CHARACTERISATION AND DIAGNOSTIC LEACHING OF GOLD-BEARING MINERAL ORE, EAST COAST PENINSULAR MALAYSIA

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Abstract. This study reports the refractory characteristic of newly found gold ore deposits from the East-Cost of Peninsular Malaysia. The diagnostic leaching was conducted on the concentrated samples to assess the refractoriness of the ore. The bulk, concentrated, and tailing samples were characterised by mineral phase and chemical composition analysis. The surface morphology also was studied using microstructural analysis. As a result, a few significant elements were found in the sample, with the highest is silicon, followed by iron, aluminium, potassium, sulfur and arsenic. For mineral phase analysis, the dominant mineral in the sample is quartz, followed by other gangue minerals such as pyrite, arsenopyrite and magnetite. The direct cyanidation at the first step gives 38% of gold recovery, indicating the gold associated with oxide. In contrast, the second higher of gold recovery was 31.7% after the leaching of sulphide minerals. Hence, most gold is associated with sulphide minerals such as pyrite and arsenopyrite due to significant sulfur content and gold associated with iron. This ore can be classified as a refractory ore from diagnostic leaching that needs a pretreatment process before cyanide leaching for gold extraction.

Keywords: gold ore, diagnostic leaching, gold processing, cyanide leaching, refractory ore

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Article Info

Received 29th September 2021
Accepted 1st December 2021
Published 20th December 2021

ISSN: 1823-7010, eISSN: 2600-7444

Introduction

Gold is a valuable and malleable metal. Gold can be found in ore as a pure metal, as an alloy with other metals such as silver (electrum), or as a mineral in rare cases [1]. Gold particles in sulphide minerals such as pyrite and arsenopyrite can be coarse and liberated through grinding, or finely disseminated sub-microscopic particles (often referred to as solid solution or refractory gold) [2]. The most important minerals associated with gold are pyrite (FeS_2), galena (PbS), sphalerite (ZnS), arsenopyrite (FeAsS), stibnite (Sb_2S_3), pyrrhotite (Fe(1-x)S), and chalcopyrite ($CuFeS_2$). Magnetite (Fe_3O_4) and various selenium minerals may also be present [1-2].

Cyanidation has been widely used in the gold extraction process. This metallurgical technique would form a stable gold-cyanide complex ion and be able to leach gold from low-grade ores [1]. The reaction of gold with cyanide represented by the following reaction in Equation 1.

$$2Au + 4NaCN + ½\,O_2 + H_2O + 2OH^{-} \rightarrow 2Au(CN)^{-2} + 4NaOH \qquad (1)$$

However, cyanide leaching is ineffective in extracting gold from refractory and carbonaceous ore [1]. The challenges associated with gold extraction involve various types of refractory gold ore. The ore is refractory if the gold recovery through cyanidation is less than 80% [3]. Carbon, sulphide, and tellurium ores are the three types of gold refractory ores [4]. Gold with physical encapsulation is the most common refractory ore. This is due to gold that is interlocked in sulphide minerals or bound with preg-robbing minerals. The preg-robbing gold binds to organic carbon and carbonaceous matter, which adsorbs the leached gold during cyanidation. It would adsorb the aurocyanide ($Au(CN)^{2-}$) during the cyanidation process and result in poor gold recoveries. The presence of slow-dissolving gold minerals such as aurostibite ($AuSb_2$), maldonite (Au_2Bi), and gold tellurides makes antimony, bismuth sulphide, and telluride gold ores somewhat refractory [2]. This ore type will cause the leaching kinetics to slow and the gold recovery to be lower. Other refractory gold ores are associated with minerals that consume a large amount of oxygen, cyanide, and lime, such as pyrrhotite, marcasite, enargite, and arsenopyrite [5]. As a result, these types of gold ore must first be pre-treated to overcome refractoriness and make the gold accessible to the cyanide leaching for gold recovery. For complex sulphide gold ore, suitable pretreatment processes such as ultrafine grinding, modified cyanidation, roasting, pressure oxidation, and bacterial oxidation are required to destroy the sulphide matrix and liberate the gold [5]. Other pretreatment processes for different types of refractory ores that have been are reported elsewhere [4, 6].

Based on the problem highlighted on cyanidation and refractoriness of the gold ore, diagnostic leaching was studied to distinguish the nature of refractory gold ores before extracting gold. The diagnostic leaching study involves a selective oxidative acid leach for eliminating minerals (from least to most stable in the sample matrix). Each stage is followed by cyanidation of the residue to recover liberated gold. Mineralogical studies are essential in determining the deportment of gold minerals and other associated gangue minerals to design the diagnostic leaching experiment using Lorenzen's guidelines [7] as a reference, as shown in Table 1. The leaching study provides information on the mode of occurrence of gold, where gold is associated with other minerals in varying degrees of liberation, resulting in the refractoriness of the ore. However, there is some overlap in the extent of leaching where it may dissolve several minerals. Nonetheless, it is generally limited to 90% of the required mineral and 10% of the following most stable mineral [4]. This overlapping minerals problem, according to Henley et al. [8], can be minimised by analysing the leached solution for the elements features of the minerals and using mineralogical knowledge to estimate the gold distribution among the minerals.

For every diagnostic leaching, the pretreatment stages used are specifically selected for that particular ore. This is because the different mineralogy of the ore reacted to the acid differently. A detailed understanding of the mineralogy is needed before designing a diagnostic leaching procedure. Based on the previous application of diagnostic leaching, all the pretreatment stages started with cyanide leach, where the free gold was removed first [9-14]. Leachable gold is gold that can be leached without any problem. Usually, leachable gold is the highest portion out of all others. The following acid that was used is different based on the different mineralogy of the ore. Anggraini [11-14] used sodium hydroxide (NaOH) as the next pretreatment stage to liberate the gold in preg-robbing material. In other procedures, HCl is used to attack the gold that disseminates in calcite and dolomite [9-12]. Celeb and Serbest [13] used potassium hydroxide (KOH) in their diagnostic leaching test design to the ore containing gold associated with the iron oxide mineral phase. This mineral phase can be leached in an alkaline solution. KOH was needed to dissolve the iron oxide mineral phase first. This alkaline leaching is a new approach in diagnostic leaching, and it significantly increases the recovery. KOH as a pretreatment stage in diagnostic leaching, exposes the refractoriness of iron oxy/hydroxide carrying gold/silver ore and a pretreatment strategy to overcome the refractoriness, according to the authors.

Malaysia has a long history of widespread small-scale gold mining throughout the country, particularly in Peninsular Malaysia's Central Belt, a high-potential region for the gold mining industry. The Central "Gold" Belt is a 20km wide, major N-S trend of gold mining districts that demonstrates the critical role of hydrothermal fluids in the development of gold in Peninsular Malaysia, particularly in North Pahang and Kelantan [1]. A review of the literature on the geology of old deposits mineralisation is commonly associated with hydrothermal quartz vein systems, skarn, and volcanogenic massive sulphides. Malaysia, particularly the East Coast of Malaysia, has produced gold over million tonnes, primarily from alluvial and high-grade hard rock deposits. However, as high-grade deposits have become scarce, the need to mine low-grade gold deposits and refractory ore has gained prominence. In this study, characterisation and diagnostic leaching were performed on a gold deposit sample from the East Coast of Peninsular Malaysia to determine the refractoriness of the sample and suggest the possible pretreatment process for gold leachability.

Table 1. Selective pretreatment leach stages and the minerals dissolved.

No.	Pretreatment stage	Minerals likely to be destroyed	References
1.	NaCN washes	Precipitated gold	[7]
2.	NaCN	Gold	[7]
	KOH	Iron oxy/hydroxide	[10][12][15]
3.	Na_2CO_3	Gypsum and Arsenates	[7]
4.	Hydrochloric Acid	Pyrrhotite, Calcite, Dolomite, Galena, Goethite, Calcium Carbonate	[4][12][3][7]
5.	$HCl/SnCl_2$	Calcine, Haematite, Ferrites	[4][7]
6.	Sulfuric Acid	Uraninite, Sphalerite, Labile Copper Sulphides, Labile Base Metal Sulphides, Labile Pyrite	[4][7]
7.	$FeCl_3$	Sphalerite, Galena, Labile Sulphides, Tetrahedrite, Sulphide Concentrates	[4][7]
8.	Nitric Acid	Pyrite, Arsenopyrite, Marcasite	[4][7]
9.	Oxalic Acid washes	Oxide Coating	[4][7]
10.	HF	Silicates	[4] [7]
11.	Acetonitrile elution	Gold adsorbed on Carbon, Kerogen, Coal	[4][7]

Materials and Methods

Sample preparation. The samples were collected from the new site located at Pahang, Malaysia. At the site, the grab sampling method was used to collect the representative samples. About 11 points were chosen for samples collecting location. The samples acquired from each point were about 3 kg, so the overall samples collected were about 40 kg on average. The ore was sampled to obtain the homogeneity and desired sample size for analysis. A riffled portion with particle size around 100 to 150 μm was pre-concentrated by the gravity separation method and produced three types of concentrate, middling, and tailing. In this study, all pre-concentrated samples were then undergone a further grinding process until 80% passing 75 μm and subjected to the physical and chemical characterisation analysis. The ore grade analysis was conducted on concentrate samples by fire assay technique and cyanidation test.

Characterisation of samples. For chemical composition analysis, bulk samples were analysed using X-ray fluorescence (XRF, Rigaku RIX 3000). All samples, including residue samples after diagnostic leaching, were analysed to determine the mineral phases using X-ray diffraction analysis (XRD, Bruker D8 Advanced XRD) with a Cu-anode and Kα radiation in the 2θ angle range of 10^o -90 ^{o}. All minerals were identified with PANalytical X'Pert HighScore Plus software (v2.2e) and the PANalytical ICSD database.

The morphological of gold ore samples were studied using the optical microscope (OM, Olympus BX41) and field emission scanning electron microscopy with energy dispersive X-ray analysis (FESEM/EDX, Zeiss Supra 35VP-24- 58). First, the samples must be coated with sputter coating with platinum to cover a specimen with a layer of conducting materials. The coating process takes about an hour to finish. Then, the samples are observed and imaged at a different working distance and accelerating voltage to produce Backscattered Images (BSI) for each sample. FESEM used to locate the image in an ore microscope while the possible minerals are spotted with EDX to determine the composition of the samples [5,11].

Fire assay analysis. A fire assay analysis is a quantitative determination that uses fusion processes to separate metals from impurities. To determine the amount present in the original sample, the sample was weighed and mixed with a flux containing soda ash, borax, litharge, silica, and nitrate [16]. Before firing at temperatures ranging from 1000 ºC to 1200 ºC, the mixture must be homogeneous. The hot molten metal was then poured into the mould. When the molten cold was reached, the lead button was separated from its slag and subjected to the cupellation process. It is then digested with aqua-regia, and the concentration is determined using inductive coupled plasma-based analysis (ICP-OES). However, accuracy in sampling, preparation, assaying, handling and weighing is critical in fire assay. If done incorrectly, errors in the 0.0001 g final gold bead can have astronomical proportions.

Cyanidation test. The cyanidation test aims to determine the recovery of the gold and the amount of free gold content in the sample that can be leached by cyanide. Cyanidation is a process that only leaches out the free gold in the form of Au^{2+}. The sample was finely ground until 80% passing 75µm, to ensure that the gold was liberated and able to leach by the cyanide in the bottle roll leaching. About 50 g of bulk sample with 150 ml of cyanide solution (using 1:3 ratio) was mixed in the plastic bottle while adding lime to ensure the solution pH was maintained at pH 11. Once the pH was stabilised at 11, the slurry was transferred to a $1dm^3$ Nalgene roll bottle, and the lid was sealed using masking tape to prevent it from leaking. The bottle was then placed on a bottle roller at a rotation of 50 rpm for 24 hours. After 24 hours, the sludge was filtered using a vacuum pump using filter paper No. 1 (Whatman, Sigma-Aldrich), and the pregnant solution was obtained. The solvent extraction procedure was carried out using DIBK, KCl and aliquot 336 to purify gold solution, and the organic solution was analysed using an ICP-OES machine.

Diagnostic Leaching Method. Diagnostic leaching is a type of analysis that can help determine which minerals are associated with gold. To determine which minerals are associated with the desired precious metal, a specific mineral is first eliminated using a selective oxidative leach. The gold liberated by the leaching of this mineral is extracted using cyanide leaching. First, the least stable mineral in the sample matrix is solubilised in an aqueous acid medium. The leaching experiment was carried out in a 1 dm^3 four neck glass reactor equipped with a condenser, thermometer, and a 600 rpm overhead stirrer. Once the solution reached the temperature specified in Table 2, approximately 150 g of sample (for the first step) was charged into the glass reactor. The residue is filtered, washed, cyanided to extract the liberated gold, and then washed with the diluted cyanide solution. A more oxidative acid leach of hydrochloric, sulfuric, and nitric acid was used until all re-cyanided gold was leached, producing an accurate record of the amount of gold associated with each mineral. At the end of the stage, the sample is subjected to a fire assay, and all solutions collected at each stage are analysed using an ICP-OES (Perkin Elmer Optima 4300 DV ICP System).

Table 2. Experimental design and condition of diagnostic leaching for this study (Adapted from Lorenzen [7]).

Steps	**Reagent**	**Leaching conditions**	**Concentration**	**Minerals likely to be eliminated**
Cyanide Leaching I	NaCN	24 h, pH 10.5	1.5 g/L	Free gold
Acid leaching-I	HCl	L/S=2:1, 8 h, 60°C	12% (V/V)	Calcite, dolomite, galena, pyrhotite, hematite
Cyanide Leaching II	NaCN	24 h, pH 10.5	1.5 g/L	Liberated gold
Acid leaching-II	H_2SO_4	L/S=2:1, 5 h,80°C	48% (V/V)	Cu-Zn sulphides, labile pyrite
Cyanide Leaching III	NaCN	24 h, pH10.5	1.5 g/L	Liberated gold
Acid leaching-III	HNO_3	L/S=2:1, 6 h,60°C	33% (V/V)	Pyrite, marcasite arsenopyrite
Cyanide Leaching IV	NaCN	24 h, pH 10.5	1.5 g/L	Liberated gold
Fire Assay	Flux	-	-	Silicates

Results and Discussion

Physical and chemical characterisation . The chemical composition of the bulk sample is shown in Table 3 in terms of major constituents and minor elements. The ore was found to be majorly composed of SiO_2 (55.5 wt%), followed by Al_2O_3 (8.58 wt%) and Fe_2O_3 (22.56 wt%). The presence of gold is indicated by the higher concentrations of SiO_2 and Fe_2O_3 as pathfinder elements. Pathfinder elements are always present in greater concentrations than target elements and originate from the same source or were deposited at the same time and location. In low-sulfidation deposits, gold ore is commonly associated with quartz and feldspar and calcite or sericite as primary gangue minerals. The minor element analysis reveals that arsenic (908ppm), fluorine (1060ppm), and sulphur (1297ppm) are present in significant amounts in the bulk samples. It is also possible that the sample contains arsenopyrite (FeAsS) and pyrrhotite (Fe(1-x)S). Gold also can occur at a different proportion of non-refractory and refractory gold in iron sulphide or arsenic sulphide ores [2]. Figure 1 depicts a diffractogram scan of bulk samples, gravity separation products (concentrated ore, middling, tailing), and diagnostic leaching residues. The analysis of the bulk sample reveals that it is primarily composed of quartz, with minor amounts of magnetite, pyrite, and arsenopyrite. Gold is usually associated with pyrite and arsenopyrite as they are carries for sub-microscopic gold [2]. This finding is consistent with the chemical composition of bulk samples as determined by XRF analysis. While minerals quartz in concentrated and middling samples decreased after gravity separation, other identified gold-associated minerals remained. The gravity separation process successfully separated quartz as gangue mineral into the tailing product, as shown by the XRD diffractogram. The residues after the diagnostic leaching process are also composed of main quartz minerals. Other associate minerals, such as pyrite, magnetite, and arsenopyrite, were not found in tailings or residue samples. According to Coetzee et al. [12], this physical separation process is critical in determining gold's deportment and should be included in the processing route.

Table 3. Chemical composition of gold ore bulk sample.

Major Constituents	Wt. %	Minor Element	ppm
SiO_2	55.5	As	908
Fe_2O_3	22.6	Ce	361
Al_2O_3	8.6	Cl	168
TiO_2	0.3	Cr	325
MnO	0.01	Cu	109
MgO	0.23	F	1060
CaO	0.11	Ni	116
Na_2O	0.05	Rb	217
K_2O	1.84	S	1297
P_2O_5	0.19	Sr	93
LOI	5.67	Y	104
		Zr	209

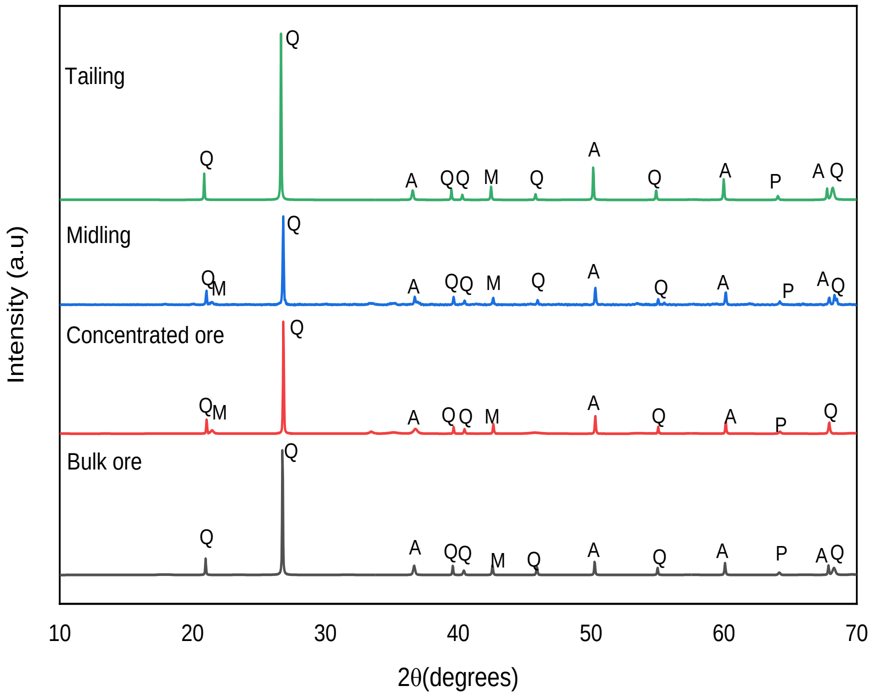


Figure 1. XRD diffractogram shows the bulk sample's mineral phases, pre-concentrated samples (concentrated ore, middling, tailing), and diagnostic leaching residue. (Q: quarzt, M: magnetite, A: arsenopyrite, P: pyrite).

Mineral Liberation Study. Mineral liberation is critical in the design of mineral processing plants. It is determined by the ore's mineralogical and textural characteristics. Estimating the liberation degree is critical for estimating the separation process's efficiency [17]. The mineral liberation percentage of each size fraction is calculated using an SEM micrograph and image J software. The mineral liberation calculation is based on the colour of the mineral that appears on the SEM image (Tabletop). Backscattered electron micrographs were used to identify and quantify this gold ore in twelve polished sections. The magnification used ranged from 100x to 1000x magnification. Each polished section's mineral surface was depicted in 50 scenes in the acquired images. It shows the difference in colour for each image for all of the images required. The lighter images indicate the presence of the metal element in the sample. As the study is about gold ore, it can be determined as the light in colour is present as pyrite and arsenopyrite, as these minerals are associated with gold formation. As the dark in colour, the mineral present as quartz as the formation of the soil is mainly from silica sand commonly [18]. This mineral identification is also based on the result of XRD.

The measurement of the degree of liberation was carried out using Equation 2.

$$\text{Degree of Liberation, \%} = \frac{Total\ Area\ of\ Liberated\ Particle}{Total\ Area\ of\ Liberated\ and\ Locked\ Particle}\ x\ 100 \qquad (2)$$

Figure 2 and Table 4 show the percentage of mineral liberation (Equation 2) at a given size fraction. The mineral liberation starts after size reduction to below 4.25 mm. The degree of liberation seems to increase with size reduction. However, the percentage of mineral liberation is relatively low, even at -0.075 mm. Only crushing processing takes place in this research work. The prediction of per cent of mineral liberation indicates that -0.075 mm particles produce good liberation (21.2%) of the mineral interest even though individual minerals' components were not distinctly separated. This study assumes that gold could occur in fined-grained, locked/attached gold associated with silicates [2]. The crushing or gravity concentration cannot recover sub-microscopic gold associated with relatively fine-grained sulphide [19]. However, further study on mineral liberation should be carried out with finer grain after the grinding process.

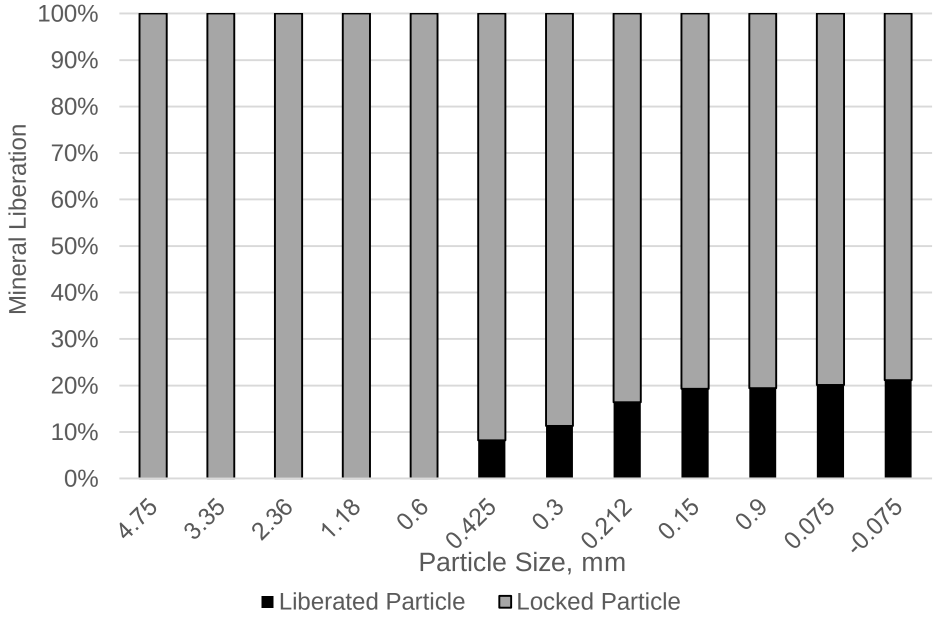
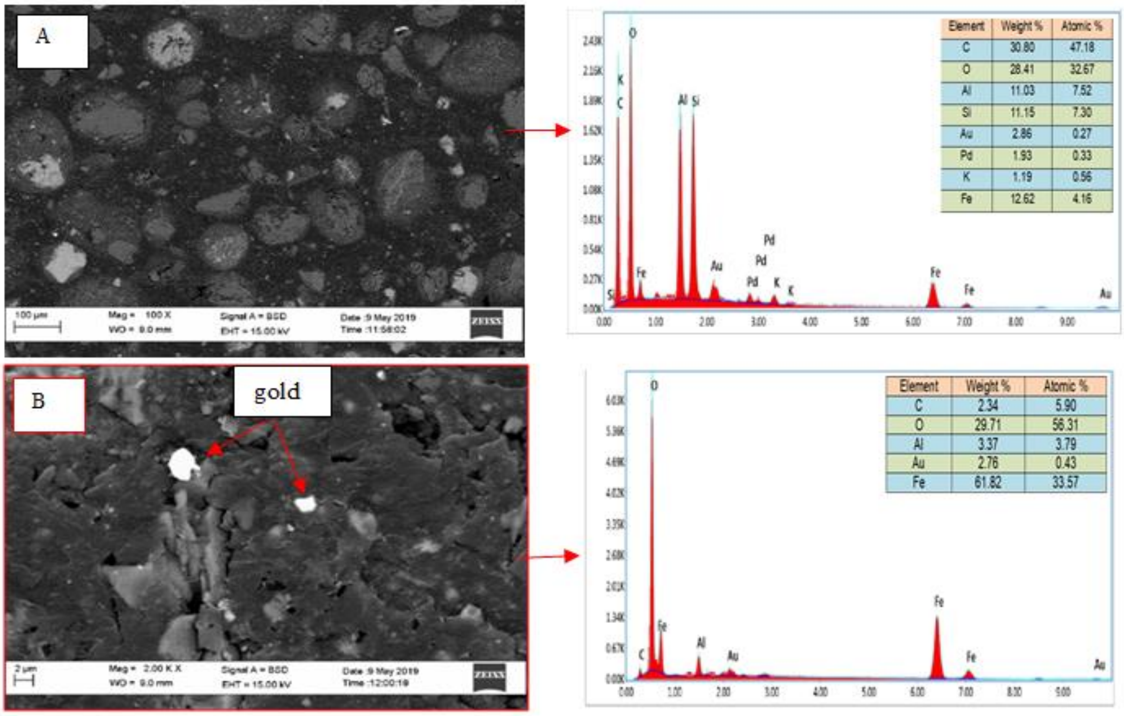


Figure 2. Resulting data from image analysis of the particle show as a percentage of the mineral liberation at particle size fraction of -4.75+3.35 mm, -3.35+2.36 mm, -2.36+1.18 mm, -1.18+0.6 mm, -0.6+0.425 mm, -0.425+0.3mm, -0.3+0.212 mm, -0.212+0.15 mm, -0.15+0.09 mm, 0.09+0.075 mm and -0.075 mm.

Table 4. Percentage of the liberated and locked particle at given size fraction.

Particle size (mm)	Liberated particle (%)	Locked particle (%)
+4.75	0	100
-4.75+3.35	0	100
-3.35+2.36	0	100
-2.36+1.18	0	100
-1.18+0.6	0	100
-0.6+0.425	8.22	91.78
-0.425+0.3	11.30	88.70
-0.3+0.212	16.37	83.63
-0.212+0.15	19.29	80.71
-0.15+0.9	19.41	80.59
-0.09+0.075	20.14	79.87
-0.075	21.15	78.85

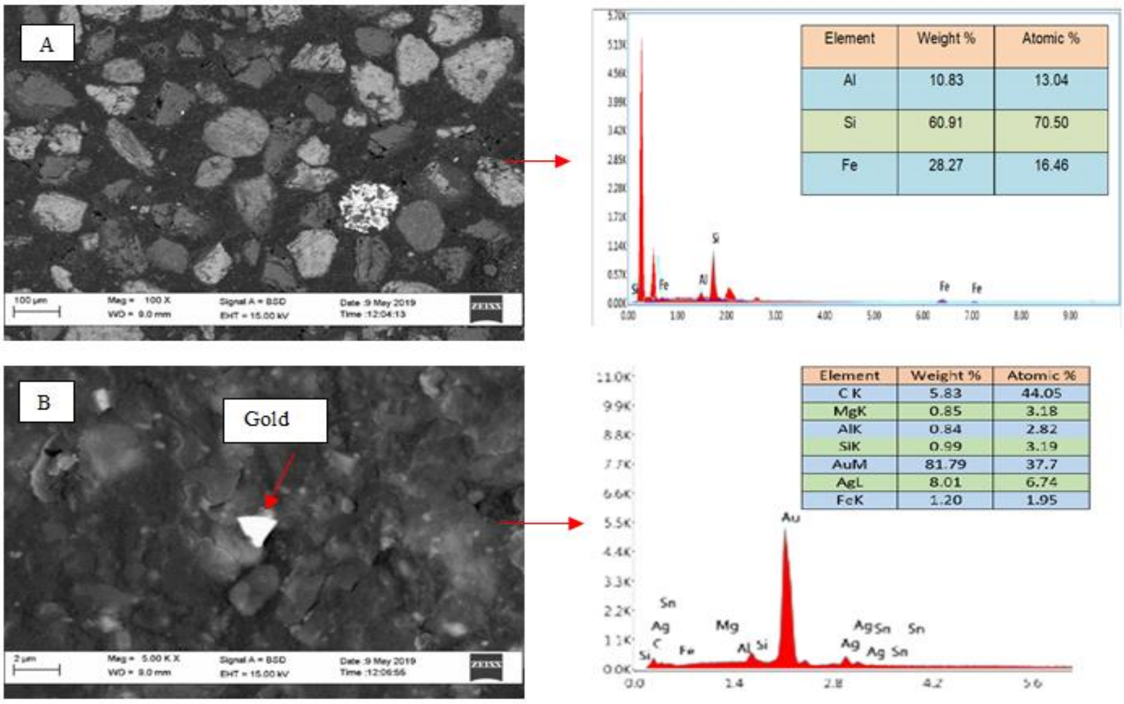
Ore morphological studies. The polish section for the bulk and concentrated samples was coated with platinum and scanned using the FESEM-EDX at different magnifications. Figure 3(A) shows the particle grain of bulk samples and EDX analysis, predominantly carbon and oxide, followed by aluminium, silicon, iron, potassium, trace amount of gold, and palladium. Figure 3(B) revealed that gold grain was detectable with a bright white shade. EDX analysis shows a higher percentage of Fe (62%) and oxygen (30%). It suggests that gold may be associated with iron-oxides minerals. The elemental composition determined by EDX analysis is consistent with XRF and XRD analysis, indicating a significant amount of iron oxides. Gold is frequently associated with an iron-sulphide mineral, such as pyrite or arsenopyrite, or an iron oxide/hydroxide mineral, like goethite, hematite, and limonite [20]. Figure 4(A) shows the particle grain of concentrated ore samples, which mostly have angular and sub-angular shapes. EDX analysis shows that Si (61%) is the dominant element identified, followed by Fe (28%) and Al (11%). At higher magnification (Figure 4(B)), gold grains were depicted with 82% Au via EDX scanned analysis. The gold grains depicted here could be linked to silicate minerals due to the presence of silver (Ag). It suggests that this ore contains a wide range of gold occurrences in silicates and sulphide minerals. According to Larrabure and Rodríguez-Reyes [20], silicon-containing minerals are frequently found alongside sulphides and oxides in low-grade silver or gold ores.



Element	Weight %	Atomic %
C	30.80	47.18
O	28.41	32.67
Al	11.03	7.52
Si	11.15	7.30
Au	2.86	0.27
Pd	1.93	0.33
K	1.19	0.56
Fe	12.62	4.16

Element	Weight %	Atomic %
C	2.34	5.90
O	29.71	56.31
Al	3.37	3.79
Au	2.76	0.43
Fe	61.82	33.57

Figure 3. Photomicrograph of backscattered image of bulk sample with magnification of (A) 100x and (B) 2000x.



Element	Weight %	Atomic %
Al	10.83	13.04
Si	60.91	70.50
Fe	28.27	16.46

Element	Weight %	Atomic %
C K	5.83	44.05
MgK	0.85	3.18
AlK	0.84	2.82
SiK	0.99	3.19
AuM	81.79	37.7
AgL	8.01	6.74
FeK	1.20	1.95

Figure 4. Photomicrograph of backscattered image of the concentrated sample with the magnification of (A) 100x and (B) 5000x.

Determination of head assay and free gold. This study determined the gold content of the collected and concentrated samples by fire assay and cyanidation test techniques. The prill obtained from fire assay was digested by aqua regia solution and analysed by ICP-OES. The gold content in each sample was found to be in the range of 0.1 to 0.4 g/t approximately (Table 5). The cyanidation test was carried out to determine the amenability of the gold ore to direct cyanidation and quantify the gold that can be extracted via direct cyanidation known as free milling gold. Free milling gold is the gold particles that are free and exposed, liberated through the milling process, and cyanide can reach and leach the gold. From this analysis of head assay and cyanidation test, approximately 50% of gold from the concentrated sample can be recovered by cyanide. Gold recovery of >90% can be readily achieved with a conventional cyanide leaching of free milling ores. However, refractory gold ores are often characterised by low gold extractions (<50-80%) in cyanide leaching [3].

Table 5. The gold extracted (g/t) in the bulk, concentrated and leach residue samples assayed by fire assay and cyanidation.

No	Samples	Gold Concentration (mg/dm^3)	Gold Concentration (g/t)
1	Bulk Sample 1	0.087	0.146
2	Bulk Sample 2	0.141	0.234
3	Concentrate	0.213	0.355
4	Diagnostic Leaching residual	0.056	0.093
5	Concentrate (Cyanidation test)	-	0.182 (average)

Diagnostic Leaching. A sequence of acid treatment with NaCN followed by HCl, H_2SO_4, HNO_3 and fire assay that improves the extraction of gold to 100% was observed (Figure 5). The initial cyanide leaching of the concentrated sample resulted in low gold extraction (38%), indicating the free milling gold recoverable by cyanide [4]. In the second stage, 16.7 % (0.115 g/t) of gold dissolution was achieved by HCl treatment. This step mainly dissolved carbonates, some chlorite and some oxides. (Eq.3) [4,12]. Next, in the third stage, sulphuric acid treatment extracted about 15.7% (0.108 g/t) of gold. It is assigned to decompose sulphide minerals, as shown in Equation 4 below. In the fourth stage, samples were treated with nitric acid, which disintegrated pyrite (Eq.5) [4]. This treatment extracted 16% of the gold. The remaining samples were treated with fire assay and associated with silicates [6,11]. This treatment dissolved 13.6% of the gold extract (0.093 g/t). Fine gold grains locked in remnant silicates such as quartz and feldspar are recovered from the final residue [9,12,21]. Some of these fine gold grains could only be liberated through ultra-fine grinding [2,12]. According to the diagnostic leaching data in Table 6 and Figure 5, the ore's refractoriness can be attributed to the association of gold with sulphide (31.7%), carbonates and oxides (16.7%), and silicates (13.6%). These findings are consistent with the findings of the characterisation analysis, which confirmed the presence and association of gold within sulphide, oxides, and silicates. Overall, the diagnostic leaching analysis suggested that without any pretreatment (free milling gold), only 38% of the gold could be recovered[2]. This ore can recover more than 86% of its gold using a standard pretreatment process to liberate the locked gold particles and oxidise sulphide. Other researchers studied the pretreatment process for refractory gold ore and discovered that gold recovery increased after pretreatment [4-6,19].

$$CaCO_{3(s)} + 2HCl_{(aq)} \rightarrow CaCl_{2(aq)} + CO_{2(g)} + H_2O_{(l)} \quad (3)$$

$$MS_{(s)} + H_2SO_{4(aq)} \rightarrow MSO_{4(aq)} + H_2S_{(g)} \text{ (M: zinc, copper, lead, etc)} \quad (4)$$

$$2FeS_{2(s)} + 10HNO_{3(aq)} \rightarrow 2Fe^{3+}{}_{(aq)} + 2H^{+}{}_{(aq)} + 4SO_4^{2-}{}_{(aq)} + 4H_2O_{(l)} \quad (5)$$

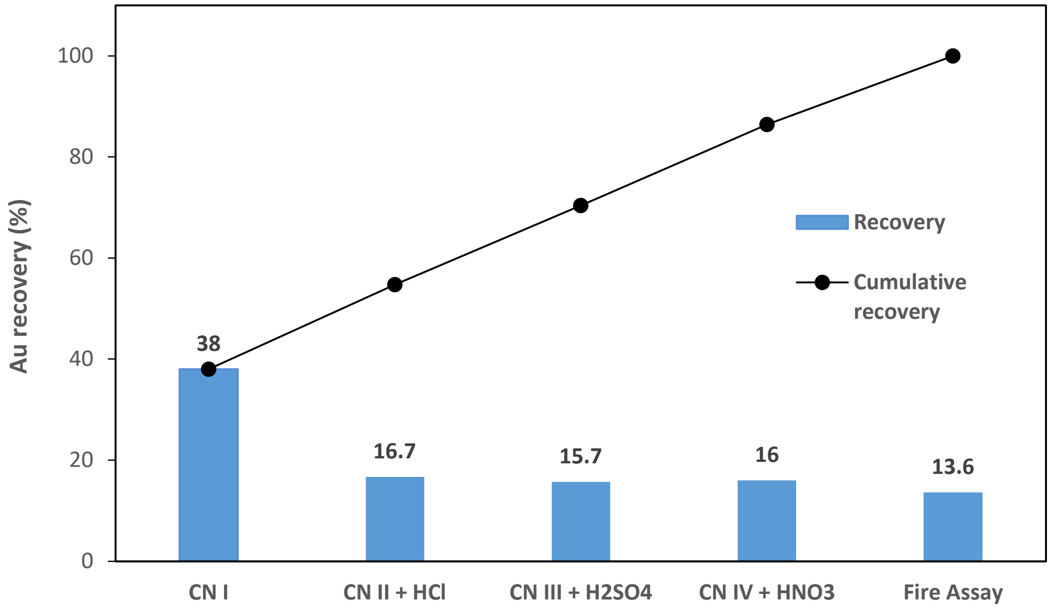
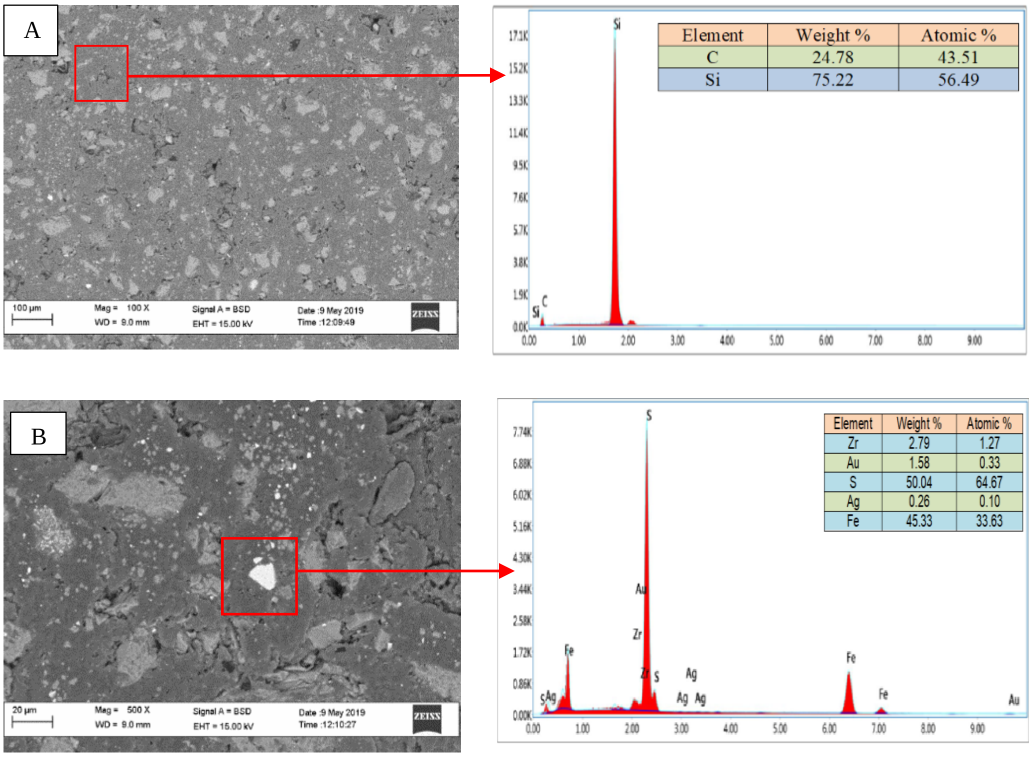


Figure 5. Extraction of gold following each stage of acid treatment stage of diagnostic leaching.

Table 6. The amount of gold extracted from each stage of cyanidation in diagnostic leaching

Diagnostic Leaching Stage	**Nature of Liberated Gold**	**% Au Extracted**
Fire Assay	Head Grade	100.00
Stage 1 - NaCN	Free milling gold	38
Stage 2 - HCl / CN	Gold in hematite,calcite,galena	16.7
Stage 3 - H_2SO_4 / CN	Gold in base metal sulphides	15.7
Stage 4 - HNO_3 / CN	Gold in pyrite, arsenopyrite	16
Residue by Fire Assay	Gold in silicate	13.7

Figure 6 shows SEM images and EDX analysis of residue of final diagnostic leaching. According to EDX analysis, the elemental analysis at the selected area corresponds to silicon and carbon composition. In contrast, the bright shade of a grey area corresponds to iron and sulfide composition (Figure 6(B)). This EDX analysis agrees well with stage 4 of the diagnostic leaching step, where the pyrite/arsenopyrite and silicate has been eliminated [13]. The diagnostic leaching test result agrees well with the cyanidation test, where this ore can be assumed refractory ore due to low recovery by the cyanide leaching process[21].



Element	Weight %	Atomic %
C	24.78	43.51
Si	75.22	56.49

Element	Weight %	Atomic %
Zr	2.79	1.27
Au	1.58	0.33
S	50.04	64.67
Ag	0.26	0.10
Fe	45.33	33.63

Figure 6. Backscatterred image of leaching residue with a magnification of (A) 100x and (B) 500x.

Conclusion

This work studied the refractoriness of gold ores from the East Coast of Peninsular Malaysia. The sample is mainly composed of silica, iron, aluminium, and potassium; however, no gold was detected due to low concentration or gold interlocked with other minerals. In mineral phase analysis, quartz predominates the sample, followed by arsenopyrite, pyrite, and magnetite. There are gold particles that interlock with silicates and occur as submicroscopic gold in sulphide. Diagnostic leaching revealed that only 38% of free milling gold extraction was achieved, and this ore can be classified as a highly refractory ore. After leaching sulphide minerals with sulphuric acid, 31.7% of gold was recovered, while the remaining was locked in silicates. The total gold liberated from its host or associated mineral after diagnostic leaching is 86%. This gold-bearing ore from East Coast Peninsular Malaysia is classified as refractory gold-bearing ore that requires pretreatment before gold extraction can begin. This study has shown that diagnostic leaching is critical for evaluating new gold ores' extraction potential.

Acknowledgements

The author would like to thank Universiti Sains Malaysia (USM) for the financial support through School of Materials and Mineral Resources Engineering and Selinsing Gold Mine Manager Sdn Bhd for providing samples.

Author Contributions

All authors contributed toward data analysis,drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of Conflict of Interest

The authors have no disclosures to declare.

Compliance with Ethical Standards

The work is compliant with ethical standards.

References

[1] Ariffin, K.S., (2012). Mesothermal Lode Gold Deposit Central Belt Peninsular Malaysia. *Earth Sciences*. InTech, pp. 313–342. Available from: http://www.intechopen.com/books/ earth-sciences/mesothermal-lode-gold-deposit-central-belt-peninsularmalaysia. (ISBN: 978-953-307-861-8)

[2] Zhou, J., Jago, B. & Martin, C., (2004). Establishing the process mineralogy of gold ores. *Technical Bulletin*, SGS Minerals 3 1-16.

[3] Marsden, J.O. & House, C.I. (2006). The chemistry of gold extraction, *Society of Mining*, Metallurgy and Exploration Inc. (SME), Colorado, USA.

[4] Saba, M., MohamedYousefi, A., Rashchi, F., Moghaddam, J. (2011). Diagnostic pretreatment procedure for simultaneous cyanide leaching of gold and silver from a refractory gold/silver ore. *Minerals Engineering*, 24 1703-1709.

[5] Zhou, J.Y. & Cabri, L.J. (2004). Gold process mineralogy: Objectives, techniques and applications. *Technical Bulletin*, SGS Minerals 14 1-6.

[6] Nazari, A.M. Ghahreman, A. & Bell, S. (2017). A Comparative Study of Gold Refractoriness by the Application of QEMSCAN and Diagnostic Leach Process. *Inter. J. of Min. Pro*, 169 35-46.

[7] Lorenzen, L. (1995). Some guidelines to the design of a diagnostic leaching experiment. *Minerals Engineering* 8 (3) 247-256

[8] Henley, K. J. Clarke, N. C. & Sauter, P. (2001). Evaluation of a diagnostic leaching technique for gold in native gold and silver tellurides. *Minerals Engineering* 14(1) 1-12.

[9] Teague, A. J. Swaminathan, C. and Van Deventer, J. S. J. (1998). The behaviour of gold bearing minerals during froth flotation as determined by diagnostic leaching. *Minerals Engineering* 11(6) 523-533.

[10] Goodall, W. R., Scales, P. J., & Butcher, A. R., (2005). The use of QEMSCAN and diagnostic leaching in the characterisation of visible gold in complex ores. *Minerals Engineering,* 18(8) 877-886.

[11] Celep, O. Alp, I. Deveci, H. & Vicil, M. (2009). Characterisation of refractory behaviour of complex gold/silver ore by diagnostic leaching. *Trans.of Nonferrous Met.Soc. of China (English Edition)* 19(3) 707-713.

[12] Coetzee, L.L. Theron, S.J. Martin, G.J. van der Merwe, J.D. & Stanek, t. (2011). Modern gold deportments and its application to industry. *Minerals Engineering* 24(6) 565-575.

[13] Celep, O. & Serbest, V. (2015). Characterisation of an iron oxy/hydroxide (gossan type) bearing refractory gold and silver ore by diagnostic leaching. *Trans. of Nonferrous Met. Soc. of China (English Edition)* 25(4) 286-297

[14] Anggraini, A.G., (2020). Study of gold ore characterisation from Tanggamus regency by using diagnostic leaching method. Available at: http://digilib.unila.ac.id/62434/,(Accessed 12th July 2020)

[15] Celep, O. Alp, I. & Deveci, H. (2008). Application of diagnostic leaching technique for refractory gold ores. *Fen Bilimberi Institüsü Dergisi*, 16th September, p.81-90

[16] Hoffman, E.L. Clark, J.R. & Yeager, J.R. (1998). Gold analysis- Fire assaying and alternative methods. *Exploration Mining and Geology* 7(1-2) 155-160.

[17] Olubambi, P.A., Ndlovu S., Potgieter J.H., & Borode J.O. (2008). Mineralogical characterisation of Ishiagu (Nigeria) complex sulphide ore. *International Journal of Minerals Processing* (87) 83-89.

[18] Ismail, S, Hashim,S.F.S,Hussin, H, & Abdullah, N.S.(2016). Chemical and Mineralogical Characterisation of Malaysian Low-Grade Manganese Ore. *Periodico di Mineralogia* (85) 277-288.

[19] Nie, Y. Chen, J. & Wang, Q. (2021). Use of dry grinding process to increase the leaching of gold from a roasted concentrate containing hematite in the thiosulfate system. *Hydrometallurgy*, 201 (May 2021)

[20] Larrabure, G. & Rodríguez-Reyes, J.F.C. (2021). A review on the negative impact of different elements during cyanidation of gold and silver from refractory ores and strategies to optimise the leaching process. *Minerals Engineering* (173) 107194.

[21] Meakin, R. Chapman, N. Frangeskides, Z. & Kuhar, L.(2012). Maximising the value of gold diagnostic leaching. *Proceeding of ALTA 2012 Gold Conference,* ALTA Metallurgical Services, Burtswood Convention Center, Perth Australia, 31st May – 1st June, pp.99-110.