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## Effect of hydrogen peroxide and lead(II) nitrate on gold cyanide leaching of Malaysian mesothermal deposit gold ore

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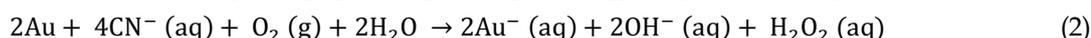
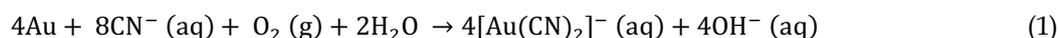
**Abstract:** The gold extraction from Malaysian mesothermal lode gold ore through the cyanidation method was performed. The effect of hydrogen peroxide ( $H_2O_2$ ) and lead nitrate  $Pb(NO_3)_2$  were investigated on the percentage of gold recovery. The statistical analysis method using a response surface design-central composite design was applied to find the optimum condition for gold extraction. The studied parameters were NaCN concentration (300-500 ppm),  $H_2O_2$  concentration (205-410 ppm), and  $Pb(NO_3)_2$  concentration (50-150 ppm). It was found that increasing the amount of NaCN and  $H_2O_2$  as well as decreasing the amount of  $Pb(NO_3)_2$  in the studied range, increased the gold recovery. The analysis of variance suggested the linear model for the gold extraction with the optimum condition at 467.3 ppm NaCN, 94.96 ppm  $Pb(NO_3)_2$ , and 340 ppm  $H_2O_2$ . The gold extracted from the ore at the optimum condition was 88.97% as compared to 62.02% from conventional cyanidation. The characterization study of the gold ore indicated that the finely gold particles interlocked in the aluminosilicate phase (grain size of  $\sim 10.0 \mu m$ ). The Van't Hoff differential method justified that the cyanidation was of second order with a specific reaction rate of 0.0501/hour.

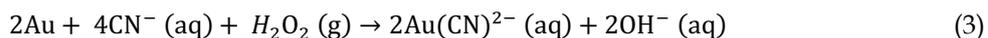
**Keywords:** gold cyanidation, mesothermal lode gold, statistical analysis, design of experiment, Van't Hoff differential method

### 1. Introduction

Cyanide lixiviant has been used as a leaching medium in the extraction of gold from the ore for more than a century due to the selectivity and chemical stability of the aurocyanide compound. The gold cyanide complexes such as  $[Au(CN)_2]^-$  and  $[Au(CN)_4]^-$  having stability constant of  $2 \times 10^{38}$  and  $\sim 10^{54}$ , respectively (Marsden and House, 2006). According to Ofori-Sarpong and Osseo-Asare (2013), cyanidation is economically value, and gives a better understanding of the chemical process. Although cyanide is considered a toxic and harmful chemical, however proper handling of the substance will not harm the ecosystem.

The cyanidation process for gold ore is described by Elsner's equation as Eq. 1, in which oxygen plays an important role in gold cyanidation (Liu and Yen, 1995). Later studies suggested that the cyanidation process occurs in two steps:  $O_2$  is reduced and hydrogen peroxide ( $H_2O_2$ ) is formed at the beginning of the reaction as Eq. 2, and further proceeds with the reduction of  $H_2O_2$  as shown in Eq.3.  $H_2O_2$  is used in this process to assist in the gold dissolution process (Guzman et al., 1999; Yang et al., 2010; Esmkhani, 2013). Moreover, it is environmentally safe because of the decomposition of  $H_2O_2$  yields only  $O_2$  and  $H_2O$ . It should be noted that the oxygen or air aeration in the cyanidation on site is an unfavourable option owing to its prohibitive costs especially in remote areas (Deschênes and Wallingford, 1995; Deschênes et al., 2009).





The recovery of refractory gold ore by direct cyanidation method was only about 66.0% as compared to free milling ore (Mahmoodi et al., 2010). Gold extracted from free milling ore could be up to 90.0% when the ore is ground to 80.0% passing 75  $\mu\text{m}$  (Karimi et al., 2010). As mentioned by La Brooy et al. (1994) the locking of gold in sulphide and silicates prevent the leach solutions from dissolving the gold. The sulphide mineral forms a passive layer on the surface of the gold particles resulting in low recovery of gold (Rees and van Deventer, 2000; Senanayake, 2005). Fine grinding (<38  $\mu\text{m}$ ) neither improve the gold recovery, nor overcomes the detrimental effect of the sulphide minerals towards the gold dissolution (Senanayake, 2004; Celep, 2009). It has reported that the roasting of the gold sulphide ores prior to cyanidation may remove the arsenic and sulphide as a volatile  $\text{As}_2\text{O}_3$  and  $\text{SO}_2$  components, respectively (Aylmore, 2001; Mular et al., 2002). However, partial oxidation of sulphides produces sulphur that may react with cyanide to form thiocyanate which is adversely affecting the cyanidation process. Furthermore, high operating costs and emission of toxic gaseous from roasting are unfavourable. Therefore, the addition of lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) in cyanidation able to overcome the detrimental effect of sulphide minerals and increases the gold recovery (Deschênes et al., 1998; Dai and Breuer, 2013).

The injection/aeration of  $\text{Pb}(\text{NO}_3)_2$  and  $\text{O}_2$  in cyanidation has been extensively studied (Kondos, 1996; Ellis and Senanayake, 2004); however, the information about the effect of  $\text{H}_2\text{O}_2$  and  $\text{Pb}(\text{NO}_3)_2$  together on the gold extraction from cyanidation process is limited. In this study, the effect of lead nitrate addition together with  $\text{H}_2\text{O}_2$  on the cyanidation process and the reaction kinetic were investigated.

## 2. Materials and methods

### 2.1. Gold ore

The gold ore used in this study was provided from a local gold mine in Peninsular Malaysia. The gold ore was characterized using X-ray fluorescence (XRF, Rix 3000, Rigaku), X-ray diffraction (XRD, PW Philip 1820) with  $\text{Cu-K}\alpha$  radiation (1.5406 Å), and scanning electron microscope/energy dispersive X-ray (SEM/EDX, Gemini SUPRA 35VP-ZEISS) to identify the department of gold and association of other minerals, the elemental composition of the ore, and the available phases.

### 2.2. Determination of gold concentration in gold ore

All the chemical reagents were provided by Merck, Germany as an analytical grade. The concentration of gold in the gold ore was determined by bulk leach extractable gold (BLEG) and fire assay methods. The BLEG method was conducted using 1000 ppm of NaCN solution. As for the fire assay method, ~50.0 g of gold ore was mixed with a flux (48.0-50.0%  $\text{Na}_2\text{CO}_3$ , 25.0%  $\text{PbO}$ , 20.0%  $\text{Na}_2\text{B}_4\text{O}_7$ , 0.5%  $\text{AgNO}_3$ , and 4.5-6.50% flour) before fusion at 1000-1100 °C for 1 h using a Lenton muffle furnace, UK. The prill obtained from the fire assay method was digested with the acids and analysed using inductively coupled plasma-optical emission spectroscopy (ICP-OES, Optima 7300 DV, Perkin Elmer).

### 2.3. Gold cyanidation

The cyanidation experiments were conducted under stirred condition at 450-500 rpm with solid:liquid (S:L) ratio of 1:3, pH 11, and room temperature (26.5 °C). The filtrates obtained were pre-concentrated using diisobutyl ketone (DIBK) by solvent extraction technique. The DIBK was treated with 1.0% Aliquat 336 and about 20.0% KCl (25.0%) solution before extraction of aurocyanide into the organic phase, which were later analysed by flame atomic absorption spectrometry (FAAS, AA 700, Perkin Elmer).

### 2.4. Pre-leaching experiments

The preliminary cyanidation experiments were conducted to study the effect of factors that mostly affecting the gold leaching process including particle size, concentration of NaCN, concentration of

$\text{Pb}(\text{NO}_3)_2$ , and concentration of  $\text{H}_2\text{O}_2$ . Samples were taken for each experiment after 2, 4, 6, 8, and 24 h to investigate the behaviour of gold dissolution rate during the experiment.

## 2.5. Statistical approach

The statistical analysis using response surface design–central composite design (RSM-CCD) was used in this study. The RSM-CCD is a strong tool for optimizing the industrial and research processes (Um and Bae, 2011; Im et al., 2012; Asadollahzadeh et al., 2014). According to the results of pre-leaching experiments, the effect of three main factors on the percentage of gold recovery was investigated with Design Expert 11 software (State-Ease, USA). The studied factors with their respected levels were NaCN concentration (300-500 ppm),  $\text{H}_2\text{O}_2$  concentration (205-410 ppm), and  $\text{Pb}(\text{NO}_3)_2$  concentration (50-150 ppm). The total number of experiments as summarized in Table 1 consisted of a  $2^3$  full factorial CCD points, 6 axial points, and 6 replicates of centre points indicating 20 experiments were calculated from Eq. 4. The centre points were included to provide a reasonably stable variance of the predicted response (Aslan, 2007). The optimum conditions for the cyanidation process were obtained by using the Design Expert 11 software.

$$N = 2^n + 2n + n_c \quad (4)$$

where, N is total number of experiments; n is the number of studied factors, and  $n_c$  is the number of centre points.

## 2.6. Kinetic study

The reaction order of the isothermal cyanidation was determined by using the Van't Hoff differential method with reference to Eq. 4, where V is the reaction rate, C is the reactant concentration, n is the reaction order and k is the reaction rate constant. In order to find the reaction order, Eq. 5 can be written as Eq. 6.

$$V = kC^n \quad (5)$$

$$\log V = \log k + n \log c \quad (6)$$

Table 1. Experimental data of the design matrix generated by RSM-CCD and the corresponding gold extracted from cyanidation experiments

Run	A: Hydrogen Peroxide (ppm)	B: Lead nitrate (ppm)	C: Sodium Cyanide (ppm)	Au Extracted (%)
1	205.00	150.00	300.00	27.09
2	410.00	50.00	300.00	31.86
3	307.50	100.00	400.00	45.94
4	410.00	150.00	500.00	65.86
5	410.00	150.00	300.00	30.19
6	205.00	50.00	300.00	33.40
7	307.50	100.00	400.00	50.62
8	205.00	150.00	500.00	57.89
9	307.50	100.00	400.00	50.64
10	410.00	50.00	500.00	79.81
11	307.50	100.00	400.00	53.53
12	205.00	50.00	500.00	70.28
13	307.50	100.00	400.00	44.24
14	135.00	100.00	400.00	42.37
15	307.50	100.00	400.00	53.80
16	307.50	16.00	400.00	52.74
17	480.00	100.00	400.00	48.87
18	307.50	100.00	568.20	62.95
19	307.50	185.00	400.00	53.41
20	307.50	100.00	231.80	21.10

If the  $V$  is known at the various values of  $c$ , a plot of the  $\log V$  versus  $\log c$  will result in a straight line and the slope of the linear trend will determine the reaction order with respect to the concentration of the substance.

### 3. Results and discussion

#### 3.1. Characterization of gold ore

The total concentration of gold present in the gold ore was found to be 1.94 g/t by performing the BLEG and fire assay techniques. The XRF analysis result is tabulated in Table 2. It was found that the gold presents in traces amount, whereas, Si (34.19%) and Al (9.25%) are the major components in the gold ore. The elemental composition of S, Fe, and As obtained from the XRF analysis indicated the elemental composition of sulphide elements.

Table 2. Elemental composition of XRF analysis results of the gold ore

Element	Weight (%)	Element	Weight (%)
Na	0.04	Mn	0.01
Mg	0.12	Fe	1.99
Al	9.25	As	0.14
Si	34.19	Rb	0.02
P	0.02	Y	0.01
S	0.01	Zr	0.01
Cl	0.01	W	0.01
K	2.79	Pb	0.01
Ca	0.01	V	0.01
Ti	0.18	Ni, Cu, Zn, Ga, Sr, Au	Traces

XRD analysis represents that the main phases present in the gold ore were quartz (Q:  $\text{SiO}_2$ ) and aluminium silicate (kyanite, A:  $\text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ ) as shown in Fig. 1. Moreover, small peaks related to aluminium oxide ( $\text{Al}_2\text{O}_3$ ) was observed. The powder diffraction file numbers for the minerals are 98-003-4023 (quartz), 98-004-5600 (aluminium silicate), and 98-002-2959 (aluminium oxide).

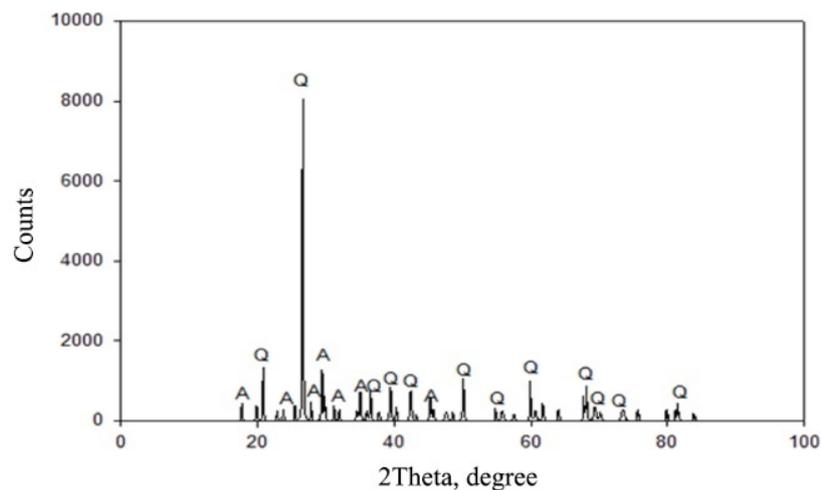


Fig. 1. XRD diffractogram showing phases of quartz (Q) and aluminium silicate (A) present in the gold ore

The SEM/EDX analysis was performed to study morphology and elemental analysis of gold ore as shown in Figs. 2 and 3. The result in Fig.2 illustrates that the gold particles were interlocked in Al, Si, and O, which is indicated by the label A. As it can be seen the matrix is mainly contained Si and O. The gold particles were found to be finely disseminated with the grain size of  $\sim 10.0 \mu\text{m}$ . On the other hand, the SEM photomicrograph in Fig. 3 reveals the presence of sulphide minerals.

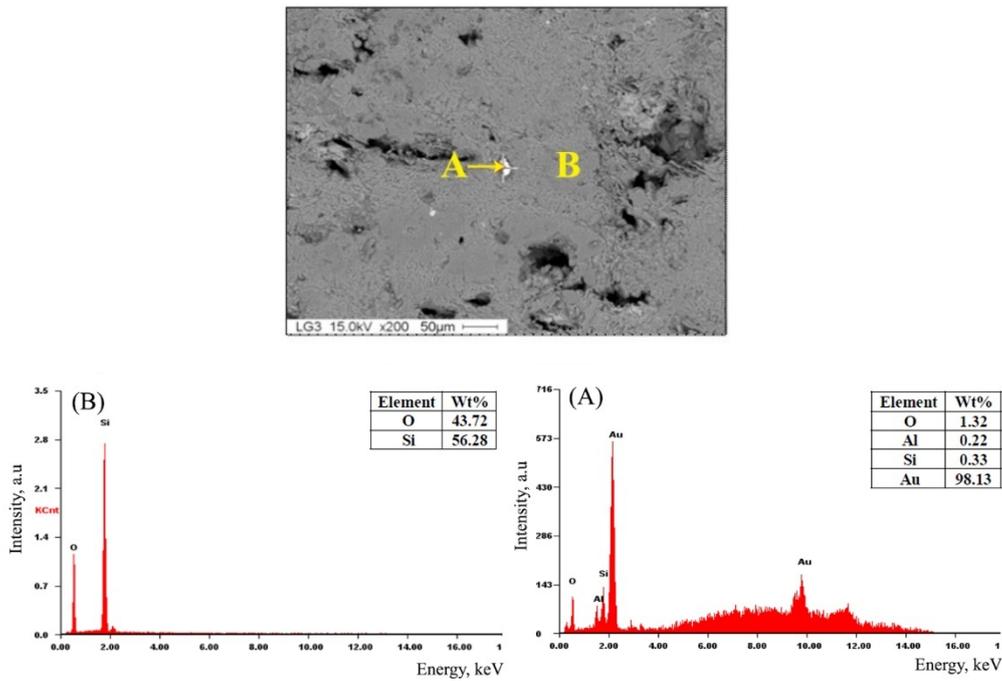


Fig. 2. SEM/EDX image of the gold ore polished section showing the presence of (a) O, Al, Si and Au at labelled spot A and (b) Si and O at labelled spot B

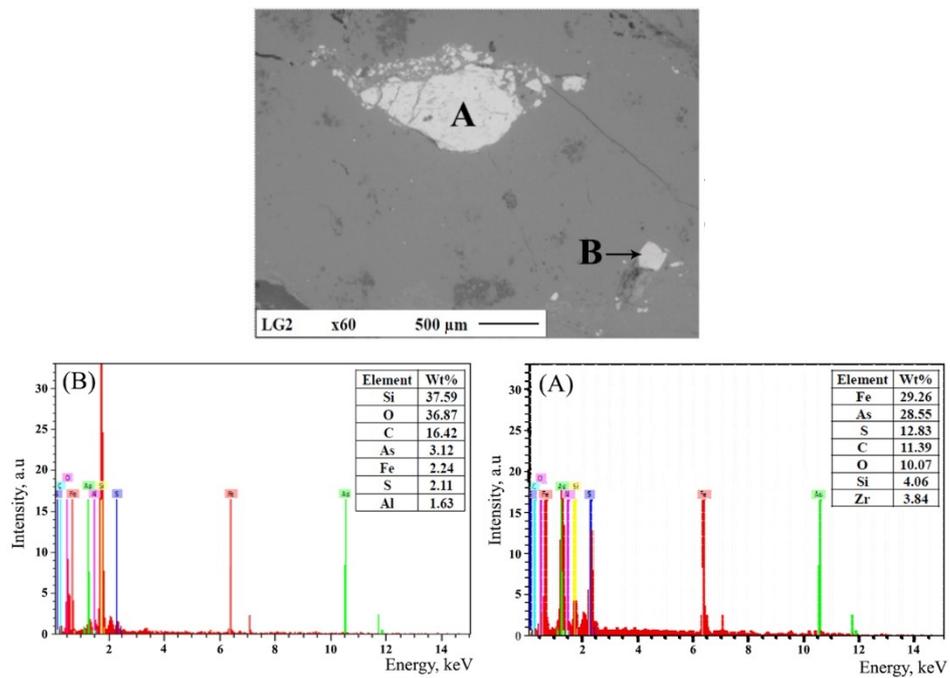


Fig. 3. SEM/EDX image of the gold ore polished section showing the presence of (a) Fe, As, S, Zr, Si, and O, at labelled spot A and (b) Si, O, C, As, Fe, S and Al at labelled spot B

### 3.2. Pre-leaching study

#### 3.2.1. Effect of particle size fraction

The dissolution of gold is directly proportional to the exposed surface area of gold which determines the liberation characteristic of the low-grade gold ore. Figure 4 represents the effect of two different particle size fractions on the percentage of gold extraction. The total percentage of gold recovery were 39.04% (80.0% <75 μm) and 49.79% (90.0% <75 μm). It can be seen that the percentage of gold extraction

after 4 h is almost 10% higher for finer particles and there is no specific change with increasing the time up to 24 h. This is due to the larger surface area of the gold particles were in contact with the leaching solution, and the flattening or physical breakage during the grinding process. These results suggest that particle size of 90.0% <75  $\mu\text{m}$  can increase the dissolution of gold due to the higher amount of gold particles liberated from the interlocking of silicates minerals.

### 3.2.2. Effect of cyanide concentration

The percentage of gold extraction is proportional to the cyanide concentration; however, regarding the environmental concerns, the low cyanide concentration range was chosen in this research. The NaCN concentrations were 300, 400, and 500 ppm. From Fig. 5, it was found that the gold extraction rate increased with an increase in the NaCN concentration from 300 to 500 ppm during the 24 h of the experiment. The gold dissolution amounts for NaCN concentrations of 400 and 500 ppm reached the equilibrium after 4 h with an extraction amount of 49.79% and 56.29%, respectively. The amounts of gold recovery were not promising due to the refractoriness of the gold ore samples, which discussed in the previous section. The sulphide minerals that present in the gold ore might compete with the gold to complex with the cyanide. On the other hand, the NaCN concentrations were at the low range. Hence, the using of NaCN alone with a low concentration was not enough to react with the gold and other gangue elements present in the gold ore sample, which are also the cyanide consumers.

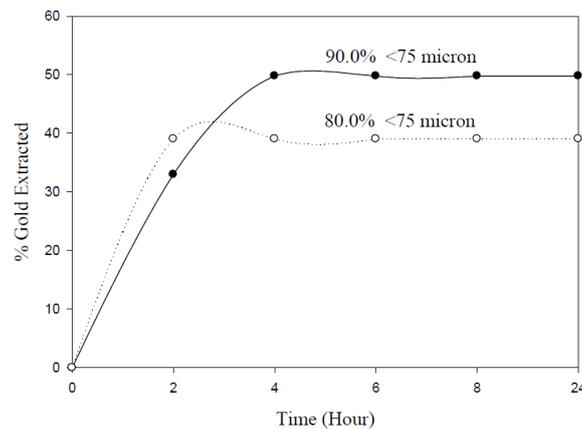


Fig. 4. Effect of particle size on the gold cyanidation over time. (cyanidation condition: 400 ppm NaCN concentration, S:L = 1:3; pH = 11; temperature: 26 °C)

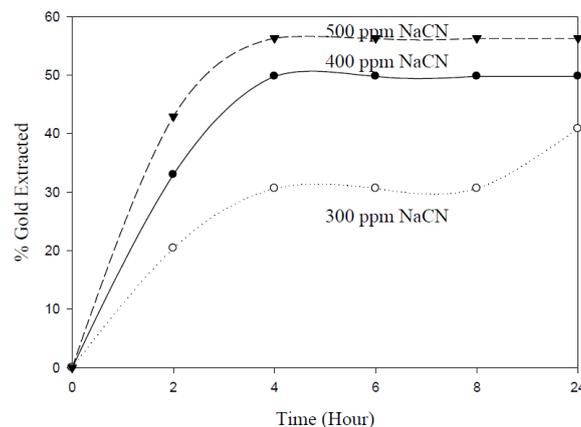


Fig. 5. Effect of NaCN concentration on the gold cyanidation over time. (cyanidation conditions: S:L = 1:3; pH = 11; temperature: 26 °C)

The gold dissolution rate for each NaCN concentration is tabulated in Table 3. The rate difference in the gold dissolution for 300 and 400 ppm cyanide was 0.0073 g/t per hour. Meanwhile, the rate

difference associated with the 400 and 500 ppm cyanide was 0.0051 g/t per hour. It can be seen with an increase in the cyanide concentration, the differences in the gold dissolution rates were decreased. It has reported that the excess amount of NaCN has no effect on the gold dissolution rate, when maximum cyanide concentration of 600 ppm was reached (Shrithammavut, 2008). The excessive cyanide results in the dissolution of sulphide ions, which react with the free cyanide and consume oxygen to form thiocyanate as shown in Eq. 7. This is also a process of passivation film formation which inhibits the gold dissolution process (Deschênes and Wallingford, 1995).

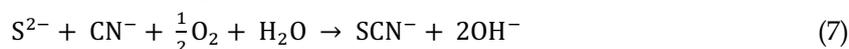


Table 3. Gold dissolution rate for cyanidation of the gold ore sample at varying concentrations of cyanide solution

CN <sup>-</sup> concentration (ppm)	Gold dissolution rate (g t <sup>-1</sup> h <sup>-1</sup> )
300	0.0329
400	0.0402
500	0.0454

### 3.2.3. Effect of hydrogen peroxide addition

The H<sub>2</sub>O<sub>2</sub> which contributes to an increase in the dissolved oxygen concentration in the leaching solution was added to the cyanide leaching process to enhance the gold dissolution rate based on Elsner's equation (Eq. 1). To investigate the effect of H<sub>2</sub>O<sub>2</sub> on the gold extraction, the NaCN concentration was set to 400 ppm with varying the concentration of H<sub>2</sub>O<sub>2</sub>. Figure 6 indicates that the addition of 205 ppm H<sub>2</sub>O<sub>2</sub> had little effect on the gold extraction process compared to the control experiment of 400 ppm NaCN without the addition of H<sub>2</sub>O<sub>2</sub>. With an increase in the concentration of H<sub>2</sub>O<sub>2</sub> from 205 to 410 ppm, the amount of gold recovery was increased from 48.52% to 68.85%. A comparison between the cyanidation leaching using 500 ppm NaCN and the cyanidation process with 400 ppm NaCN and 410 ppm H<sub>2</sub>O<sub>2</sub> showed that the gold recovery in the later one is higher by 12.56%. According to Dai and Breuer (2013), the addition of a low concentration of H<sub>2</sub>O<sub>2</sub> does not directly enhance the dissolution rate of gold, but it may act as an additional oxidant when the amount of dissolved oxygen is low. The H<sub>2</sub>O<sub>2</sub> can decompose to oxygen, which is a process that can be catalysed by the gold ore sample.

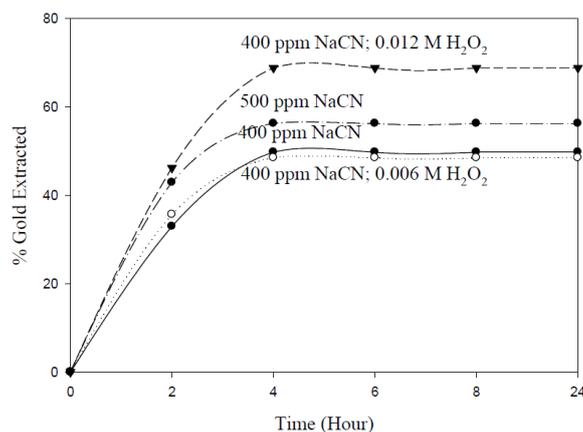


Fig. 6. The effect of H<sub>2</sub>O<sub>2</sub> concentration on the gold extraction. (cyanidation condition: 400 ppm NaCN concentration; pH = 11; S:L = 1:3; temperature: 26 °C)

Table 4 summarizes the gold dissolution rate for various amounts of H<sub>2</sub>O<sub>2</sub> in the cyanide leaching solution. It is evident that the gold dissolution rate increased with the addition of a higher concentration of H<sub>2</sub>O<sub>2</sub>. However, the gold dissolution rate dropped when 476 ppm H<sub>2</sub>O<sub>2</sub> was used. It was reported that the high concentration of H<sub>2</sub>O<sub>2</sub> (476 ppm) may retard the gold dissolution due to cyanide oxidation by peroxide (Guzman et al., 1999). Thus, a suitable concentration of H<sub>2</sub>O<sub>2</sub> should be carefully selected for the experiments on optimization of the gold cyanidation process which will be discussed later.

Table 4. Gold dissolution rate for cyanidation of the gold ore sample by varying hydrogen peroxide concentrations. (cyanidation conditions: 400 ppm cyanide concentration; S:L = 1:3; pH = 11; temperature: 26 °C)

H <sub>2</sub> O <sub>2</sub> concentration (ppm)	Gold dissolution rate (g t <sup>-1</sup> h <sup>-1</sup> )
205	0.0392
306	0.0402
410	0.0556
476	0.0394

### 3.2.4. Effect of lead nitrate addition

The addition of lead nitrate in the gold cyanidation process was carried out to overcome the detrimental effects of the sulphide minerals. The addition of 150 ppm Pb(NO<sub>3</sub>)<sub>2</sub> resulted in an increase in the gold recovery (59.81%) compared to 50 ppm Pb(NO<sub>3</sub>)<sub>2</sub> (49.79%) and control experiment (without Pb(NO<sub>3</sub>)<sub>2</sub>) as shown in Fig. 7. At 400 ppm NaCN with 150 ppm Pb(NO<sub>3</sub>)<sub>2</sub>, the gold extraction was even higher than 56.29% gold recovery associated with the leaching process with 500 ppm NaCN without the addition of Pb(NO<sub>3</sub>)<sub>2</sub>. At 500 ppm NaCN, the curve reached the plateau after 4 h compared to 400 ppm NaCN and 150 ppm Pb(NO<sub>3</sub>)<sub>2</sub>, where equilibrium was observed after 6 h. Previous study by Breuer et al. (2008) reported that the reaction between Pb(NO<sub>3</sub>)<sub>2</sub> with the sulphide minerals forms lead sulphide, which acts as a catalyst for sulphide oxidation, releasing the lead ions to further react with sulphide ions preventing the sulphide ions from passivating the gold surface. In other words, lead ions can react with the sulphide films in the gold ore and making the passivated gold become reactive in the absence of sulphide formation.

Although the addition of Pb(NO<sub>3</sub>)<sub>2</sub> can assist in accelerating the gold dissolution, passivation of the gold surface can also occur in a high concentration of lead. According to De Andrade Lima and Hodouin (2005), the addition of sufficient amount of lead nitrate can minimize cyanide consumption by the sulphides. The addition of a small quantity of lead salt to the cyanide leaching solution can stabilize the copper and iron dissolution by forming a passivation layer at the surface of the sulphide minerals, and preventing the formation of a passive layer on the surface of gold (Deschênes et al., 1998). It can be seen from Table 5 that the gold dissolution rate increased with the addition of lead nitrate in the gold cyanidation process. It was reported that the Pb(NO<sub>3</sub>)<sub>2</sub> plays an important role in enhancing the gold dissolution rate not only in passivating the sulphide minerals surface, but also forming a precipitate on gold grains to form a corrosion pile which makes gold anodic not easy to dissolve (Deschênes et al., 2000).

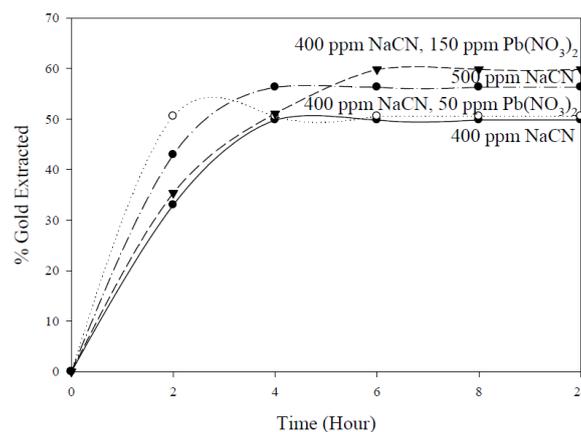


Fig. 7. Effect of Pb(NO<sub>3</sub>)<sub>2</sub> addition on the gold extraction. (cyanidation condition: 400 ppm NaCN concentration; pH = 11; S:L = 1:3; temperature: 26 °C)

### 3.3. Statistical analysis

The results of pre-leaching experiments highlighted the effect of NaCN, Pb(NO<sub>3</sub>)<sub>2</sub>, and H<sub>2</sub>O<sub>2</sub> concen-

Table 5. Gold dissolution rate for cyanidation of the gold ore sample by varying  $\text{Pb}(\text{NO}_3)_2$  concentrations. (cyanidation conditions: 400 ppm cyanide solution; pH = 11; S:L = 1:3; temperature: 26 °C)

$\text{Pb}(\text{NO}_3)_2$ concentration (ppm)	Gold dissolution rate ( $\text{g t}^{-1} \text{h}^{-1}$ )
0	0.0402
50	0.0408
100	0.0402
150	0.0483

tration on the percentage of gold recovery. The optimization process of the cyanidation leaching for the maximum extraction of gold was carried out with the design of experiment (DOE) method. Noteworthy that based on the pre-leaching experiments, the particle size and leaching duration were set to constant amount of 90.0% <75  $\mu\text{m}$  and 24 h for all experiments. The results showed that the lowest gold recovery (21.10%) obtained in run #20, where the main factors of  $\text{H}_2\text{O}_2$ ,  $\text{Pb}(\text{NO}_3)_2$ , and NaCN concentrations were 306 ppm, 100 ppm, and 231.80 ppm, respectively. On the other hand, the maximum amount of gold recovery (79.81%) was associated with the run #10 with the experimental conditions of 410 ppm  $\text{H}_2\text{O}_2$ , 50.00 ppm  $\text{Pb}(\text{NO}_3)_2$ , and 500.00 ppm NaCN. The analysis of variance (ANOVA) results by Design Expert 11 software suggested the Quadratic model for inverse transformation of gold cyanidation based on the low standard deviation, high  $R^2$  and adjusted- $R^2$  values, lowest p-value, and with probability  $F < 0.0001$ , which indicates the model is significant (Ong et al., 2015). The models were developed based on the highest order polynomials, where additional terms were significant and the model was not aliased as shown in Table 6.

Table 6. ANOVA analysis for various response surface models for gold cyanidation

Source	Std. Dev.	$R^2$	Adjusted $R^2$	Predicted $R^2$	PRESS*	Sequential p-value	
Linear	0.0041	0.8113	0.7760	0.6840	0.0005	<0.0001	
2FI	0.0045	0.8150	0.7297	0.6247	0.0005	0.9659	
Quadratic	0.0017	0.9795	0.9610	0.8987	0.0001	<0.0001	Suggested
Cubic	0.0016	0.9906	0.9643			0.4281	Aliased

\* Predicted Residual Error Sum of Squares

The value of the signal to noise ratio was 27.036, which indicates an adequate signal. In general, a ratio greater than 4 is desirable and confirms that the predicted model can be used to navigate the design space defined by CCD (Liu et al., 2011). The proposed quadratic model demonstrated a low standard deviation value of 0.0017, a high  $R^2$  value of 0.9795, and an adjusted  $R^2$  value of 0.9610. The Predicted  $R^2$  of 0.8987 is in reasonable agreement with the Adjusted  $R^2$ ; i.e. the difference is less than 0.2. The  $R^2$  value should be closed to 1 to show a good correlation between the experimental and predicted values. The empirical model for gold extracted is a second order polynomial model as shown in Eq. 8 (coded factor), where, A =  $\text{H}_2\text{O}_2$  (ppm), B =  $\text{Pb}(\text{NO}_3)_2$  (ppm), and C = NaCN (ppm).

$$\begin{aligned} \% \text{Au Extracted} = & 0.0202 - 0.0008 A + 0.0010 B - 0.0092 C - 0.0007 AB - 0.0002 AC - 0.0004 BC \\ & + 0.0005 A^2 - 0.0006 B^2 + 0.0039 C^2 \end{aligned} \quad (8)$$

The adequacy of the model was further justified through ANOVA where the 'Model F value' of 53.02 implies that the model is significant. There is only a 0.01% chance that the 'Model F value' this large could occur due to noise for gold cyanidation.

The adequacy of the suggested model can be examined by applying the normal probability plot of residuals and actual versus predicted value plot. A normal probability plot in Fig. 8(a) suggests that in terms of graphical analysis the residuals follow a normal distribution with the points follow the straight line without any specific pattern. The actual experimental data versus predicted data plot which illustrated in Fig. 8(b) was in good agreement between actual experimental data and the data predicted from the model.

The perturbation plot in Fig. 9 illustrates the effects of the main factors on the gold recovery. It is clear that the NaCN concentration (factor C) has the most significant impact on the gold extraction with

respect to the slope of the reference lines. In regard to the slope of the lines A and B, it is evident that  $\text{Pb}(\text{NO}_3)_2$  has a higher slope comparing to  $\text{H}_2\text{O}_2$  concentration. Therefore, it is placed as the second factor which has a higher impact on the gold extraction rather than  $\text{H}_2\text{O}_2$  concentration. Moreover, the positive slope of the lines C and A indicates that an increase in the NaCN and  $\text{H}_2\text{O}_2$  concentrations results in the increasing of the gold extraction. On the other hand, an increase in the  $\text{Pb}(\text{NO}_3)_2$  concentration follows by the slight decrease in the gold extraction.

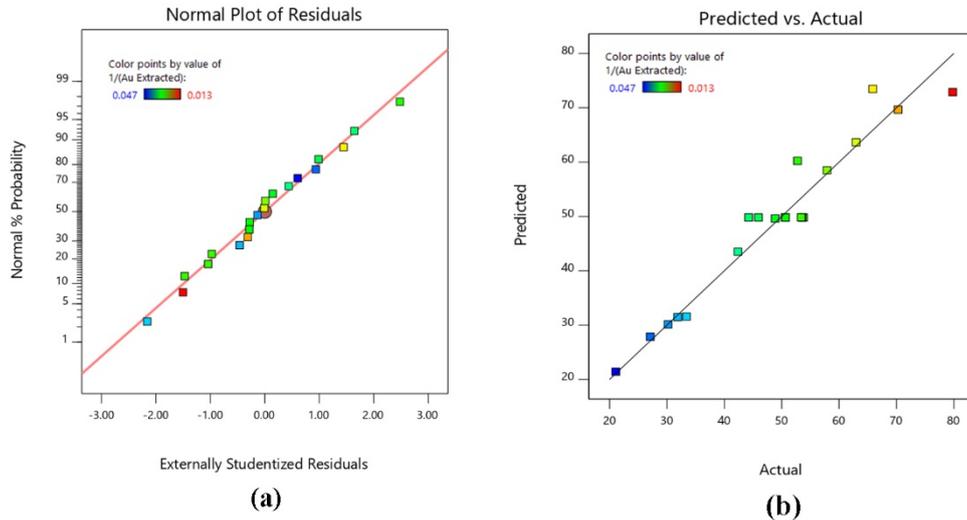


Fig. 8. Normal plot for (a) residuals for gold extracted, and (b) actual versus predicted value for gold cyanidation

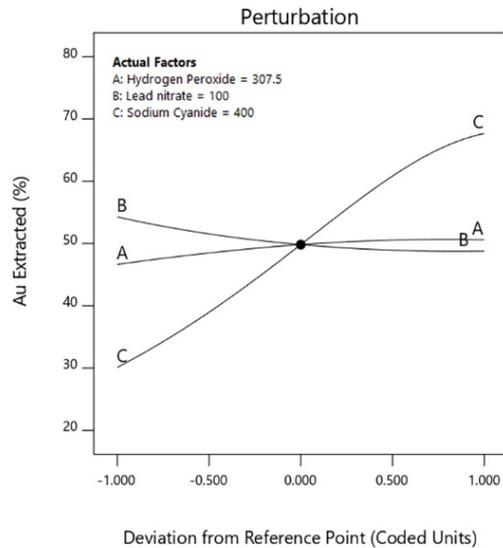


Fig. 9. Perturbation plot for gold recovery at reference point: 0.009 M  $\text{H}_2\text{O}_2$ ; 100 ppm  $\text{Pb}(\text{NO}_3)_2$ ; 400 ppm NaCN

The ANOVA analysis showed that the best response surface model for the gold extraction was the second order polynomial model. The contour and 3D surface plots for gold extraction is shown in Fig. 10. These plots are used to find the direction of potential improvement for the gold cyanidation using a path of steepest ascent in which two variables were set to constant values while the others fluctuated in the examined ranges. The presence of curvatures in the plots is associated with the interactions between the variables. According to Figs. 10(a) and 10(b) the maximum amount of gold recovery can be obtained at the highest NaCN concentration with a minimum amount of the  $\text{Pb}(\text{NO}_3)_2$  and maximum amount of  $\text{H}_2\text{O}_2$ , respectively. With respect to the effect of  $\text{H}_2\text{O}_2$  and  $\text{Pb}(\text{NO}_3)_2$  concentrations on the gold recovery, Fig. 10(c) indicates that there is no significant change in the gold extraction with the change in the associated factors.

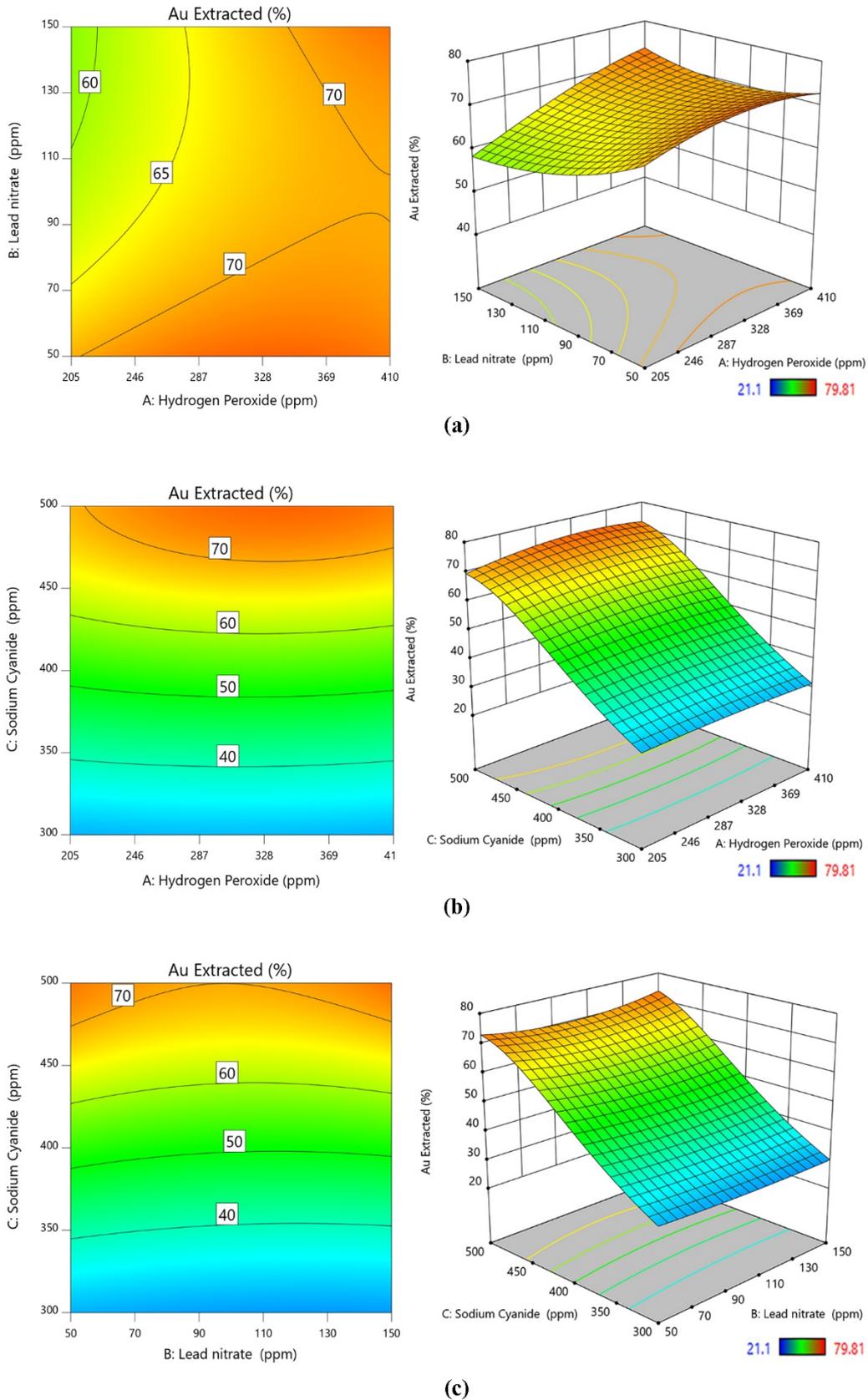


Fig. 10. Contour plot and 3D surface plot for gold cyanidation as a function of (a)  $H_2O_2$  and  $Pb(NO_3)_2$  concentrations at 500 ppm, (b) NaCN and  $H_2O_2$  concentrations at 100 ppm  $Pb(NO_3)_2$ , and (c) NaCN and  $Pb(NO_3)_2$  concentrations at 307.5 ppm  $H_2O_2$

### 3.3.1 Optimization

The ultimate goal of this study is to maximize the gold extraction with respect to the specific level of each parameters. The statistical analysis applied in the previous section to predict the model for leaching process, which diagnostics validated statistically. In order to optimize the process, numerical hill-climbing algorithms was applied in design-expert software to find out the most desirable results. Figure 11 represents the optimal conditions with associated desirability values for maximizing the gold recovery from leaching process. According to the optimization process, the best condition for gold cyanidation was found at 528.75 ppm NaCN, 185 ppm  $\text{Pb}(\text{NO}_3)_2$ , and 480 ppm  $\text{H}_2\text{O}_2$ . About 1.67 g/t of gold extracted which is equivalent to 86.80% of gold extracted from the gold as compared to 62.02% from conventional cyanidation method (Ding and Sartaj, 2016). It should be noted that Fig. 11 indicates the significant gold recovery up to 82.44% could be also obtained with 508.30 ppm NaCN, 16 ppm  $\text{Pb}(\text{NO}_3)_2$ , and 292.73 ppm  $\text{H}_2\text{O}_2$  with desirability of 0.94.

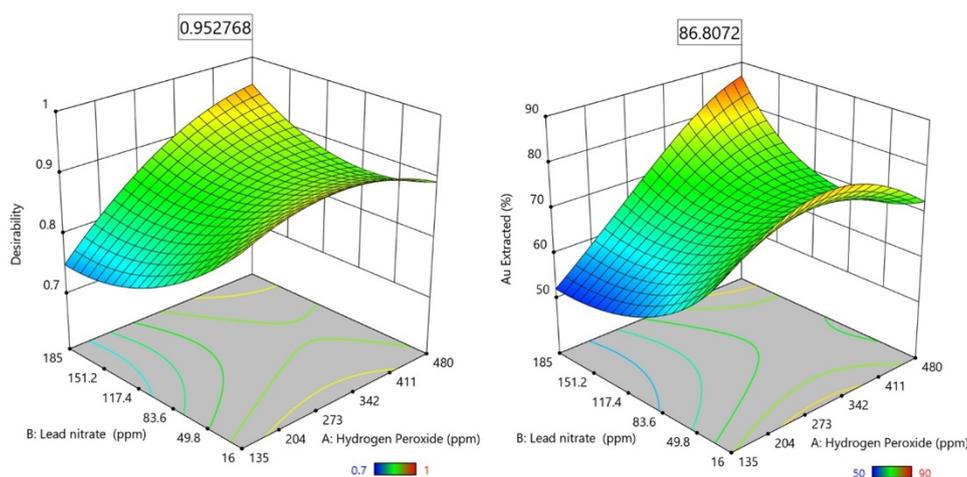


Fig. 11. 3D view of most desirable operating conditions (best condition at  $\text{H}_2\text{O}_2$ : 480 ppm, NaCN: 528.75 ppm,  $\text{Pb}(\text{NO}_3)_2$ : 185 ppm)

### 3.4. Van't Hoff differential method

The reaction order of gold cyanidation at the optimum condition was determined by adopting Van't Hoff differential method. This method is used when the temperature is constant (isothermal reaction) (Upadhyaya and Dube, 1977). Table 7 represents the amount of  $\log c$  and  $\log V$  for different points of the gold extraction at optimum condition over time. The sampling was done at different time intervals according to Fig. 12(a). The linear graph of  $\log V$  against  $\log c$  is plotted as Fig. 12(b). The slope of the diagram is 1.5 ( $\sim 2.0$ ) which can be regarded as the second order reaction. The intercept of the line at  $\log c = 0$  is equivalent to -1.3. With reference to Eq. 5, the specific reaction rate,  $k$  can be calculated by using the intercept value. From this method, it is verified that the reaction order for gold cyanidation at the vicinity of optimum condition is of second order with the specific reaction rate of  $k = 0.0501/\text{hour}$ .

Therefore, it can be concluded that the gold complex extracted from cyanidation was based on the CN concentration and dissolved oxygen produced by  $\text{H}_2\text{O}_2$ . Moreover, the  $\text{Pb}(\text{NO}_3)_2$  is used to stabilize copper and iron which have a passivation effect on the gold particle surface.

Table 7. concentration of gold extracted and rate of gold extracted at the optimum condition

Point No.	Time (h)	Gold Extracted (%) (C)	Log c	Slope of the Tangent	Rate of Gold Extracted % Au/hour (V)	LogV
i	2	48.45	1.69	-16.0	16.00	1.20
ii	4	23.24	1.37	-11.67	11.67	1.07
iii	6	12.94	1.11	-0.90	0.90	-0.05
iv	24	4.34	0.64	-0.53	0.53	-0.28

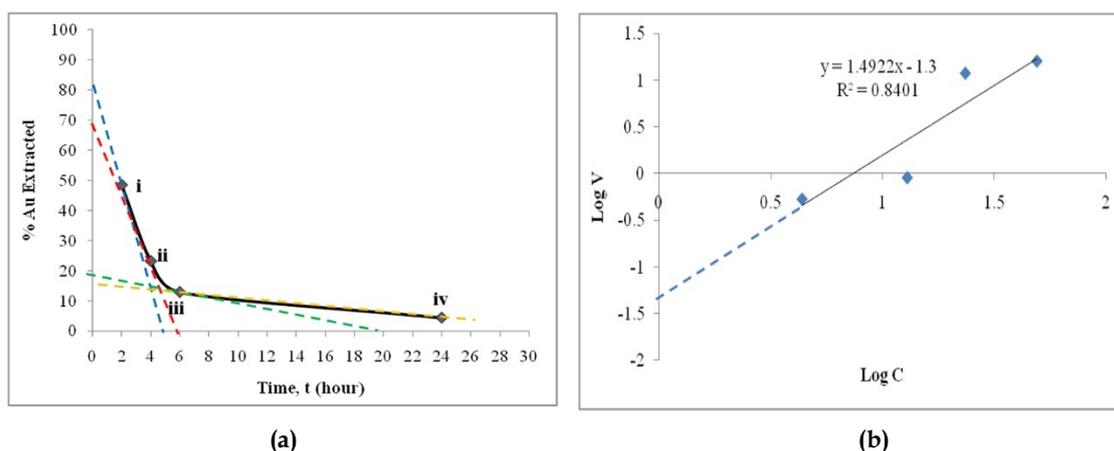


Fig. 12. (a) the percentage of gold extracted at the optimum condition with respect to time, and (b) the rate of gold extracted (V) with respect to the percentage of gold extracted

#### 4. Conclusions

The statistical analysis of gold extraction from mesothermal lode gold ore via the cyanidation process in presence of  $H_2O_2$  and  $Pb(NO_3)_2$  was performed to identify the optimum condition for gold recovery. The characterization study indicated that the gold particles interlocked in sulphide minerals and aluminosilicates minerals. Moreover, the XRD spectrum and elemental composition analysis revealed that the major elements were Si and Al in the form of aluminosilicates minerals. The statistical approach using the response surface design–central composite design suggested a first order linear model between the experimental parameters and the gold extraction percentage. The optimization of the derived model showed that the maximum gold recovery of 88.97% (1.72 g/t Au) was obtained at 467.3 ppm NaCN, 94.96 ppm  $Pb(NO_3)_2$ , and 340 ppm  $H_2O_2$ . The reaction order was claimed to be of second order with a specific reaction rate of 0.0501/hour using Van't Hoff differential method.

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