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Mineralogy, petrology, and chemistry studies to evaluate oxide copper ores for heap leaching in Sarcheshmeh copper mine, Kerman, Iran

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Abstract

In recent years, as a result of biological, environmental, and economic considerations, available copper in copper oxide ores that could not be recovered by pyrometallurgical methods was accumulated in so-called oxide dumps. Suitable material is treated with dilute sulfuric acid in a heap-leaching process, whereupon the copper content of the rock slowly dissolves in the acidic solution. The performed investigations show that one needs to consider the action of the acid on the copper oxide-containing rocks at the microscopic level.

In this paper, we describe research carried out on oxide samples from the western dump of the Sarcheshmeh copper mine. Each sample was split into two parts and a portion of each was exposed to heap-leaching conditions in a column. Subsequently, polished sections, thin sections, and powdered samples were subjected to chemical analysis as well as petrographic and mineralogical considerations. Changes in the weight percentages of non-metal and metal minerals before and after acid treatment were measured. Microscopic studies have indicated that chemical analyses do not provide a complete picture of the effects of acid on the rock. Thus, microscopic studies on sections are shown to be a necessary requirement, neglection of which can have negative economic and environmental effects.

Keywords: Mineralogy; Petrography; Chemistry; Heap leaching; Oxide copper ore

1. Introduction

Heap-leaching operations have gained increasing importance in the Iranian mining industry [1], in recent years, as they offer the possibility of treating oxide copper ores. From a metallurgical point of view [2], key parameters controlling heap-leaching operations are the crushed rock size distribution and resulting porosity, permeability and height of the heap, acid pre-treatment, acid concentration and irrigation rate, total and soluble copper contents of the ore, and the bulk rock mineralogy and chemistry [3].

Sarcheshmeh copper mine is located 160 km south-west of Kerman region (Fig. 1). Geologically (Fig. 2), in a vertical section, it is divided into three zones. named oxide, supergene, and hypogene (Fig. 3). A vast amount of copper has hitherto been extracted from sulfide rocks (Fig. 4), but in recent years copper has been recovered by hydrometallurgical methods from oxide rocks [4].

Hydrometallurgical methods include leaching, solvent extraction, deposition, and electrolysis filtering [5]. Hydrometallurgical copper recovery starts with exposure to sulfuric acid as a solvent for copper oxide rock mass. The copper-containing solution is recovered in subsequent hydrometallurgical processes. At the Sarcheshmeh copper mine, copper oxide rocks that have been excavated in past years are kept in the oxide dump and are exposed to a gradual flow of dilute sulfuric acid (Fig. 5). In this process, the copper dissolves in the acid and the solution produced is collected from this section. Cathodic copper is deposited after a special electrolytic process. This method is preferred to a pyrometallurgical one on both economic and environmental grounds.

The exploitation of copper oxide ores was not economical in the past, but heap leaching now has the advantages of low investment, better working conditions, and lower energy consumption [6]. This process is continuously being developed to improve the removal of copper from a wide variety of ores; in many cases the ore is microscopically inseparable mixture of reactive gangue

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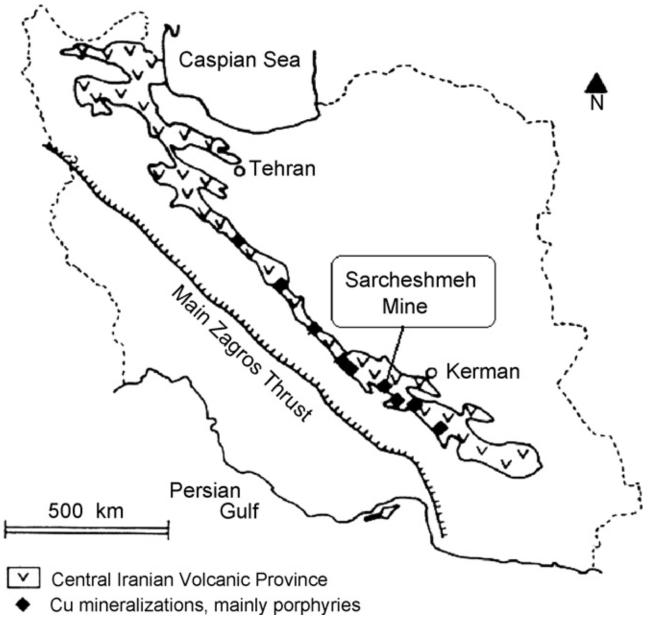

Fig. 1. Sketch map showing the location of the Central Iranian Volcanic Province and porphyry copper-type mineralization relative to the main Zagros thrust.

Fig. 3. Three vertical zones of the Sarcheshmeh copper mine [18].

minerals such as smectites, white micas, chlorites and zeolites [7]. It can also be presented as microcrystalline Cu phases or Cu retained already in cation exchange sites prior to mixing and processing. Gangue reactivity produces mainly acid consumption loss of permeability copper exchange and adsorption and finally, copper retention in the pad [8]. The best solvent for copper oxide rocks is dilute sulfuric acid. However, the most important parameters in the dissolution process are the size (Fig. 6 shows curves labelled with pH) and gradation of the copper oxide rock types and the amount of gangue, the way in which the oxide dump is laid out in the place of heap leaching, the temperature, the way in which the acid is applied, pH effects, and the duration of the leaching, as well as the nature of the oxide rock used for the heap-leaching process and how it was excavated from in situ oxide rock masses (Fig. 7).

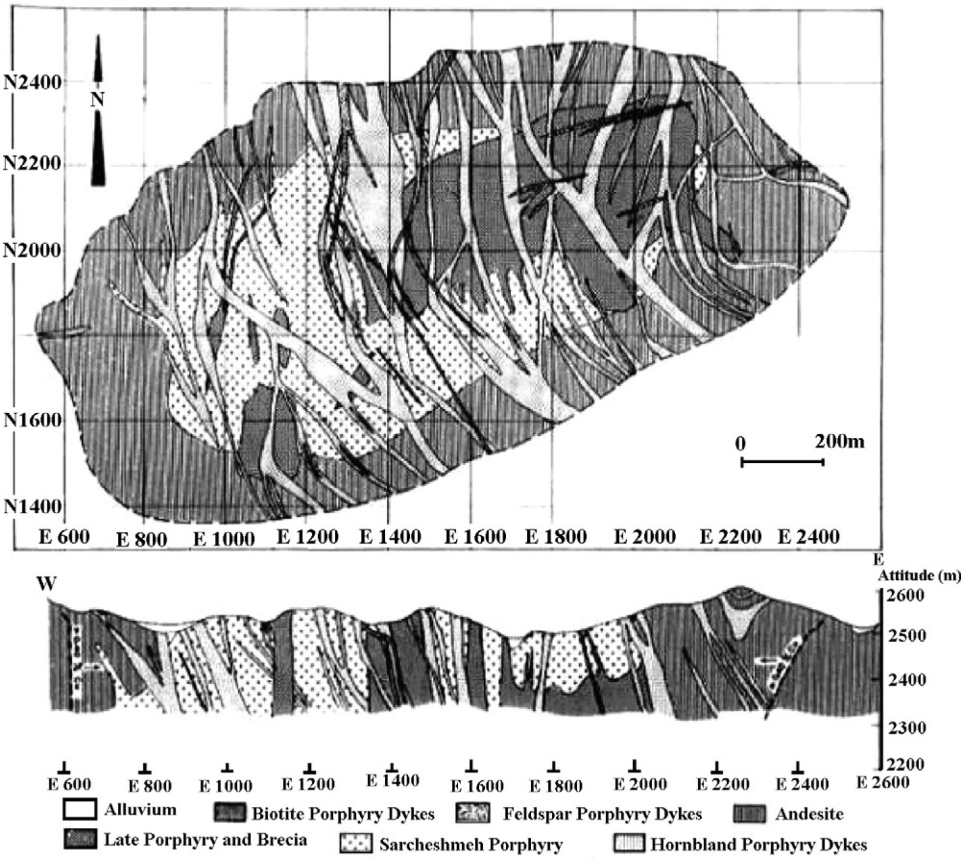

Fig. 2. Distribution of rock types at 2400 m elevation within outer 0.40% copper line Sarcheshmeh deposit with geological section [17].

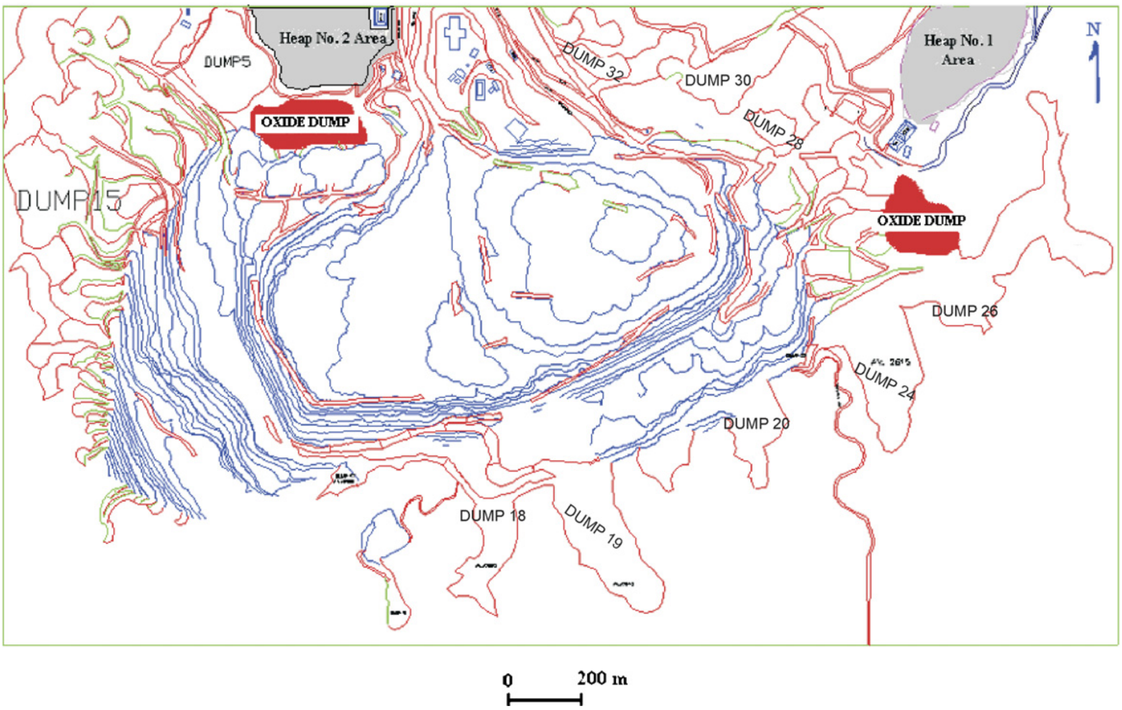

Fig. 4. The locations of the western dumps and the heap-leaching facilities at the Sarcheshmeh copper mine.

Fig. 5. Heap leaching with drop emitters on the ore surface at Sarcheshmeh porphyry copper mine (looking toward the north).

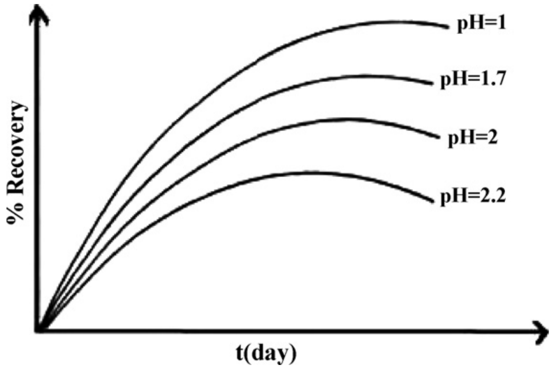

Fig. 6. Graph showing the effect of concentration on recovery [13].

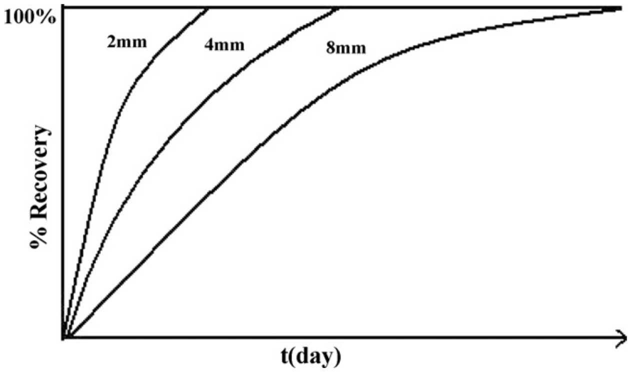

Fig. 7. Graph showing the effect of grain size on recovery [13].

Indeed, the rock samples can have wide ranges of size and gradation. Rock samples ranging from boulders of size 1 m to small soil particles are settled under special circumstances and treated with acid for about 60 days. In this period, as the acid penetrates inside the rock mass and soil, it gently dissolves certain components of the rock and transports them out of the mass. A collector system then collects the copper-containing solution, referred to as PLS (pregnant leach solution), and copper is recovered in a subsequent process.

2. Materials and methods

In order to evaluate the effect of dilute sulfuric acid on the mineralogy, petrography, and chemistry of rock fragments, 15 oxide rock samples in the size range 3–3.5 in. (7.5–9.0 cm) were selected from the western dump of the Sarcheshmeh copper mine. Each sample was split into two parts so that there were two

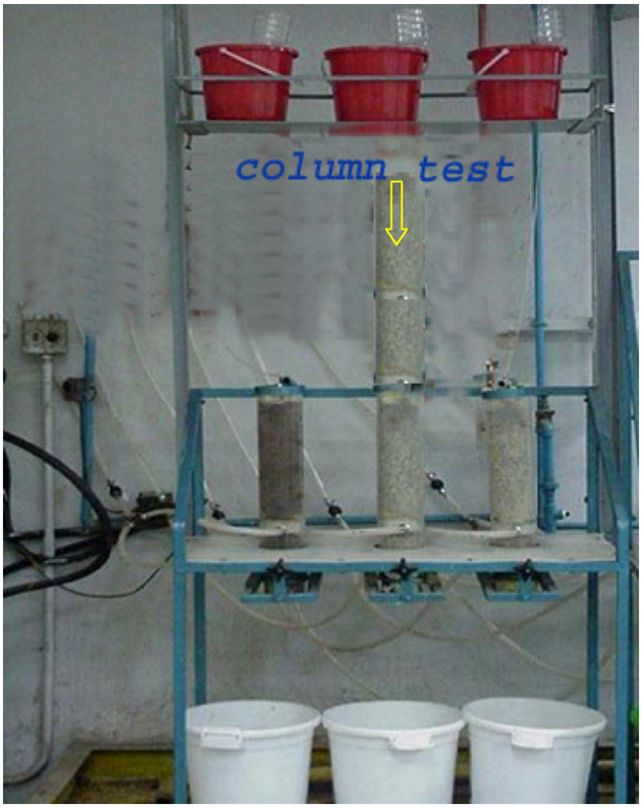

Fig. 8. Leaching system and solution collector [13].

groups of 15 portions. The first group of samples were placed in a laboratory-scale cylindrical column in a set up simulating a pilot plant and irrigated with sulfuric acid at pH 1.9 for 60 days at an outlet flow rate of 1 cm^3/min. In order to assess the effect of the acid on different portions, the samples were placed in a 1.5 m high column (Fig. 8). After irrigating with 8 l of acid, the pH of the PLS (pregnant leach solution) produced was measured and controlled. If the pH had increased upto 3–3.5, then the pH of the incoming solution was reduced to 1.3. Figs. 9 and 10 show the changes effected on the samples arranged from the top to the bottom of the simulated column.

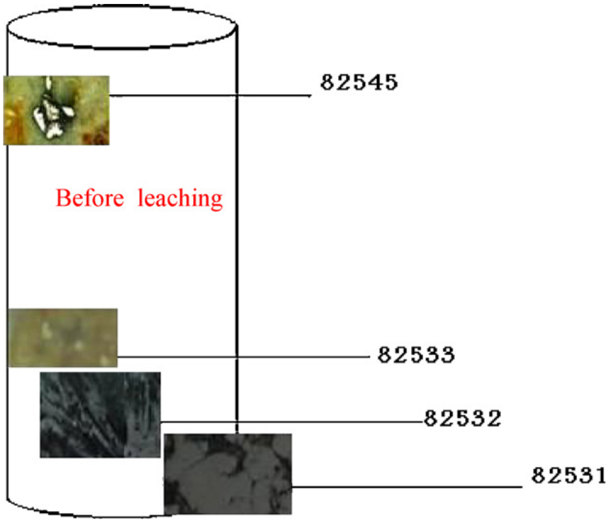

Fig. 9. Schematic depiction of an experimental column and the relative positions of the samples before acid leaching.

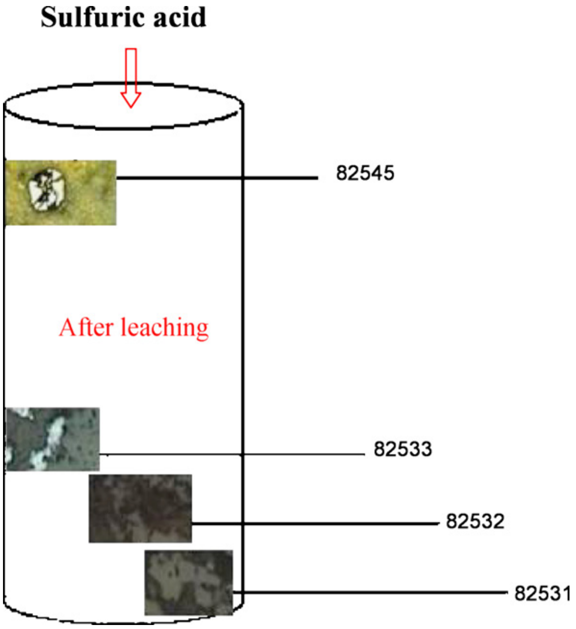

Fig. 10. Schematic depiction of an experimental column and the relative positions of the samples after acid leaching.

From the other group of samples, more than 30 polished and 25 thin sections were prepared and carefully studied by means of ore and polarized microscopy (Olympus BM-60M). The remaining part was analyzed chemically by means of atomic absorption spectroscopy using a Varian Spectra A.A. 220 with graphite furnace. After the first samples had been subjected to acid treatment for 60 days, sections of these were also prepared, powdered, and chemically analyzed in order to assess the changes that had occurred. Analytical results concerning the percentages of Fe, Cu (total Copper), and CuO (soluble Copper) measured throughout this process are plotted in Figs. 11–13, respectively, and collected in Table 1. Also, changes in the weight percentages

Table 1
Chemical composition in terms of Fe, CuO (Soluble), and Cu (Total) contents before and after acid leaching on west dump at Sarcheshmeh

Number of sample	Before acid leaching			After acid leaching		
	%Cu	%CuO	%Fe	%Cu	%CuO	%Fe
82531	0.39	0.39	0.06	0.09	0.06	0.19
82532	2	1.64	1.44	0.07	0.02	1.25
82533	0.13	0.07	1.66	0.04	0.01	2.35
82534	4.1	1.13	0.9	1.57	0.98	1.46
82535	0.16	0.1	1.15	0.04	0.01	1.9
82536	0.2	0.12	1.4	0.05	0.02	1.7
82537	1.24	1	2.71	0.87	0.6	2.8
82538	0.24	0.22	0.3	0.13	0.05	0.6
82539	0.2	0.16	2.84	0.05	0.02	2.5
82540	0.17	0.09	8.4	0.03	0	2.95
82541	0.16	0.07	3.76	0.11	0.03	2.53
82542	0.1	0.06	2.32	0.04	0.01	1.7
82543	3.35	2.94	4.94	0.06	0.05	0.1
82544	0.11	0.09	0.18	0.02	0	1.02
82545	0.36	0.17	5.06	0.1	0.01	3.8

Table 2
Weight percentages of limonite, sphalerite, hematite, and magnetite before and after acid leaching

Number of sample	Limonite		Sphalerite		Hematite		Magnetite	
	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching
82531	0.138	0.35	0.051	0	0.033	0.345	0.021	0.017
82532	0.424	0.562	0.092	0.23	0.052	0.343	1.003	0.504
82533	2.042	1.177	0.072	0.711	0.375	0.369	0.03	1.236
82534	0.236	0.277	0.028	0.18	0.276	0.121	0.011	0.725
82535	0.3	1.364	0.182	0.393	0.636	0.279	0.09	0.609
82536	1.058	1.568	0.015	0	0.27	0.342	0.038	0.132
82537	1.961	1.324	0.11	0.563	1.123	0.138	0.19	1.657
82538	0.114	0.225	0.009	0.051	0.263	0.027	0.015	0.311
82539	1.159	0.65	0.208	0.229	0.052	0	0	0.158
82540	7.288	1.603	0.077	0.564	0.193	0.25	3.774	1.773
82541	4.886	2.308	0.31	0.077	0.216	0.321	0.32	0.826
82542	1.542	1.427	0.76	0.296	0.634	0.117	0.126	0.116
82543	2.887	0.084	0.096	0.1	2.196	0.009	0.158	0.004
82544	0.016	0.261	0.003	0.107	0.018	0.035	0	0.025
82545	6.707	4.019	0	0.597	0.661	0.364	0.347	0.834

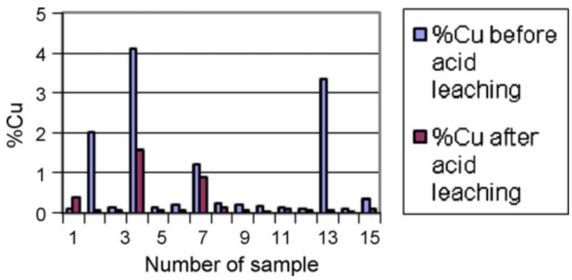

Fig. 11. Plot of Cu (total) content before and after acid leaching.

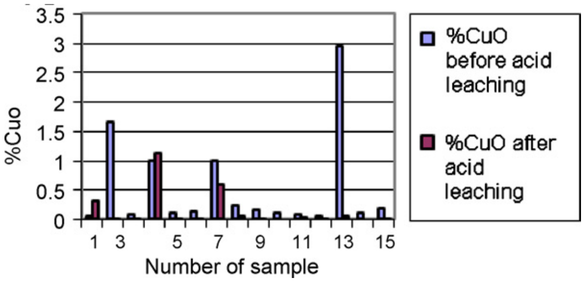

Fig. 12. Plot of CuO (soluble) content before and after acid leaching.

of various metal minerals were estimated (Tables 2–4) before and after the acid leaching (Figs. 14–23) in the same way as in Table 1. Finally, the results of both methods (microscopic as well as analytical), carried out simultaneously and in parallel on the same samples, were carefully compared to check the accuracy of the work.

3. Removal of iron from copper leach solution

The large amount of ferrous sulfate generated by oxidation of pyrite is oxidized to ferric sulfate and hydrolyzed to jarosite (Fig. 24). This precipitate forms within the dump, but generally outside the rock pores under aerobic conditions:

Table 3
Weight percentages of calcocite, covellite, chalcopyrite, and native copper before and after acid leaching

Number of sample	Calcocite		Covellite		Chalcopyrite		Native copper	
	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching
82531	0	0	0.022	0	0.04	0.231	0.021	0
82532	0.157	0	0.099	0	0.487	0.144	0	0
82533	0	0	0	0	0.05	0.087	0.043	0
82534	0	0.052	1.332	0.532	1.587	0.13	1.685	0
82535	0.026	0	0.019	0.022	0.026	0.045	0.018	0
82536	0	0.008	0	0.003	0.131	0.062	0.035	0
82537	0	–	0	0	0.693	0.78	0	0
82538	0.008	0.038	0	0.015	0.019	0.082	0.007	0
82539	0.009	0	0.004	0.003	0.088	0.059	0	0
82540	0	0	0.082	0	0.073	0.087	0	0
82541	0	0.052	0	0.005	0.085	0.052	0.06	0
82542	0.022	0.024	0	0	0.032	0.016	0.011	0
82543	0	0.012	0	0	1.184	0.002	0	0
82544	0	0.022	0	0	0.058	0.008	0	0
82545	0.029	0	0	0	0.026	0.086	0.0137	0

Table 4
Weight percentages of bornite and pyrite before and after acid leaching

Number of sample	Bornite		Pyrite	
	Before acid leaching	After acid leaching	Before acid leaching	After acid leaching
82531	0	0	0.09	0.119
82352	0	0	0.532	0.436
82533	0	0	0.13	0.627
82534	0	0	0.133	1.295
82535	0	0	0.874	0.675
82536	0	0	1.019	0.749
82537	0	0	0.685	0.699
82538	0	0.019	0.055	0.383
82539	0	0.012	4.308	4.107
82540	0	0	1.972	0.751
82541	0	0.027	0.456	0.478
82542	0	0.008	1.402	1.226
82543	0	0	2.343	0.038
82544	0	0	0.298	1.695
82545	0	0	0.206	0.46

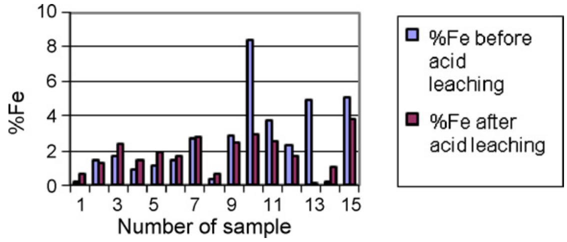

Fig. 13. Plot of Fe content before and after acid leaching.

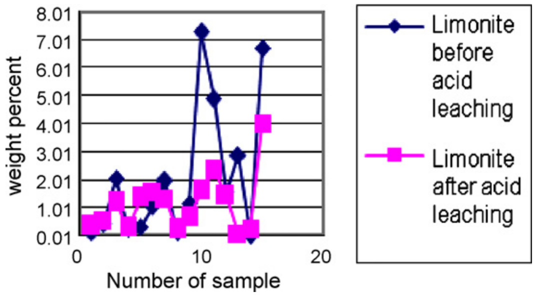

Fig. 14. Plot of weight percent of limonite before and after acid leaching.

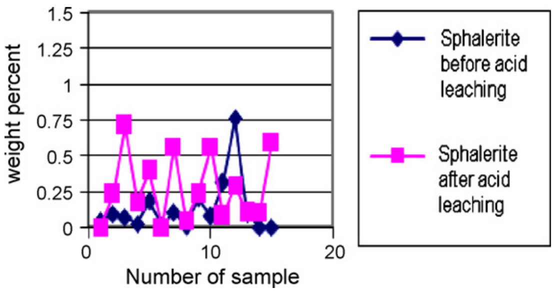

Fig. 15. Plot of weight percent of sphalerite before and after acid leaching.

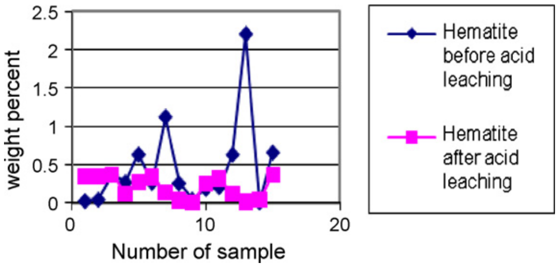

Fig. 16. Plot of weight percent of hematite before and after acid leaching.

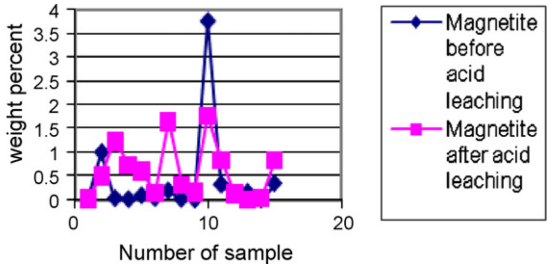

Fig. 17. Plot of weight percent of magnetite before and after acid leaching.

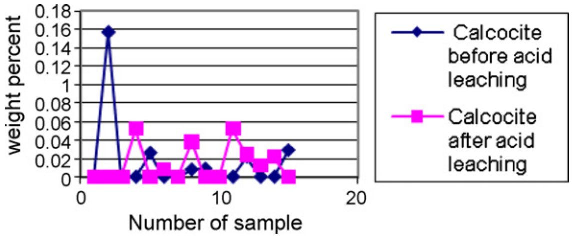

Fig. 18. Plot of weight percent of calcocite before and after acid leaching.

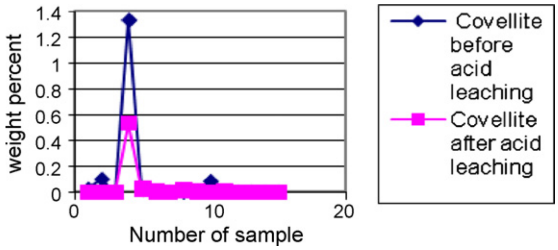

Fig. 19. Plot of weight percent of covellite before and after acid leaching.

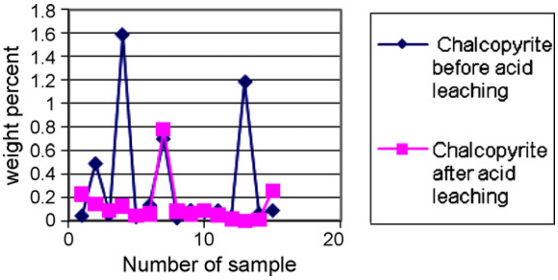

Fig. 20. Plot of weight percent of chalcopyrite before and after acid leaching.

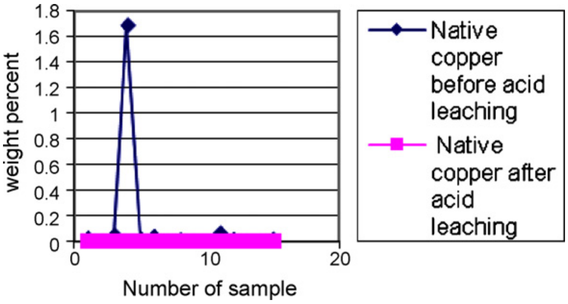

Fig. 21. Plot of weight percent of native copper before and after acid leaching.

Oxidation:

$$2FeSO_4 + H_2SO_4 + 1/2O_2 = Fe_2(SO_4)_3 + H_2O \quad (1)$$

Precipitation of jarosite:

$$3Fe_2(SO_4)_3 + 0.8K_2SO_4 + 12H_2O = 2K_{0.8}H_{0.2}Fe_3(SO_4)_2(OH)_6 + 5.8H_2SO_4 \quad (2)$$

Hydrolysis and precipitation prevent iron, dissolved from both pyrite and chalcopyrite, from accumulating in the solution, beyond the equilibrium value for ferric iron and other ferric complexes (Fig. 25). Total soluble ferric iron is increased by sulfate complexing, and mature dump leach solutions have a high concentration of sulfate ions due to the presence of large amounts of magnesium sulfate and aluminum sulfate salts [2].A typical leaching dissolution chemical reaction is:

$$CuSiO_3{\cdot}2H_2O + H_2SO_4 = CuSO_4 + 3H_2O + SiO_2 \quad (3)$$

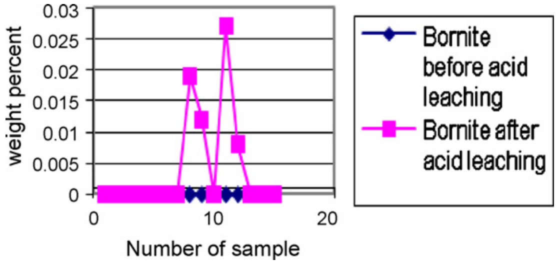

Fig. 22. Plot of weight percent of bornite before and after acid leaching.

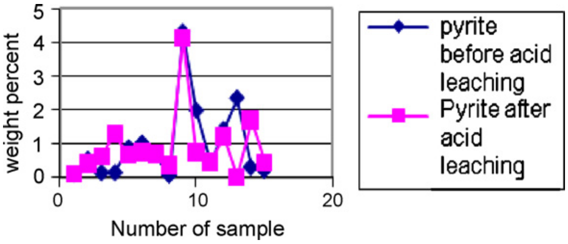

Fig. 23. Plot of weight percent of pyrite before and after acid leaching.

As the acid initially enters the rock pores, it is quickly consumed by rapid copper oxide mineral dissolution reactions before it can penetrate to the center of the rock (Figs. 26–28). Thus, the initial penetration of even a strong acid solution will produce a leached rim or shell surrounding an unaffected core of the rock (Fig. 29)

So, what is the mechanism for extracting copper from a simple ore fragment during leaching? As it dissolves, copper leaves the rock by diffusion through the stagnant pore liquor [9]. Although additional acid cannot flow into the rock pores, acid can diffuse into the rock pores under an acid concentration gradient. Hence, there is countercurrent chemical diffusion of hydrogen and copper(II) cations through the stagnant liquid solution in the rock pores [6].

Sawing a partially leached rock readily shows the leached rim (bleached of color) and the unleached core colored blue–green by the copper oxide minerals. The smallest rocks show leached rims and progressively smaller fractions of the copper extracted with increasing rock size [10].

The preceding results relating to column leaching tests indicate that increasing heap height in oxide copper ore percolation leaching will not increase the extraction rate proportionately.

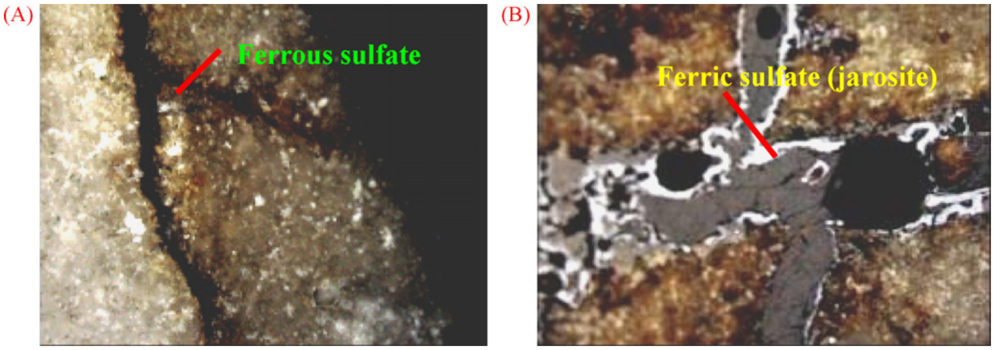

Fig. 24. (A) Photomicrograph showing ferrous sulfate generated by oxidation of pyrite is oxidized to ferric sulfate and hydrolized to jarosite (B). This has been formed at the expence of sulfide minerals around the rim (XPL. 10×).

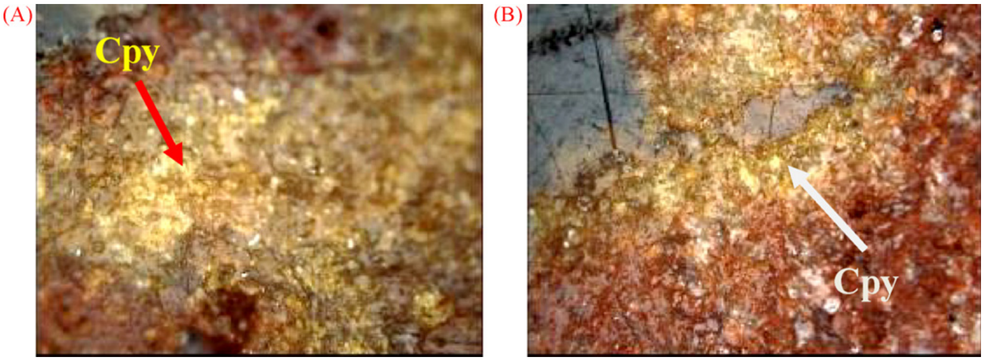

Fig. 25. (A) Photomicrograph showing chalcopyrite before acid leaching; (B) photomicrograph of the sample after acid leaching, in which the amount of chalcopyrite is seen to be unchanged, and the amount of oxides has decreased concomitantly (PPL. 5×).

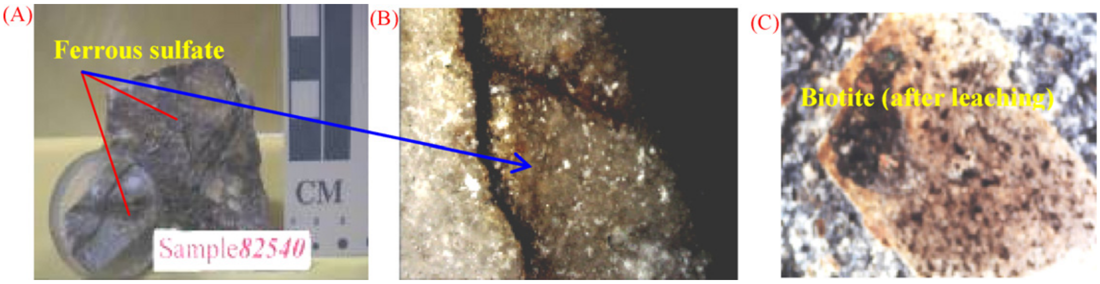

Fig. 26. (A) Polished section along with the hand specimen; (B) photomicrograph of the same polished section; (C) petrography of the same sample. Significantly, (B) (XPL. 10×) and (C) (XPL. 20×) show that the amount of ferrous sulfate (A and B) are considerable before the leaching, (Idiomorphic biotite is preserved in (C)).

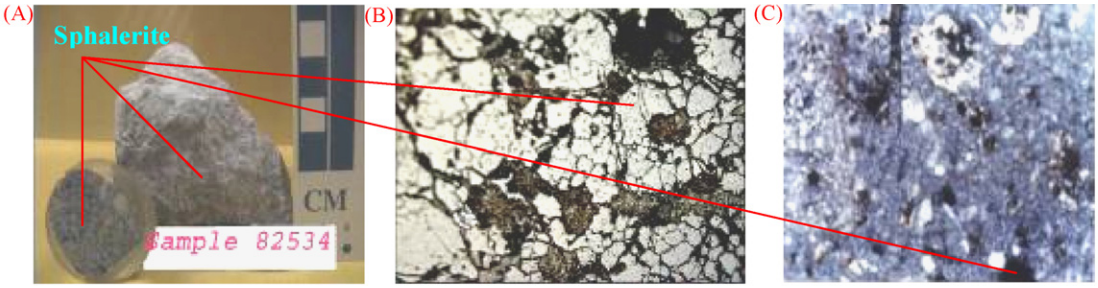

Fig. 27. (D) Polished section along with the hand specimen; (E) photomicrograph of the same polished section showing the sphalerite after acid leaching; (F) petrography of the same sample. Significantly, (E) (XPL. 10×) and (F) (XPL. 5×) indicate that the amounts of calcocite and covelite are decreased and the amount of sphalerite has been increased. The rock type is the Sarcheshmeh porphyry.

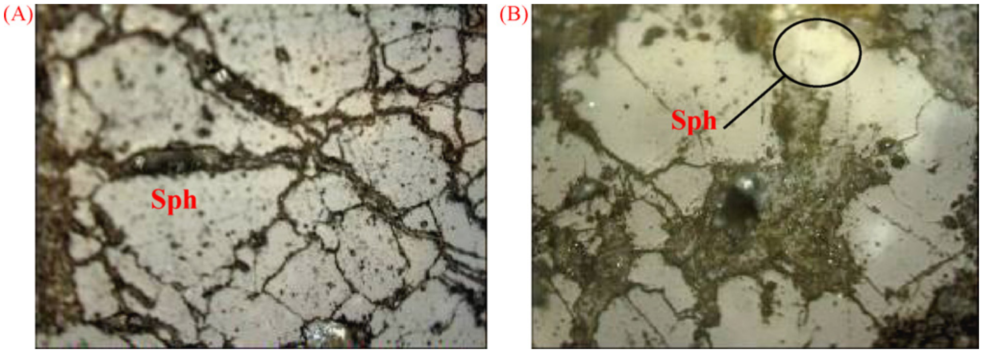

Fig. 28. (A) Photomicrograph of sphalerite sample (number 82538) before acid leaching (PPL. 10×); (B) the same sample with little change in color, perfect IR and the “triangle zoning” of shalerite crystal after acid leaching (XPL. 20×).

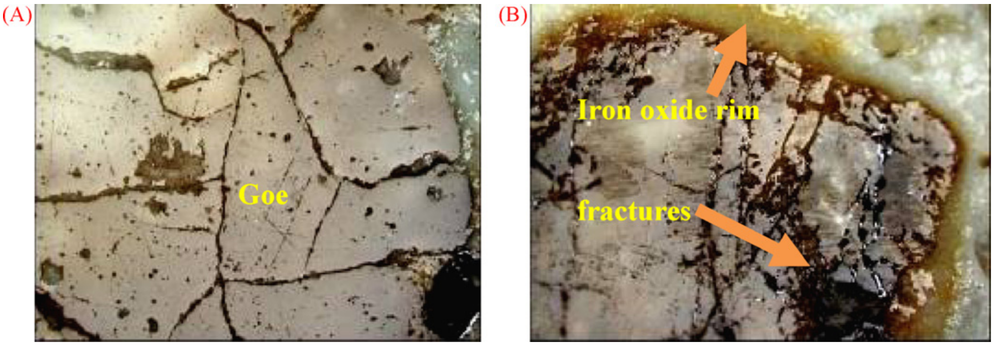

Fig. 29. (A) Goethite crystal assembly before acid leaching; (B) the same sample after acid leaching with more fracture and leaching iron oxide rim; it can be seen that Oxihydroxide fills open spaces around the quartz grains (XPL. 10×).

Typical copper mine oxide ore has an average copper content of 0.2% and the amount of copper extracted by a leaching method is often less than 0.1% [11].

Consequently, a simple, inexpensive process consists of the sequence: pregnant liquor pond → copper recovery plant → barren liquor pond.

4. Results and discussion

- Investigation of this process has shown that the size of the rock can affect copper recovery. Acid penetration to the interior of a large rock mass needs more time than for smaller rocks. Increasing time reduces the economic efficiency of this method. Thus, economic and optimum comminution can be helpful; microcracks reduce the penetration time and make the dissolution process easier.
- Also, changes in acid concentration have an effect on this process. In the laboratory-scale experiment [12], it was noticeable that the percentage Fe content increased in the lower half of the column. This may have been due to a reduction in the acid concentration as a result of its reaction with soluble contents. As the concentration decreases, it leads to a reduction in the Fe content of the solution. A significant decrease causes Fe to become deposited as its basic sulfate, which covers the surface of minerals and limits the effectiveness of the acid.
- This problem can be solved by measuring the pH of the effluent and controlling the pH of the incoming acid accordingly. Acid in leach liquor attacks gangue minerals within the heap [13], where Cu(II) is generated at rates that depend on pH and vary among the numerous gangue minerals present [14]. Generally, the rate of acid attack and neutralization increases with increasing acid concentration (lower pH). A steady-state condition is reached in each heap at a pH at which the rate of introduction of the acid balances the rate of acid consumption by gangue mineral neutralization.
- The majority of acid consumption in ore from primary deposit oxide mines is usually associated with the extraction of magnesium, aluminum and alkali metal oxides from biotite mineral [$H_2K(Mg,Fe)_3\cdot Al(SiO_4)_3$] and similar basic silicate minerals [2]. This can be readily seen in partially leached rocks that have been sectioned. The originally dark (basic) silicate minerals in the leached rim of rock are bleached of color because of the chemical reactions with the acid. Of course, when carbonate rocks are present in the oxide ore, these will be rapidly attacked by the acid (Figs. 30 and 31).
- Acid attack of gangue minerals also leads to a loss of rock integrity [15]. Both the average rock particle size and permeability to percolating leach solutions tend to decrease with extended leaching time [16]. This is a major factor preventing continued economic leaching as the mine heaps age, and it often occurs long before most of the copper has been extracted.
- The acid attack of silicate minerals is also evidenced by the high concentrations of these cations in heap leach liquors The most important oxidized copper ores are consisting mainly of [19], silicates [chrysocolla ($CuSiO_3\cdot 2H_2O$)], carbonates [malachite ($CuCO_3\cdot Cu(OH)_2$,

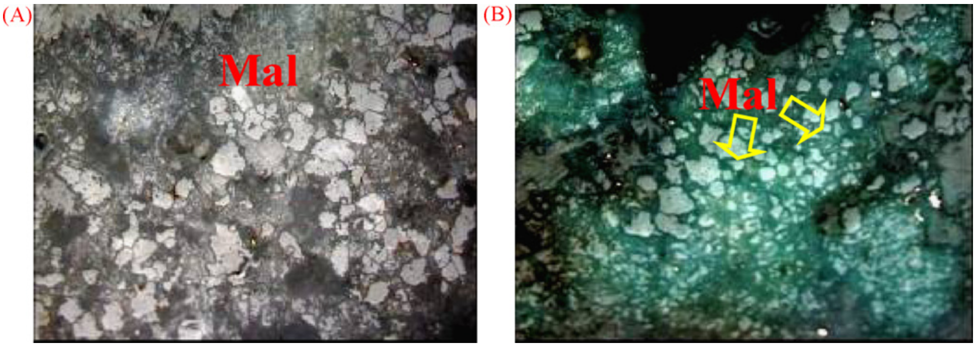

Fig. 30. (A) Photomicrograph showing malachite, which can be observed in between the silicates and near the surface before acid leaching; (B) photomicrograph of the same sample, from which the malachite has been completely preserved in acid and texture of the rock (PPL. 5×).

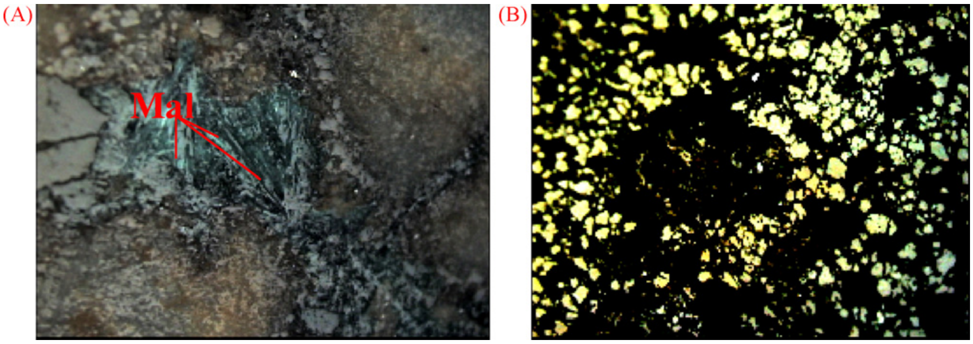

Fig. 31. (A) Malachite mineral (sample number 82532), preserved in acid leaching (PPL. 5×); (B) the yellow fragmented materials are nothing but the reflection of light through the silicate minerals in crossed nickol (XPL. 5×).

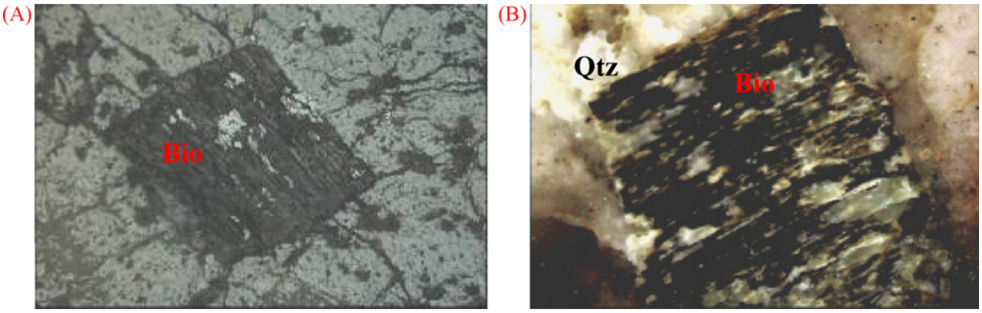

Fig. 32. (A) Microscopic image showing a biotite grain before and after acid leaching (PPL. 5×); (B) the biotite grain after acid leaching along with the quartz (XPL. 20×).

and azurite ($2CuCO_3{\cdot}Cu(OH)_2$], and chlorates [atacamite, ($Cu_2Cl(OH)_3$)] are generally treated by leaching of the crushed ore with sulfuric acid to produce the impure solutions of copper sulfate.

- As the major acid-consuming gangue mineral, the alteration chemistry of biotite is important with regard to an overall dump chemistry balance [2]. Based on structural consideration of the biotite lattice and experimental work on the dissolution of biotite (Fig. 32) under more acidic conditions than are typical of heap leaching, it is expected that biotite alters in the following manner.
- Potassium is readily exchanged by hydrogen ions or other small cations and is quickly leached from biotite. Ultimately, all of the cations from biotite other than silicon can be removed, so that the final residue would be amorphous silica [2]. These cations are not leached nearly as rapidly as potassium.
- Copper dump leaching solutions are calcium saturated. Carbonates and soluble calcium bearing silicate minerals cause precipitation of gypsum, which is very detrimental to dump permeability because the molecular volume of gypsum is much greater than the minerals it replaces.

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