Optimisation of the Leaching Parameters of a Gold Ore in Sodium Cyanide Solution

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Abstract: **The objective of the work reported in this paper is to propose ways of improving the recovery on a low grade gold ore. To date, no work on optimizing this particular ore has been done in order to improve the recovery. Five size fractions used in the leaching tests reveal that recovery is a function of operational parameters. The optimum values for these parameters that gave a maximum recovery were: particle size (-150 + 75 μm) concentration of the lixiviant (210 ppm), temperature (85°C) and mixing speed (900 rpm). We then applied the shrinking core model and ash layer diffusion is the slowest step. An empirical model was then proposed to predict the leaching behaviour of the gold ore. A value of 12, 9 kJ /mol was obtained for Activation Energy (E$_a$) which is consistent with some results from leading researchers.**

Keywords: **Gold, Cyanidation, Process Optimization, Shrinking core Model.**

1. INTRODUCTION

The present trend in the processing of minerals is the processing of low grade ores as the high grade ones become exhausted [Chenje et al., 2003]. The effective liberation of values from these low grade ores remains one of the major challenges for mineral processors, hence an interesting area of research [Danha et al., 2015]. In South Africa, the depletion of high grade gold reserves has resulted in the increase of the price for this metal mainly due to the high operational costs encountered in processing the available low grade reserves. The feasibility challenges in the gold sector, offer a strong incentive for researchers to propose efficient techniques that can be employed to commercially recover as much of the gold as possible.

Ball milling is an important unit operation for size reduction in the mining industry. It is the mostly used unit operation and is an energy inefficient process. In the recent years, attention has generally been on ways of effectively using energy during the operation. The grinding process requires a lot of energy to break up material to smaller particles. Over-grinding must be avoided as this only wastes energy and to some extent can negatively impact downstream processes. Several researchers have therefore come up with ways of improving energy utilisation based on plant experience and empirical modelling which is a major drawback.

One of the primary reasons for milling materials down to smaller particle sizes is to unlock the value material in the host matrix. Once the material is small enough, and value material has been liberated, a downstream processes such as leaching or flotation can be used as recovery unit operation. It must therefore be understood that mill products should be tailored to satisfy leaching requirements. For this to successfully happen, milling has to be carefully studied to enable the determination of the trade-off between itself and leaching.

Rademan and Groot, [2012] carried out studies on the leaching of this Far West Rand gold ore with the aim of comparing the leaching kinetics of the reef, that of the surface gold ore and a mixture of the two. Their main objective was to

minimize processing costs; risks associated with lack of safe practise and losses incurred during the transference of ores to the central point of treatment.

The work reported in this paper is on the laboratory scale experiments we carried out on a low grade gold ore, in which our objective was to propose ways of improving the gold recovery. This investigation intends to continue from where Rademan and Groot, (2012) left and add value to the understanding of the leaching kinetics as well as determine optimum conditions required for the processing of this ore.
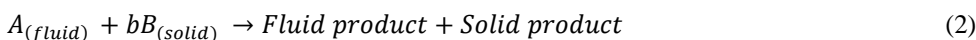
2. LEACHING

The leaching reaction of gold in a solution of sodium cyanide (NaCN) can be represented as follows:

$$4Au + 8CN^- + O_2 + 2H_2O \rightarrow 4Au(CN)_2^- + 4OH^- \qquad (1)$$
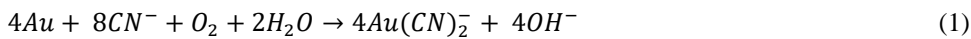
When performing the leaching process, the cyanide solution must penetrate through the ore. A number of steps can occur during the process and these are summarised as follows:

1. Diffusion of the NaCN solution from solution through the diffusion layer (Δx) to the gold ore surface layer

2. Adsorption of the NaCN on the solid;

3. Reaction of the lixiviant with the gold to form a soluble metal species;

4. Desorption of the formed complex from the gold ore surface; and

5. Diffusion of the complex through the ash layer (Δx) away from the gold surface.

The ash layer does not dissolve into the NaCN, but the reacting core (of gold) shrinks although the particle size remains constant, hence the 'shrinking core' model [Demirkiran, 2008; Koech et al.,, 2015] The model has been widely used to predict the kinetics of mineral dissolution so that researches can make comparative analyses of the dissolution kinetics of minerals. The general equation is represented as

$$A_{(fluid)} + bB_{(solid)} \rightarrow Fluid\ product + Solid\ product \qquad (2)$$

Where: A represents the solvent, B represents soluble component undergoing leaching, and b the stoichiometric coefficients of B.

It is noted that the slowest step from the listed 5 steps becomes the rate controlling or limiting step. A reaction can therefore be chemical reaction, product or ash layer diffusion or film diffusion controlled. When diffusion through the product layer controls the kinetics, the equation is given by Levenspiel [1972] as:

$$1 + 2(1 - X) - 3(1 - X)^{\frac{2}{3}} = \frac{6aMDC}{\rho r_0^2} t = k_d t \qquad (3)$$

For chemical reaction, equation (4) applies:

$$1 - (1 - X)^{\frac{1}{3}} = \frac{k_c MC}{\rho a r_o} t = k_r t \qquad (4)$$

When diffusion through the liquid film controls the reaction, the equation is given as:

$$X = \frac{3bk_g C}{\rho r_o} t = k_m t \qquad (5)$$

where: k_r, k_m and k_d = chemical rate constants (cm/ min);

X is the fraction of gold recovered in time t, D is the diffusion coefficient (m^2/min), k_c and k_g are kinetic constants, M the molecular mass , C the concentration NaCN, a is the stoichiometric coefficient of the reagent, ρ gives density of the solid, r_o the initial radius, and k the rate constant.

An Arrhenius equation is applied to calculate temperature dependence of the reaction rate constant. The expession is given as follows:

$$k = A \exp[\frac{-Ea}{RT}] \qquad (6)$$

where R is the universal gas constant, A is the frequency factor, E_a is the activation energy, and T is the absolute temperature.

A graph of ln k against 1/T gives the E_a for a leaching reaction. According to Habashi [1967] the activation enegy of an ash layer process is considered to be in the 1 - 3 kcal/mol range, while that of a chemically controlled process is generally more than 10 kcal/mol. Other researchers such as Levenspiel [1972]; Maina and Mbarawa, [2011a]; Limo and Enweremadu, [2011] have suggested that when E_a is less than 20 kJ/mol, ash layer diffusion is normally the rate controlling step.

3. EXPERIMENTAL

3.1 Material and methods:

A run-of-mine sample of gold ore is ground in the batch ball mill. The resultant gold-bearing ore is then sieved through a dry screen to separate the particles into size classes, as shown in Table 5. The size range normally used for leaching is ≤75 microns, but we also experimented with the finer and coarser size classes in order to develop a broader context within which to understand the process.

An ore sample (from each size class) weighing 200g was poured into the reaction vessel (fitted with a mechanical stirrer as illustrated in (Fig. 1), and 200ml of water was added to make slurry. The pH of the sample was then adjusted by adding lime (CaO), maintaining a pH value of ~12. The lime was added to keep the pH of the pulp above 11 in order to avert the danger of producing poisonous HCN. The leachant (NaCN) was then added according to the set parameters to start the leaching process.
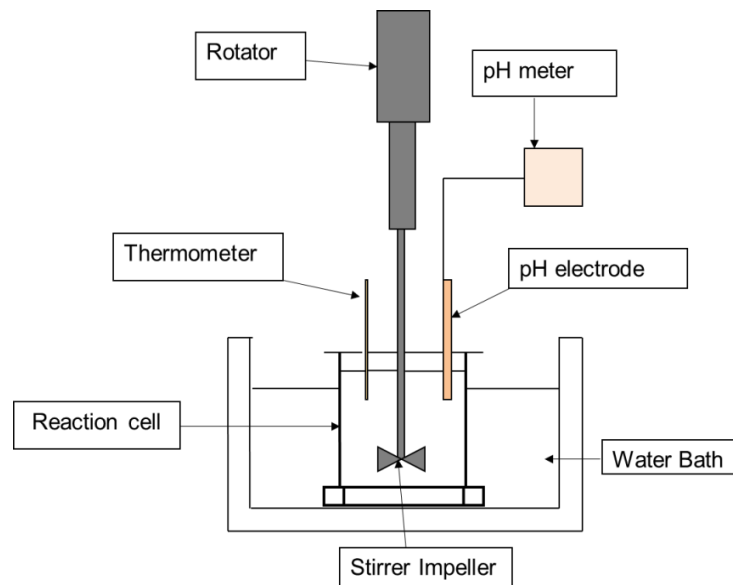

Fig. 1: Schematic diagram of equipment set-up for the gold leaching process.

A reactor containing the ore sample, distilled water and lime was conditioned for one hour before adding the calculated amounts of sodium cyanide. This was done at the start of each leach test and tests are carried out for one, two, three, five and 24 hours, on material from each of the chosen size classes. After the completion of each leaching run, the samples were filtered through a Buchner funnel to remove the leach liquor. The leach liquor was re-filtered on a Millipore filter before being taken for gold analysis, using an Atomic Adsorption Spectrometer. The same procedure was then repeated for all the samples that were being investigated. After analysing the filtrate for gold, the slurry was washed two to three times with distilled water. Thereafter the solid was dried and sent for gold analysis.

Table 1: Leaching test parameters

PARAMETER	VALUE	UNIT
Liquid to solids ratio	1:1	by mass
Dissolution period	24	hrs
Lime addition	1	kg/t CaO
pH	11	
Cyanide addition	0.3	kg/t NaCN
Grind	80% passing 75 micron	
Head Grade	Variable as per sample	

Table 2: Leaching conditions

PARAMETER	CONCENTRATION (PPM)	TEMPERATURE (^{0}C)	AGITATION SPEED (RPM)	PARTICLE SIZE (MICRONS)
Fixed value	210	25	180	-150+75µm
Other values	30, 75, 400 and 500	40, 60, 80, 100 and 120	360, 720, 900 and 990	Other 5 classes

Table 3: Particle size classes used for the leaching experiments

Size class	Upper (microns)	Lower (microns)
M1	-1700	+850
M2	-850	+300
M3	-300	+150
M4	-150	+75
M5	-75	+25
M6	-25	to pan

4. RESULTS

4.1 Dissolution characteristics:

In this section, we will use the experimental results to analyse the effects of particle size; mixing; sodium cyanide concentration and temperature on the dissolution characteristics of a gold ore. Based on our results, we will specify the optimum operational parameters that gave us a maximum gold recovery.
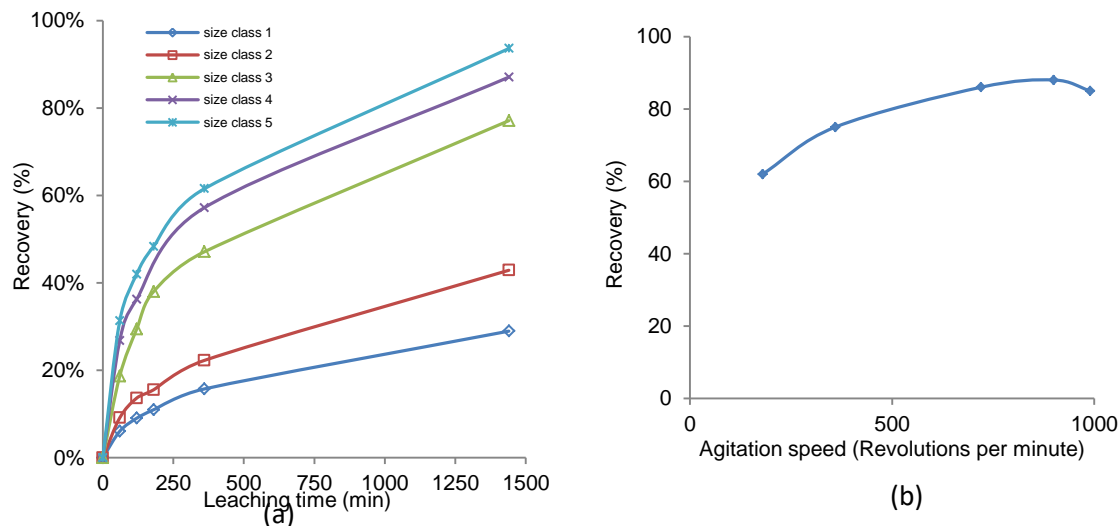

Fig. 2: (a) Gold recovery versus leaching time for different particle size classes. (b) Gold recovery versus agitation speed

We investigated the influence of particle size on the dissolution of the ore using the sample size fractions shown in Table 5. Fig. 2(a) shows the gold recovery results we obtained by varying the leaching time for different particle size classes. The experimental conditions employed were a fixed sodium cyanide concentration of 210 ppm, an agitation speed of 720 rpm and a temperature of 25^0C. The recovery calculations were based on the individual size class head assays and the gold content of the leach liquor at the end of each leach interval.

Fig. 2(a) reveals that particle radii have an effect on the dissolution of a gold ore. As shown by the results, a decrease in particle size increases the fraction of gold we recover in the liquor. This is because the smaller particles provide a larger contact surface area for the leaching solution. They also reduce the penetration distance required by the cyanide to reach the soluble gold within the particle. On the other hand milling to finer sizes tends to increase cyanide and energy consumption and though coupled with improved recovery, cost wise it might not be economic.

Fig. 2(b) on the other hand shows the influence of agitation speed on the gold recovery. For this test, we used material from the $-150 + 75$ µm size class, in a 210 ppm sodium cyanide solution and at a temperature of 25 ^{o}C. The graph reveals that the gold recovery increases with agitation speed. However, there is an optimum speed (900 rpm) that gives a maximum recovery. Further increasing the mixing speed beyond this optimum does not translate to an increase in the

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recovery. During agitation all the particles in the slurry should be sufficiently suspended to be in good contact with a fresh solution. We can see from Fig. 2(b) that on increasing the agitation speed one enhances the dissolution until external mixing is no longer a rate limiting step. We can thus infer that heavy agitation tends to reduce the thickness of the external diffusion layer, thereby improving the rate of transfer of oxygen and sodium cyanide to the particle. This also allows for the saturation of the slurry mixture with dissolved oxygen hence achieving high recoveries. At high agitation rates we waste energy without dissolving more oxygen and hence gold dissolution is not noticeable.

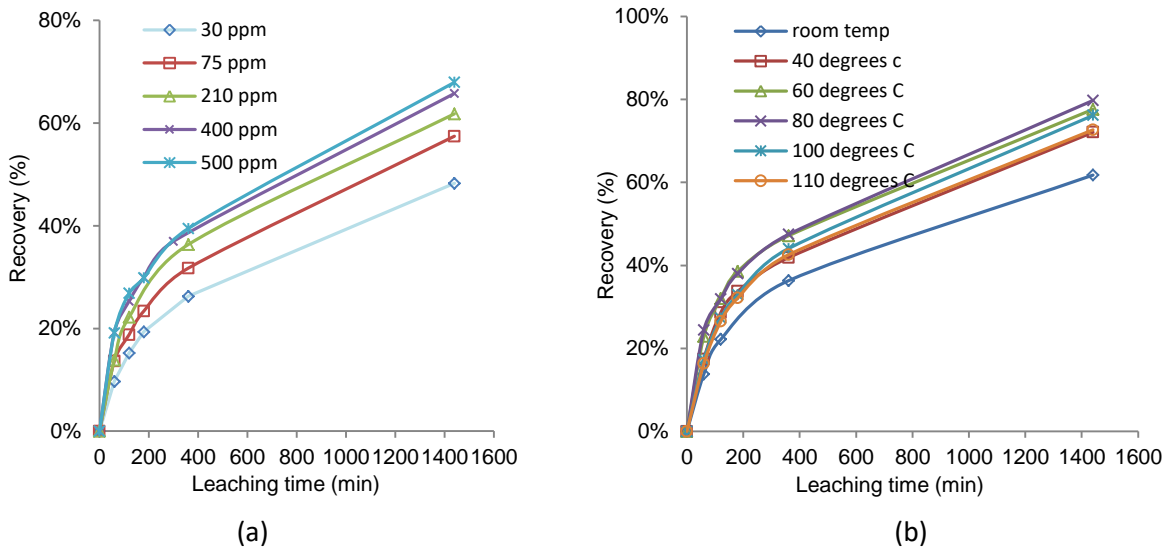(a)   (b)

Fig. 3: (a) Recovery versus leaching time for different concentrations of sodium cyanide (b) Recovery versus leaching time for different experimental temperatures

Fig 3(a) demonstrates the influence of sodium cyanide concentration on the dissolution of the gold ore, determined by evaluating the recoveries achieved over a period of leaching time, using different concentrations of the lixiviant. This set of experiments was also carried out at room temperature, and on a single size fraction (-150 + 75 μm).

The results graphically represented in Fig 3(a) show that raising the cyanide concentration improves the recovery, especially between 30 and 210 ppm while increasing the concentration further from 400 to 500 ppm did not improve the recovery much. Employing excess cyanide is accompanied by a high lixiviant usage and this has no favourable effects on gold recovery. All subsequent experiments were thus done at a cyanide concentration of 210 ppm.

Fig 3(b) shows results from a set of experiments conducted and aimed at investigating the effect of temperature on dissolution. A cyanide concentration of 210 ppm; -150 + 75 μm size class material; 720 rpm mixing speed and a temperature of 25°C were employed as the experimental conditions. Fig 4 reveals that generally the rate and the fraction of gold dissolved increases as the temperature increases but then falls off at higher temperatures. It is expected that a higher temperature will result in a higher rate of reaction because the viscosity of slurry is lowered by an increase in temperature and the rates of mass transfer are raised. The reaction rate is therefore expected to increase when temperature is raised.
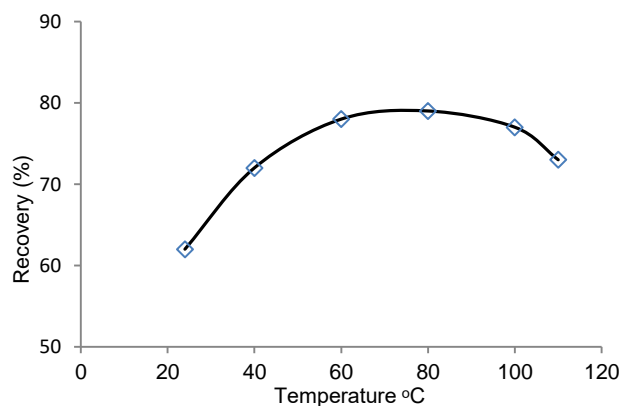

Fig. 4: Amount of gold recovered at different temperatures

Fig 4 shows that the maximum recovery of gold is obtained at ~ 80°C. It is noted that above this temperature the recovery starts decreasing and this can probably be attributed to the gas (oxygen) solubility which tends to decrease with increasing temperature.
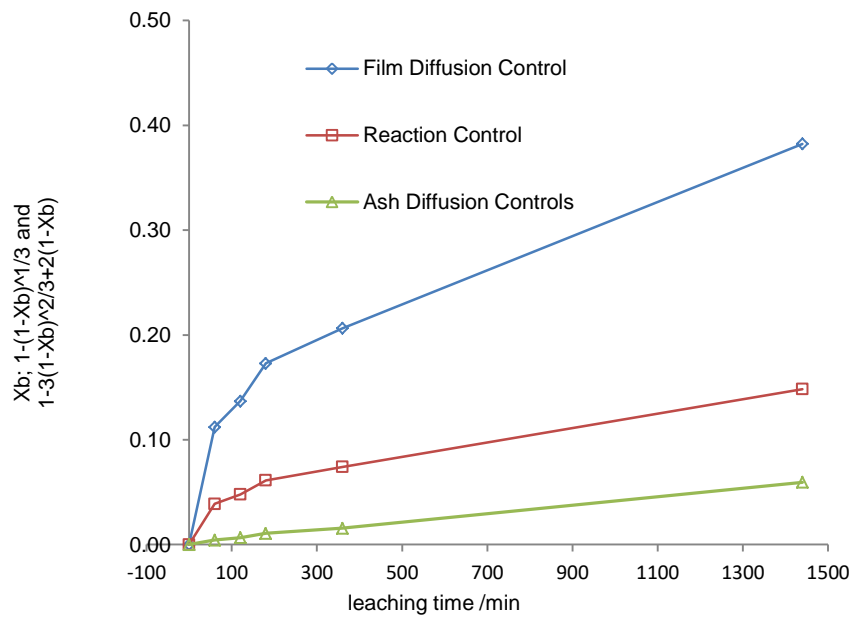

Figure 5: Variation of different shrinking core models with leaching time

Fig. 5 shows the variation of different shrinking core models with leaching time. For this set of experiments; material from the - 300 + 150 µm size class; stirring speed of 900 rpm and a reaction temperature of 25 °C were employed as the reaction conditions. To obtain Fig. 5, the left hand sides of Equations 3, 4 and 5 were plotted against leaching time. Fig. 5 reveals that the ash diffusion control model gives a straight line, which proves that it is the rate limiting step of the dissolution reaction.

Fig. 6 shows the results obtained when the ash diffusion model is applied to all the size classes, and all test conditions used in the experiment, over different leaching periods of time. A linear relationship is obtained which proves that indeed, the diffusion through the ash layer is the rate controlling step.

(a)

(b)

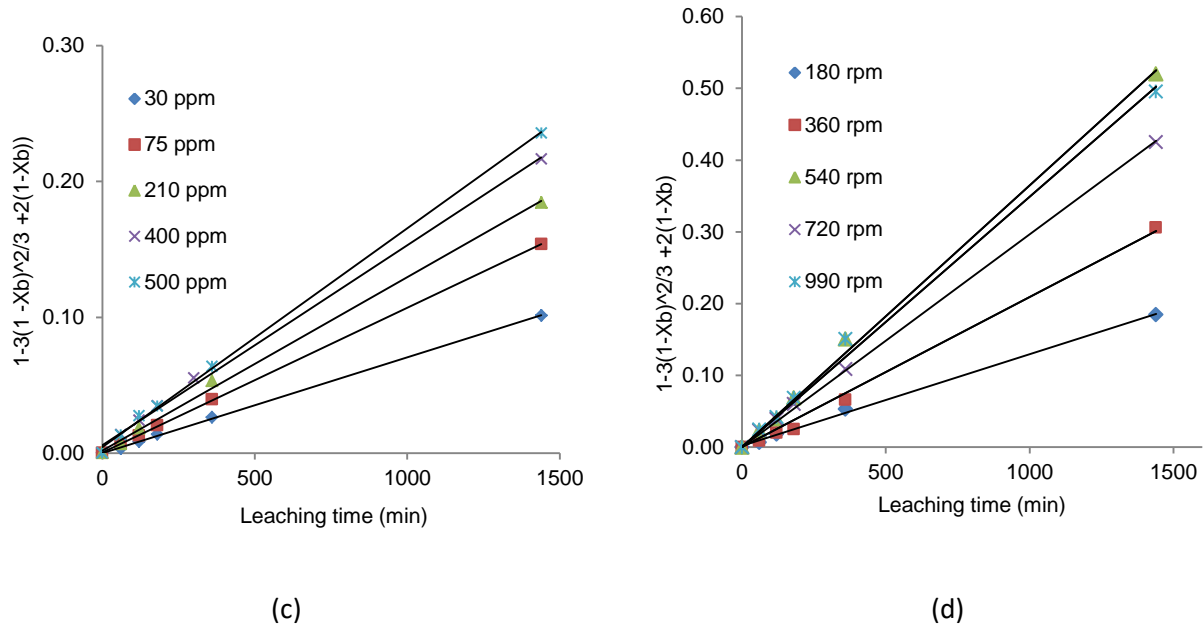(c)   (d)
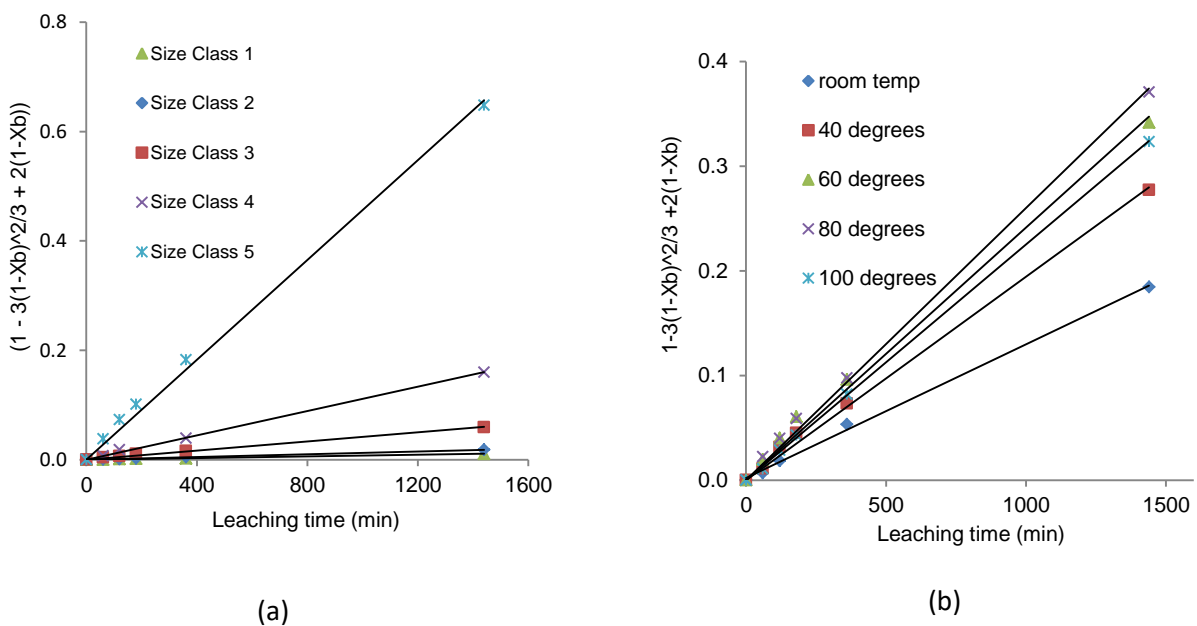
Fig. 6: Plot of the ash diffusion model against the leaching time required for gold dissolution applied to different (a) particle sizes (b) temperatures (c) concentrations (d) agitation speed

The rate constants were then determined from the gradient of the straight lines obtained using different specified conditions from Fig. 6. The values obtained are given with their corresponding correlation coefficients in Table 4.

The next step was to propose an empirical model that will predict leaching of the Far West gold ore. The model can be expressed as:

$$1 + 2(1-X) - 3(1-X)^{\frac{2}{3}} = K_0 A^a D^b C^c e^{\frac{-E_a}{RT}} t \qquad (13)$$

where A is speed of agitation, D is size of particles and C is NaCN concentration.

Each investigated parameter has a reaction order which is denoted by constants a, b and c (Equation 7). We obtain their corresponding values by plotting the natural logarithm of the rate constants versus natural logarithm of their respective parameter values. Fig. 7 shows the plots obtained for different particle sizes, speeds of agitation, temperature and NaCN concentrations.
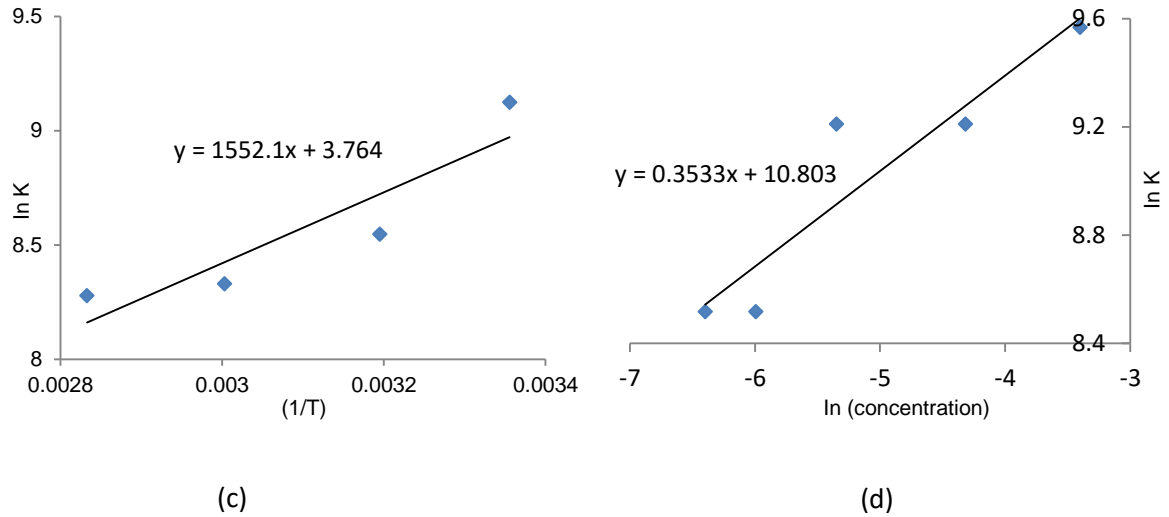

(a)   (b)

(c)

(d)
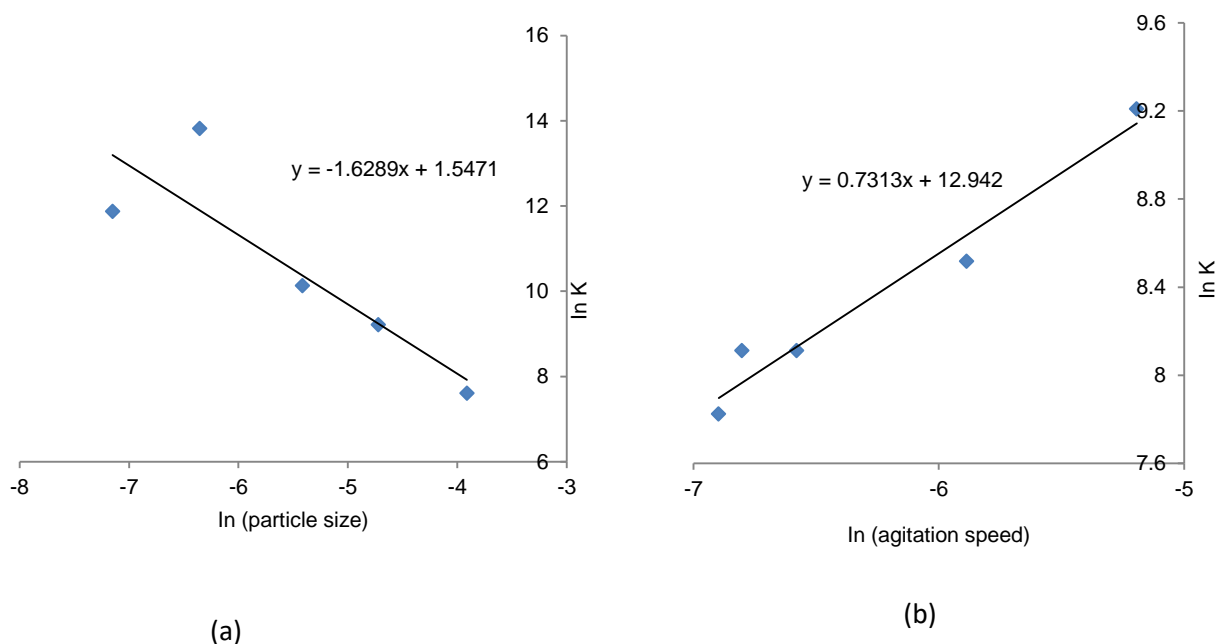
Fig. 7: Plot of the –In K against (a) -In (particle size) (b) –In (agitation speed) (c) –In (1/T) (d) –In (concentration)

The proposed semi empirical model is written as:

$$1 + 2(1-X) - 3(1-X)^{\frac{2}{3}} = K_0 A^{0.7313} D^{-1.6289} C^{0.3533} e^{\frac{-E_a}{RT}} t \qquad (14)$$

Activation Energy (E_a)
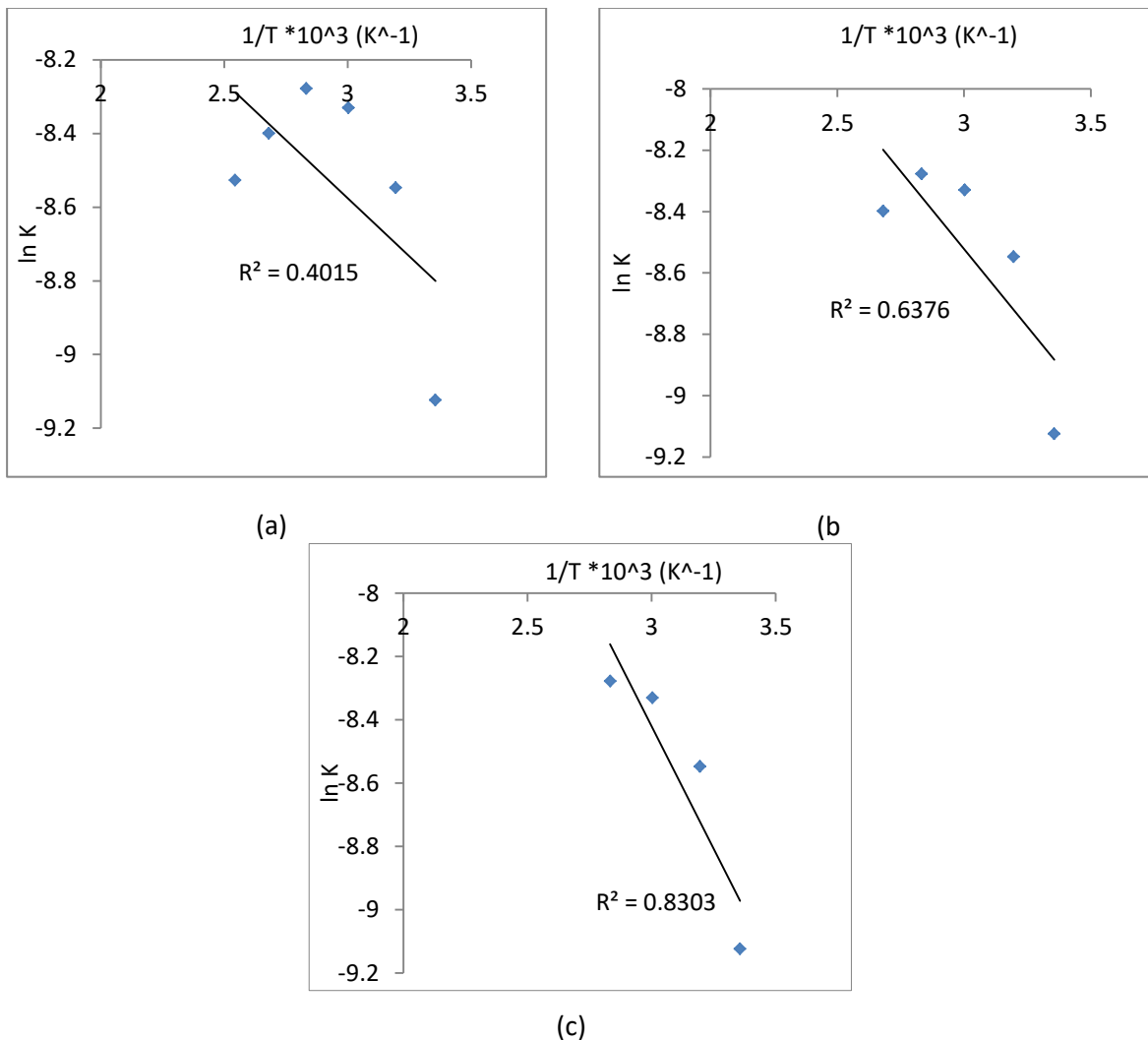

(a)

(b

(c)

Figure 8: Arrhenius plots for gold dissolution in 210ppm NaCN

The apparent rate constant derived from the slopes of the line in Fig. 8 (c) (table were used to obtain the Arrhenius relation. Due to the unusual trend on the influence of temperature on dissolution of the gold, the activation energy is only calculated for the initial four temperatures.

The Arrhenius graph of ln k against 1/T depicted by Figure 8 (c) gives an activation energy of 3,09 kcal/ mol or 12,9 kJ/mol for the reaction. As discussed earlier in section 2.2, this activation energy satisfies the ash layer process condition. Fig. 11 is a linearization of the kinetic curves done by use of Equation 9. The rate constant values are the slopes of the straight lines in Fig. 9. Plots of rate constant values for particle versus the reciprocal of the square of the particle radii ($\frac{1}{r_0^2}$) yielded a linear relationship with a correlation coefficient of 0.9901 and is shown on Fig. 11.
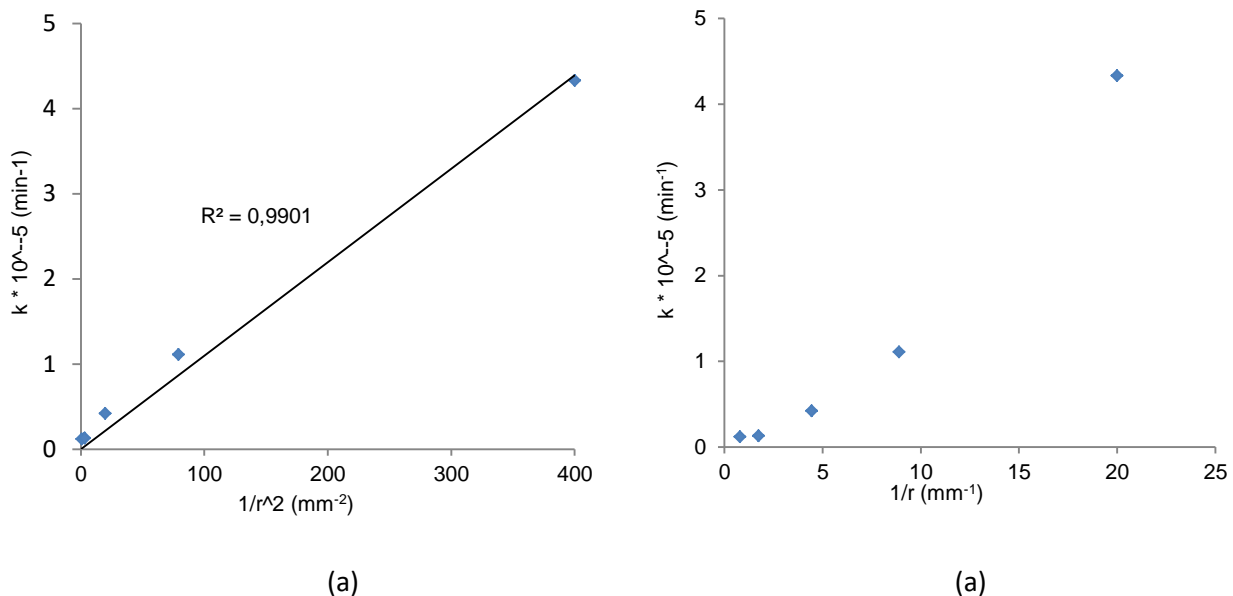

(a)   (a)

Fig. 11: Variation of rate constant k and (a) $\frac{1}{r_0^2}$ (b) $\frac{1}{r_0}$ for the leaching system

Results show that there is a straight line relationship between k (rate constant) and the reciprocal of the square of the particle radii. This is further evidence to confirm that the ash diffusion is the slowest step of the reaction. On the other hand a plot of k against the reciprocal of the particle radii (a case for chemical reaction controlling) does not give a linear relationship.

5. CONCLUSION

Leaching experiments were conducted on Far West gold ore. Leaching parameters; temperature, particle size, degree of agitation and concentration of the lixiviant showed noteworthy effect on rate of dissolution of gold. The optimum values for these parameters that gave a maximum recovery were: particle size (-150 + 75 μm) concentration of the lixiviant (210 ppm), temperature (85°C) and mixing speed (900 rpm). The dissolution followed shrinking core model with ash diffusion as the rate limiting step. Variation of k and $1/r^2$ showed linear relationship and that further confirmed the ash diffusion model. An empirical model was proposed to predict the leaching of the gold ore and can be written as:

$$1 + 2(1 - X) - 3(1 - X)^{\frac{2}{3}} = K_0 A^{0.7313} D^{-1.6289} C^{0.3533} e^{\frac{-E_a}{RT}} t$$

The E_a of 12, 9 kJ/mol was determined which is consistent with some results from leading researchers.

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APPENDIX-A

Table 4: Diffusion rate constant k for the ash diffusion control model with the correlated coefficients for gold ore dissolution

Variable	Rate constant	Correlation coeffient
Temperature (K)		
298	0.000109	0.9977
313	0.000194	0.9965
333	0.000241	0.9998
353	0.000254	0.9930
373	0.000225	0.9969
393	0.0001.98	0.9989
Concentration (ppm)		
30	0.00007	0.9996
75	0.0001	0.9999
210	0.0001	0.9977
400	0.0002	0.9936
600	0.0002	0.9970
Agitation Speed		
180	0.0001	0.9977
360	0.0002	0.9955
720	0.0003	0.9992
900	0.0003	0.9958
990	0.0004	0.9978
Mean particle size		
1275	0.000007	0.996
575	0.000001	0.9991
225	0.00004	0.9924
112.5	0.0001	0.9985
50	0.0005	0.9956