	T (^{0}C)	t (h)	P_{O2} (atm.)	L/S	Oxidized concentrates (g)	S_{tot} (%)	Alkaline Solution (ml)	
1	3000	268	300	115	1	4	10	315.2	2.48	2750	87.16
2	3000	178	200	135	1	4	15	207.8	2.26	2425	88.43
3	3000	178	200	115	3	4	15	202.7	0.98	2740	95.11
4	3000	268	300	135	3	4	10	287.6	1.01	2650	95.23
5	3000	178	200	115	1	6	15	205.8	4.36	2710	77.90
6	3000	268	300	135	1	6	10	301.6	4.11	2550	79.65
7	3000	268	300	115	3	6	10	302.6	1.22	2650	95.24
8	3000	178	200	135	3	6	15	200.7	0.35	2810	95.07
9	3000	214	240	110	2	5	12.5	246.6	1.44	2570	92.71
10	3000	214	240	140	2	5	12.5	230.0	1.14	2725	94.62
11	3000	214	240	125	0.5	5	12.5	250.2	6.06	2780	70.88
12	3000	214	240	125	3.5	5	12.5	225.9	0.76	2690	96.48
13	3000	214	240	125	2	3,5	12.5	237.6	1.52	2820	92.59
14	3000	214	240	125	2	6,5	12.5	248.2	1.58	2850	91.95
15	3000	303	340	125	2	5	8.8	347.7	2.07	2852	89.57
16	3000	165	185	125	2	5	16.2	186.3	1.16	2740	94.26
17	3000	214	240	125	2	5	12.5	248.3	1.26	2790	93.58
18	3000	214	240	125	2	5	12.5	238.7	1.78	2920	91.28
19	3000	214	240	125	2	5	12.5	239.9	1.64	2700	91.92

Table 1
EXPERIMENTAL RESULTS

the experiments the following parameters were kept constant: stirring speed, air flow, sodium carbonate quantity of 30% above the theoretical level for sulphur oxidation.

The lead concentrate was grinded in an attritor ball mill, using a ball diameter of 3 mm, in wet environment, with the balls/concentrate ratio of 14:1 and the water/concentrate ratio of 2:1. The duration of the grinding was 30 min.

After grinding, the grain size of the lead concentrate was 100% below 5 μm, the specific surface was increased from 0.81 m^2/g to approximately 7.43 m^2/g and a defective structure of solid phase was made. This had a positive influence on the material reactivity in the oxidation process.

During the experiments, a large number of samples were collected to be chemically analyzed through: Inductive Coupled Plasma Spectrometer (ICP), Direct Coupled Plasma (DCP) and (Flame Atomic Absorption Spectrometer (FAAS).

Working conditions and experimental results obtained are presented in table 1.

Results and discussions

The research studies indicated four parameters that have a significant influence on the performance of the oxidation process, which we define as the oxidation yield of the sulphur in concentrate – y. The four parameters are: temperature (^{o}C) - z_1, process duration (h) - z_2, oxygen partial pressure (atm) - z_3 and L:S ratio (dimensionless) - z_4. There are two other parameters that intervene in the process: air flow (135 L/h) and the reactant excess (Na_2CO_3 – 30 % above the theoretical level needed for sulphur oxidation), but they do not have a significant influence and were kept constant.

An active research program – an order two orthogonal program – was used in order to determine the optimal conditions for conducting the oxidation process. The program was created by supplementing a first order program of type EFC 2^n (EFC = complete factorial experiment) with certain points in the factorial space [10, -12]. The result is given by eq. (1):

$$N = N_c + N_\alpha + N_0 \quad (1)$$

where: N – represents the total number of points in the PO2 program; N_c – the number of the experiments in a program EFC 2^n ($N_c = 2^n$) or a subprogram EFF ($N_c = 2^{n-1}$), the subprogram used in this experiment with the generative relation $x_4 = x_1x_2x_3$; N_α – the number of so called "star points" ($N_\alpha = 2n$); N_0 – the number of measurements in the center of the program (parallel or repeated measurements) ($N_0 = 3$ was chosen). Because in our case n = 4, the results given in relationship (eq.2) were obtained:

$$N_c = 2^{4-1} = 8,\ N_\alpha = 2 \cdot 4 = 8,\ N_0 = 3 \text{ and } N = 19 \quad (2)$$

The steps which run the research program are the following:

a) Based on the results obtained in the previous experiments, the experimental point in the factorial space of coordinates: $z_1^0 = 125^0C$; $z_2^0 = 2h$; $z_3^0 = 5atm$; $z_4^0 = 12.5$ (dimensionless) was chosen to be the base of the experiment, and the following extensions were chosen as variation interval: $\Delta z_1 = 10^oC$; $\Delta z_2 = 1h$; $\Delta z_3 = 1atm$; $\Delta z_4 = 2.5$ (dimensionless).

b) With these extensions the factor values were determined in the eight points of the subprogram EFF 2^{4-1} - subprogram which is a constituent of PO2 - using relationship (3):

$$z_i^{(-1)} = z_i^0 - \Delta z_i; \qquad z_i^{(+1)} = z_i^0 + \Delta z_i \quad (3)$$

c) The coordinates of the eight "star points" ($z_i^{(-\alpha)}$, $z_i^{(+\alpha)}$) were established using the eq. (5), with the help of parameter α, i.e. the "star arm", having a value of $\alpha = \pm 1.471$ determined by the bi-squared eq. (4):

$$\alpha^4 + 2^{n-1}\alpha^2 - 2^{n-2}(n + 0.5N_0) = 0, \quad (4)$$

$$z_i^{(-\alpha)} = z_i^0 - \alpha \cdot \Delta z_i; \qquad z_i^{(+\alpha)} = z_i^0 + \alpha \cdot \Delta z_i \quad (5)$$

d) Because the elaborated mathematical model operates with other type of variables, the next step taken in our research was the change of variables: instead of the natural variables z_1, z_2, z_3, z_4 we switched to codified variables x_1, x_2, x_3, x_4, by using relationships (6) and (7):

$$x_i^{(-1)} = \frac{z_i^{(-1)} - z_i^0}{\Delta z_i} = -1, \qquad x_i^{(+1)} = \frac{z_i^{(+1)} - z_i^0}{\Delta z_i} = +1, \quad (6)$$

$$x_i^{(-\alpha)} = \frac{z_i^{(-\alpha)} - z_i^0}{\Delta z_i} = -\alpha, \qquad x_i^{(+\alpha)} = \frac{z_i^{(+\alpha)} - z_i^0}{\Delta z_i} = +\alpha. \qquad (7)$$

The complete matrix of the PO2 program necessary for the computation of the regression equation coefficients has 16 columns. Among them one column is for the free term, four are for the linear terms, six are for the interactions, four are for quadratic terms and one is for experimental results. The columns from table 3 for the square terms have the following form in (eq. 8):

$$x'_{iu} = x_{iu}^2 - a, \qquad a = \frac{1}{N}\sum_{u=1}^{N=19} x_{iu}^2 = 0.649, \qquad i = 1,2,3,4 \qquad (8)$$

e) On the basis of everything presented above, the PO2 program used in research is presented in table 2, and the rest of the columns necessary for the complete matrix elaboration of this program are given in table 3.

f) According to the PO2 program, the computation relations and the values of regression equation coefficients have the forms given in eqs. (9) - (12):

$$b'_0 = \frac{1}{N}\sum_{u=1}^{N} y_u = 90.191, \qquad (9)$$

$$b_i = \frac{\sum_{u=1}^{N} x_{iu} y_u}{\sum_{u=1}^{N} (x_{iu})^2},$$

$$b_1 = 0.469; \qquad b_2 = 6.908; \qquad b_3 = -1.542; \qquad b_4 = 0.497 \qquad (10)$$

$$b_{ij} = \frac{\sum_{u=1}^{N} (x_{iu} x_{ju}) y_u}{\sum_{u=1}^{N} (x_{iu} x_{ju})^2},$$

$$b_{12} = b_{34} = -0.384; \quad b_{13} = b_{24} = 0.024; \quad b_{14} = b_{23} = 2.251 \qquad (11)$$

$$b_{ii} = \frac{\sum_{u=1}^{N} x'_{iu} y_u}{\sum_{u=1}^{N} (x'_{iu})^2},$$

$$b_{11} = 0.779; \quad b_{22} = -3.834; \quad b_{33} = 0.135; \quad b_{44} = -0.029. \qquad (12)$$

The mathematical model represented by an order two regression equation with four variables has the form presented in eq. (13):

Table 2
PROGRAM PO2 (n = 4)

Exp.	Process parameters (codified units)				Process parameters (natural units)				η, (%)
	x_1	x_2	x_3	x_4	$T, (^0C)$ z_1	$t, (h)$ z_2	P_{O_2}, atm z_3	$L:S$ z_4	y
1	-1	-1	-1	-1	115	1	4	10	87.16
2	+1	-1	-1	+1	135	1	4	15	88.43
3	-1	+1	-1	+1	115	3	4	15	95.11
4	+1	+1	-1	-1	135	3	4	10	95.23
5	-1	-1	+1	+1	115	1	6	15	77.90
6	+1	-1	+1	-1	135	1	6	10	79.65
7	-1	+1	+1	-1	115	3	6	10	95.24
8	+1	+1	+1	+1	135	3	6	15	95.07
9	-1.471	0	0	0	110	2	5	12.5	92.71
10	+1.471	0	0	0	140	2	5	12.5	94.62
11	0	-1.471	0	0	125	0.5	5	12.5	70.88
12	0	+1.471	0	0	125	3.5	5	12.5	96.48
13	0	0	-1.471	0	125	2	3.5	12.5	92.59
14	0	0	+1.471	0	125	2	6.5	12.5	91.95
15	0	0	0	-1.471	125	2	5	9	89.57
16	0	0	0	+1.471	125	2	5	16	94.26
17	0	0	0	0	125	2	5	12.5	93.58
18	0	0	0	0	125	2	5	12.5	91.28
19	0	0	0	0	125	2	5	12.5	91.92

Exp.	Interactions						Factors of the process			
	x_1x_2	x_1x_3	x_1x_4	x_2x_3	x_2x_4	x_3x_4	x_1'	x_2'	x_3'	x_4'
1	+1	+1	+1	+1	+1	+1	0.351	0.351	0.351	0.351
2	-1	-1	+1	+1	-1	-1	0.351	0.351	0.351	0.351
3	-1	+1	-1	-1	+1	-1	0.351	0.351	0.351	0.351
4	+1	-1	-1	-1	-1	+1	0.351	0.351	0.351	0.351
5	+1	-1	-1	-1	-1	+1	0.351	0.351	0.351	0.351
6	-1	+1	-1	-1	+1	-1	0.351	0.351	0.351	0.351
7	-1	-1	+1	+1	-1	-1	0.351	0.351	0.351	0.351
8	+1	+1	+1	+1	+1	+1	0.351	0.351	0.351	0.351
9	0	0	0	0	0	0	1.516	-0.649	-0.649	-0.649
10	0	0	0	0	0	0	1.516	-0.649	-0.649	-0.649
11	0	0	0	0	0	0	-0.649	1.516	-0.649	-0.649
12	0	0	0	0	0	0	-0.649	1.516	-0.649	-0.649
13	0	0	0	0	0	0	-0.649	-0.649	1.516	-0.649
14	0	0	0	0	0	0	-0.649	-0.649	1.516	-0.649
15	0	0	0	0	0	0	-0.649	-0.649	-0.649	1.516
16	0	0	0	0	0	0	-0.649	-0.649	-0.649	1.516
17	0	0	0	0	0	0	-0.649	-0.649	-0.649	-0.649
18	0	0	0	0	0	0	-0.649	-0.649	-0.649	-0.649
19	0	0	0	0	0	0	-0.649	-0.649	-0.649	-0.649

Table 3
MATRIX OF THE PROGRAM PO2 (N=4)

$$\tilde{y}_1 = 90.191 + 0.469x_1 + 6.908x_2 - 1.542x_3 + 0.497x_4 - 0.384(x_1x_2 + x_3x_4) + 0.024(x_1x_3 + x_2x_4) + \\ + 2.251(x_1x_4 + x_2x_3) + 0.779(x_1^2 - 0.649) - 3.834(x_2^2 - 0.649) + 0.135(x_3^2 - 0.649) - 0.029(x_4^2 - 0.649) \quad (13)$$

respectively Eq. (14):

$$\tilde{y}_1 = 92.105 + 0.469x_1 + 6.908x_2 - 1.542x_3 + 0.497x_4 - 0.384(x_1x_2 + x_3x_4) + 0.024(x_1x_3 + x_2x_4) + \\ + 2.251(x_1x_4 + x_2x_3) + 0.779x_1^2 - 3.834x_2^2 + 0.135x_3^2 - 0.029x_4^2 \quad (14)$$

g) Eq. (14) was subject to a statistical analysis. The statistical parameters were calculated according to eqs. (15) and (16) to compute the Fisher criterion (F_c) value in relationship (17) in order to establish whether this model is adequate or not:

- reproductibility dispersion (points 17 – 19 in table 2)

$$s_0^2 = \frac{1}{3-1}\sum_{i=17}^{19}(y_i - \bar{y})^2 = 1.409; \quad (15)$$

- concordance dispersion

$$s_{con}^2 = \frac{1}{N-15}\sum_{i=1}^{N}(\tilde{y}_i - y_i)^2 = 21.538 \quad (16)$$

- value of *Fisher* criterion

$$F_c = \frac{s_{con}^2}{s_0^2} = 15.284 \quad (17)$$

Due to the fact that the calculated value (F_c) of the Fisher criterion is smaller than its value given in literature [12], ($F_T = F_{0.05(v1=4;\ v2=2)} = 19.247$ we can conclude that the eq. (14) representing the mathematical model is adequate for the studied process and can be used in optimization.

h) The last step was the establishment of optimal regimes that will lead to the maximization of sulphur oxidation yield ($\tilde{y}$). Due to the fact that the methodology of determining these regimes depends on the surface type of the response (elliptic or hyperbolic paraboloid), the mathematical model represented by the regression eq. (14) was first brought to the so called canonical form, obtaining eq. (18):

$$\tilde{y} - 95.115 = -4.141\tilde{x}_1^2 + 1.589\tilde{x}_2^2 - 0.827\tilde{x}_3^2 + 0.429\tilde{x}_4^2 \quad (18)$$

For the canonical coefficients having different signs, the response surface is a hyperbolic paraboloid, and the optimal regime determination is in this case based on a known method: the Lagrange multiplier method.

The following system of equations was consisted of (eqs. 19):

$$(b_{11} - \lambda)x_1 + 0.5b_{12}x_2 + 0.5b_{13}x_3 + 0.5b_{14}x_4 + 0.5b_1 = 0$$

$$0.5b_{12}x_1 + (b_{22} - \lambda)x_2 + 0.5b_{23}x_3 + 0.5b_{24}x_4 + 0.5b_2 = 0$$

$$0.5b_{13}x_1 + 0.5b_{23}x_2 + (b_{33} - \lambda)x_3 + 0.5b_{34}x_4 + 0.5b_3 = 0$$
$$0.5b_{14}x_1 + 0.5b_{24}x_2 + 0.5b_{34}x_3 + (b_{44} - \lambda)x_4 + 0.5b_4 = 0 \quad (19)$$

where λ represents the Lagrange multiplier, b_i, b_{ii}, b_{ij} are the mathematical model coefficients determined with the eqs. (10) – (12). Since solving the system (eqs. 19) depends on the values of the λ multiplier – in practice – in almost all problems, these values are chosen so that the following inequality is satisfied (relation 20) [10, 12-14]:

$$a_{\max} < \lambda \leq 2(a_{\max} - b_{pp}) \quad (20)$$

where a_{max} is the canonical coefficient of maximum value, b_{pp} – the coefficient of the lowest quadratic term in the regression eq. (14). In our case, (relation 20) became: $1.589 < \lambda \leq 10.847$.

A few values of λ were chosen in the considered interval and for each considered value the system was solved (eqs. 19). The following result was obtained for the optimal regime, taking into account the calculation error ($\delta_{\tilde{y}}$) of the theoretical yield ($\tilde{y}$):

$$\lambda = 1.738; \quad \hat{x}_1 = 1.314; \quad \hat{x}_2 = 0.533; \quad \hat{x}_3 = -0.217; \quad \hat{x}_4 = 1.004;$$

$$\tilde{y}_{teoretic} = 100\%; \quad \delta_{\tilde{y}} = 4.06\%; \quad \tilde{y}_{practic} \geq 95.94\%.$$

The natural values of the process parameters were established using the relationship (7).

$$\hat{z}_1 = 138^0 C; \quad \hat{z}_2 = 2.53\,h; \quad \hat{z}_3 = 4.78\,atm; \quad \hat{z}_4 = 15.$$

The subject has been also discussed in other study [15].

Conclusions

The preliminary research evidenced four parameters significantly influencing the oxidation process performance (sulphur oxidation yield - y): temperature (z_1), °C; process duration (z_2), h; oxygen partial pressure (z_3), atm; L:S ratio, dimensionless (z_4).

The mathematical model - obtained with the order two orthogonal program (PO2), presented as a second order polynomial of four variables - was subject to statistical analysis; using the Fisher criterion we verified the correlation between the model and the data.

The optimal values of the oxidation process parameters are: $\hat{z}_1 = 138^0 C$; $\hat{z}_2 = 2.53\,h$; $\hat{z}_3 = 4.78\,atm$; $\hat{z}_4 = 15:1$; taking into account the computational error of the process performance , the maximum yield expected is $\tilde{y}_{practic} \geq 95.94\%$.

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