ISIJ International, Vol. 56 (2016), No. 4, pp. 714–718

Ultrasound-intensified Leaching of Gold from a Refractory Ore

Gengwei ZHANG,[1,2,3)] Shixing WANG,[1,2,3)]* Libo ZHANG[1,2,3)] and Jinhui PENG[1,2,3)]

1) State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming, Yunnan, 650093 China.
2) National Local Joint Laboratory of Engineering Application of Microwave Energy and Equipment Technology, Kunming, Yunnan, 650093 China.
3) Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming, 650093 China.

(*Received on August 21, 2015; accepted on December 18, 2015*)

The effect of ultrasonic energy on the pretreatment and leaching of gold from the refractory ore was investigated. Firstly, the refractory ore was pretreated with sodium hydroxide under ultrasound. Secondly, the pretreated ore was leached under ultrasound. The leaching ratio of gold decreased with increasing of the ultrasound pretreatment time from 1 to 5 h and the NaOH concentration from 5 to 20 wt%. The ultrasound increased remarkably the leaching ratio of gold when the pretreated ore was subjected to ultrasound leaching. The results showed that the appropriate acoustic power was a requirement during the ultrasound-intensified leaching. The leaching ratio of gold increased with an increasing of NaCN concentration from 6 to 18 kg/t. The results demonstrated that the ultrasound is a viable extractive metallurgy technique in pretreatment and leaching for the refractory gold ores.

KEY WORDS: gold ore; intensified leaching; ultrasound.

1. Introduction

Cyanide leaching has been applied to separate gold from ore for over 120 years. Gold ores can be classified as free milling, complex, or refractory based on gold recovery achievable by direct cyanidation.[1)] High gold recoveries from free-milling gold can easily be obtained by conventional cyanide leaching.[2)] Refractory gold ores have the low gold extractions by the cyanidation and are arsenopyrites or pyrites containing various amounts of organic carbon.[3–5)]

The free-milling gold ores are being exhausted and the refractory ores are being discovered and processed. The number of new methods receives more and more attention from the metallurgist because its help to solve various problems during processing. The refractory ore was destroyed or attacked by extensive pretreatment prior to the cyanidations, such as oxidation by roasting,[6)] pressure oxidation,[7)] bio-oxidation,[8)] and ultrafine grinding.[9)] The new methods are only supplement for a long time due to its high cost. So, a suitable cyanidation process is often required to overcome the refractoriness and render the gold accessible to the lixiviante action of cyanide and oxygen.[10)]

To make the cyanidation process more efficient and increase the yield of gold, it is best to intensify the cyanidation process, improve production efficiency, reduces the scale of the project and save investment. Qiu Tingsheng and co-author introduced magnetic field to intensify the leaching process and evidently promoted the gold leaching.[11)] The magnetic field-intensified leaching increased the leaching ratio of gold by 33.08% compared to the conventional oxidation leaching. Yang *et al.* intensified gold leaching with heavy metals and hydrogen peroxide.[12)] They found that leaching time of gold was effectively shortened to no longer than 12 h from 16 to 24 h.

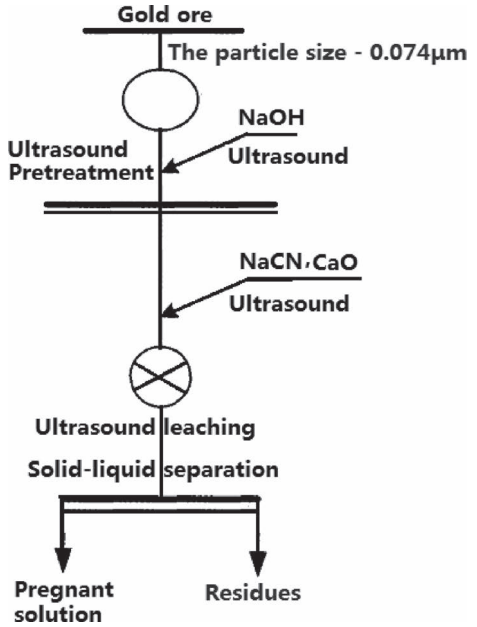
The ultrasound-intensified leaching is one of the intensified leaching processes and is becoming increasingly popular in metallurgical industry.[13,14)] The ultrasound increases dissolution rate and yields of products during leaching. The ultrasound-intensified leaching accelerates kinetics and reduces reagent consumption. The ultrasound-intensified leaching requires only the presence of a liquid to transmit its energy compared with other new technologies that require some special attribute of the system to produce an effect. The effect of ultrasound on the acid leaching of copper from copper converter slag,[15)] ammonium leaching of zinc from calamine[16)] and dissolution of colemanite in sulphuric acid[17)] have been investigated.

The aim of this study was to investigate the effect of ultrasonic energy on the leaching of gold from the refractory gold ore. **Scheme 1** showed that the processes included two stages. The first stage is ultrasound pretreatment with sodium hydroxide. The second stage is ultrasound leaching. The effects of parameters such as ultrasound pretreatment time, NaOH concentration, NaCN concentration and ultrasound power were studied.

2. Experiment

The refractory gold ores was obtained from Yunnan gold mountain co., Ltd, China. The gold ores was crushed, ground, and then sieved by using ASTM standard sieves. The gold ore with a particle size -0.074 μm was used in all experiments. The chemical composition is given in **Table 1**.

* Corresponding author: E-mail: wsxkm@sina.com
DOI: http://dx.doi.org/10.2355/isijinternational.ISIJINT-2015-476



Scheme 1. The processes of the ultrasound-intensified leaching of gold from a refractory ore.

Table 1. The chemical composition of gold ore.

Au ($\times10^{-6}$)	Ag ($\times10^{-6}$)	Cu (%)	Pb (%)	Zn (%)	Fe (%)	S (%)
3.23	2.12	0.060	0.018	0.141	6.394	1.244
As (%)	Sb (%)	CaO (%)	MgO (%)	Al_2O_3 (%)	SiO_2 (%)	C (%)
0.159	0.122	4.529	3.285	14.901	66.254	2.893

It contains Au (3.23 ppm) and silicate *etc.*

The experiments were carried out in a 1 000 ml beaker equipped with mechanical agitation, filling gas pipe and an ultrasonic generator that had cup horn type piezoelectric ultrasonic generator with 20 kHz frequency.

400 g gold ore and 800 ml water were introduced into the reactor equipped with ultrasonic generator with 1 kW power and 20 kHz frequency. Then, 60 g sodium hydroxide (15 wt% of gold ore) was added to the solution and mechanically stirred at 600 rpm under room temperature. After mixed, the ultrasonic generator was started and the flow rate of air was 120 L/h. After ultrasound pretreatment was hold for 5 h, the gold ore slurry was filtered, washed with water three times and dried at 120°C.

The second stage is ultrasound leaching. 150 g the pretreated gold ore and 600 ml water were introduced into the reactor equipped with ultrasonic generator with 180 W and 20 kHz frequency. The slurry was mechanically stirred at 600 rpm under room temperature. 1.8 g sodium cyanide and 1 g calcium oxide were added to the slurry. The concentration of sodium cyanide is about 12 Kg/t. The ultrasonic generator was started and the flow rate of air was 120 L/h. The leaching process was lasted for 5 h. At different time intervals, sample solutions were taken and filtered. The filtration was used to measure gold leaching ratios by an atomic absorption spectrometer (Shimadzu AA-6800). The solid residues were washed, dried and analyzed by XRD (model: Philips X'pert with CuKa radiation) and scanning electron microscope (JSM-5800, made by JEOL, Japan) with an energy dispersive X-ray spectrometer (Link ISIS 300, Oxford Instrument, UK).

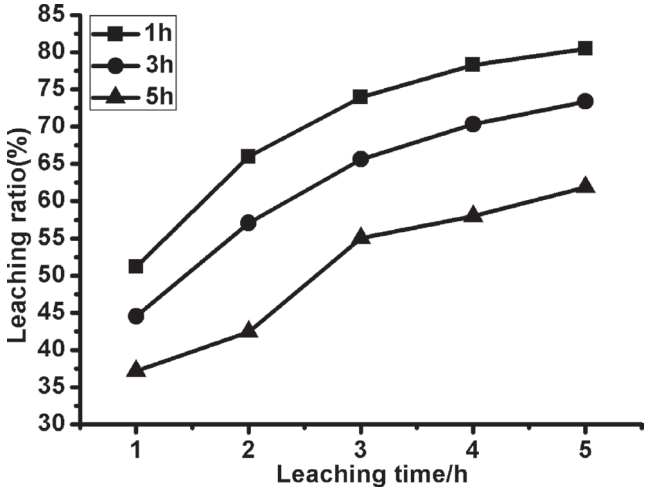


Fig. 1. Effect of ultrasound pretreatment time on the ultrasound leaching ratio.

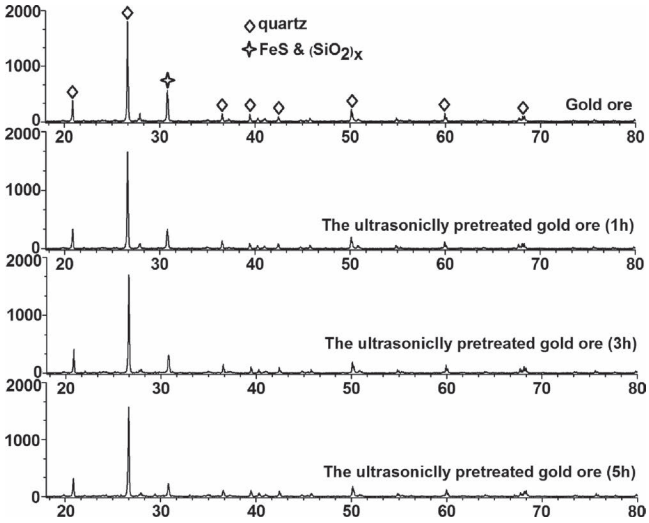


Fig. 2. XRD of the gold ore and the pretreated ores.

3. Results and Discussion

3.1. Effect of Ultrasound Pretreatment

3.1.1. Effect of Ultrasound Pretreatment Time

The effect of the ultrasound pretreatment time was studied when the NaOH concentration was 15 wt%. Increasing of the ultrasound pretreatment time had a negative effect on the leaching ratio of gold (**Fig. 1**). For example, the leaching ratio of gold decreased from 80.5 to 61.9% with increasing pretreatment time from 1 to 5 h when the pretreated ores were ultrasonically leached for 5 h. However, Fig. 1 also showed that the leaching ratio of gold increased with increasing the ultrasound leaching time.

Figure 2 is XRD of the gold ore and the pretreated ore. Figure 2 shows that the chemical composition of the gold ore is SiO_2 and FeS. The peak intensity at $2\Theta=30.811$ decrease with increasing the ultrasound pretreatment time. The peak at $2\Theta=30.811$ is the characteristic peak of (SiO_2) x and FeS. Under ultrasound, FeS reacted with NaOH prior

to $(SiO_2)x$. After FeS reacted with NaOH, the leaching ratio of gold was improved. However, SiO_2 also reacted with NaOH to generate sodium silicate with increasing the ultrasound pretreatment time. Sodium silicate absorbed gold and decreased the apparent leaching ratio of gold. Provis *et al.* indicated that the newly-formed silica gels adsorbed gold from the solution in alkaline media.[18] **Figure 3** is the morphology change of the ore sample with the ultrasound time when the NaOH concentration is 15%. In the SEM micrographs of these samples it can be observed that the size of the particles increased obviously with the ultrasound time. Energy diffraction analysis (EDX) was also carried out for selected areas as indicated by rectangles in Fig. 3(d). Energy diffraction analysis showed that the particles are mainly made of silicon and oxygen (**Fig. 4**). These results showed that silica-gel was formed after long-time ultrasonic pretreatment.

Oktay Celep and co-authors implemented sodium hydroxide pretreatment for refractory antimonial gold and silver ores.[19] They indicated that the extraction ratio of gold

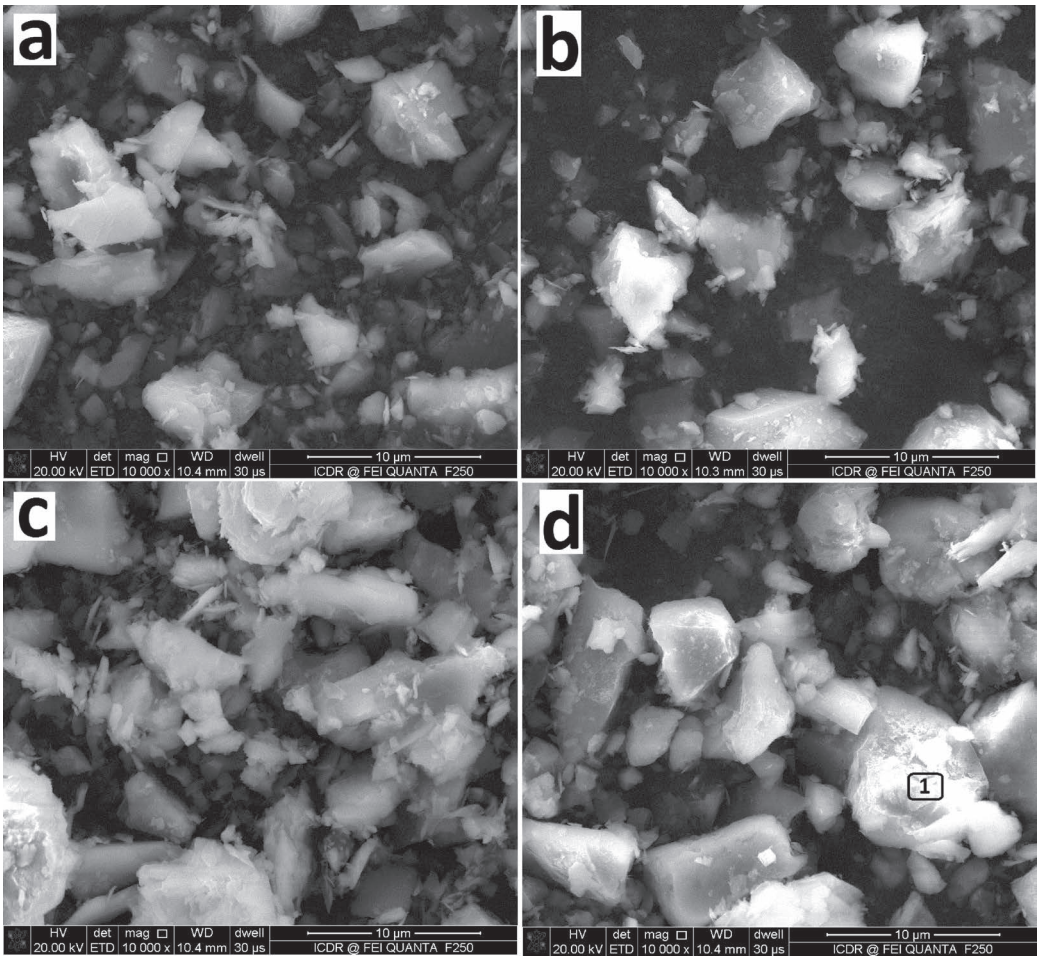


Fig. 3. Effect of the ultrasound time on the morphology of the ore sample when the NaOH concentration is 15%. a) the original ore, b) 1 h, c) 3 h, d) 5 h.

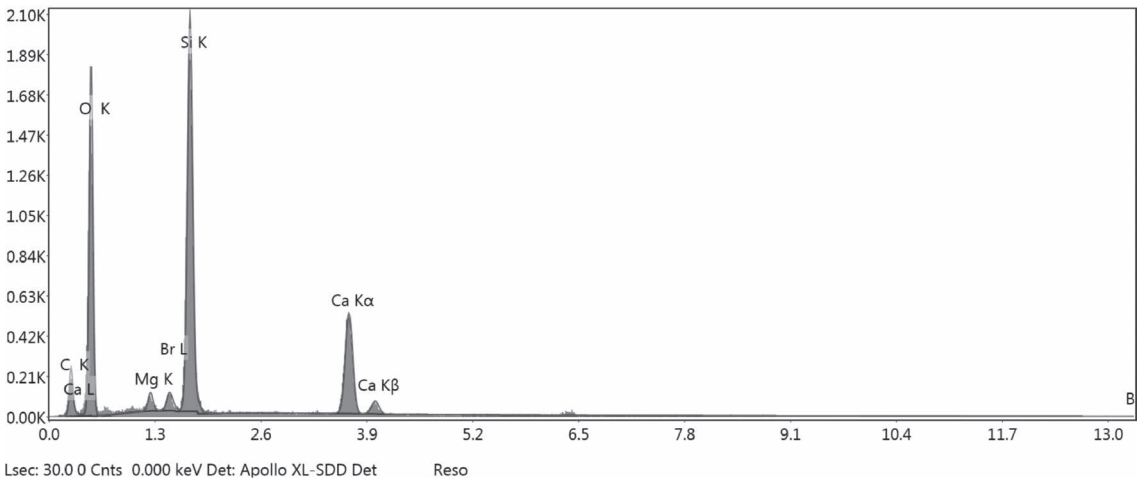


Fig. 4. Energy diffraction analysis for selected areas as indicated by rectangles in Fig. 3(d).

was improved from 49.3% to 85.4% after pretreated for 120 min with 3 mol/L NaOH at 80°C. Our experiment was carried out under room temperature and the highest gold leaching ratio was 80.5% after ultrasonically pretreated for 1 h. However, the leaching ratio of gold was 7.3% when the gold ore did not pretreated by ultrasound. The results showed that ultrasound pretreatment was effective for the refractory gold ore. The high Au extractions after ultrasound pretreatment confirmed that a large proportion of the gold was refractory in nature because of their unliberatied (*i.e.* locked) inclusions.

3.1.2. Effect of the NaOH Concentration

The effect of the NaOH concentration was studied when the ultrasound pretreatment time is 3 h. The leaching ratio of gold decreased with increasing the NaOH concentration (**Fig. 5**). For example, the leaching ratio of gold decreased from 82.5 to 72.1% with increasing the NaOH concentration from 5 to 20 wt.% when the pretreated ores were ultrasonically leached for 5 h. However, Fig. 5 also showed that the decrease of the gold leaching ratio is not obvious when the concentration of NaOH increase from 15 to 20 wt.%. The increasing of the concentration of NaOH accelerated the reaction between SiO_2 and NaOH and generated sodium silicate. Sodium silicate absorbed gold and decreased the apparent leaching ratio of gold.[18] Figure 3 showed that the small amounts of NaOH could improve the leaching ratio of gold.

The intensive and active function of ultrasound pretreatment included 1) enhancing mass transfer conditions, 2) freshly liberated FeS particles are subjected to O_2 and NaOH, 3) passivation is not possible under the high shear conditions.[20,21] The results demonstrated that ultrasound pretreatment with sodium hydroxide is a viable extractive metallurgy technique for the processing of refractory gold ores.

3.2. Effect of the Ultrasound Intensified Leaching

3.2.1. Effect of Ultrasonic Power on the Leaching Ratio

In order to improve the gold leaching ratio, application of ultrasonic power might be another viable option. The gold ore pretreated with 15 wt.% NaOH and 3 h was subjected to ultrasonically leach with different ultrasound power (1 kW, 180 W and 120 W).The leaching ratio of gold was presented in **Fig. 6**. The leaching ratio of gold increased with increasing the leaching time. After ultrasonically leaching for 5 h, the highest leaching ratio of gold (73.4%) was obtained at 180 W. However, the leaching ratio of gold only was 40.3% when the pretreated gold ore was leached without ultrasound. The results showed that ultrasound leaching is an effective way for extracting gold from ore. Thousands of tiny bubbles appear when ultrasound is applied into the liquid. This breakup process of the liquid is called acoustic cavitation. Rapid adiabatic compression of gases and vapours within the bubbles or cavities produces extremely high temperatures and pressures. The extremely high temperatures and pressures at the interface between a solution and a solid matrix, combining with the oxidative energy of radicals (H atoms and OH radicals) created during sonolysis, result in high leaching ratio. Penn *et al.* found that ultrasound waves reduced the mass transfer boundary layer and therefore efficiently increased the mass transfer.[22]

The leaching ratio of gold is higher under 180 W than that under 120 W. The leaching ratio of gold is higher under 1 kW than these under 120 and 180 W when the ultrasound

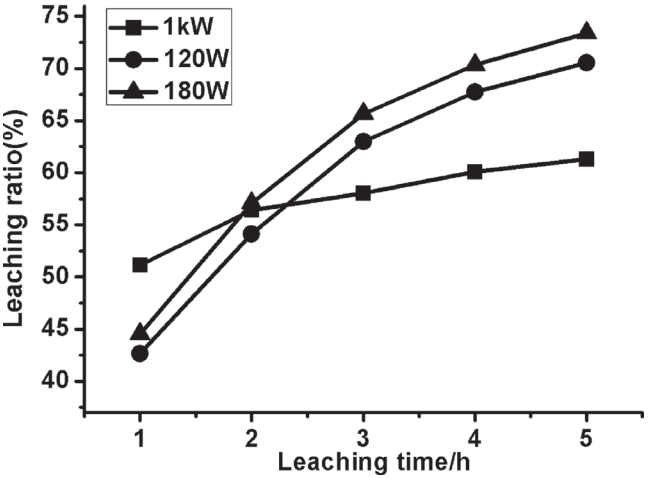


Fig. 6. Effect of ultrasonic power on the ultrasound leaching ratio.

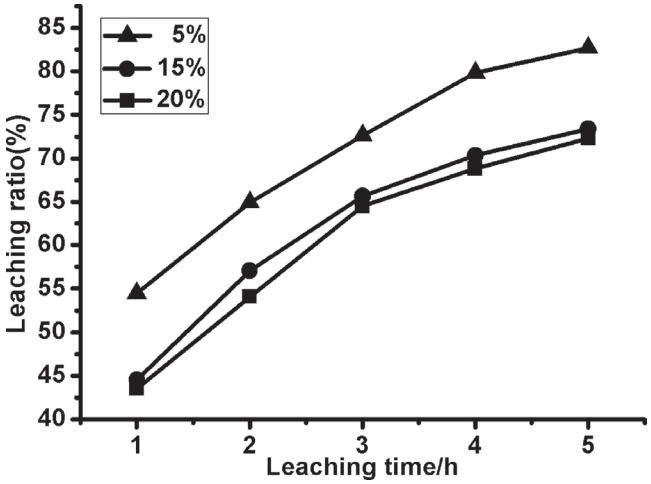


Fig. 5. Effect of the NaOH concentration on the ultrasound leaching ratio.

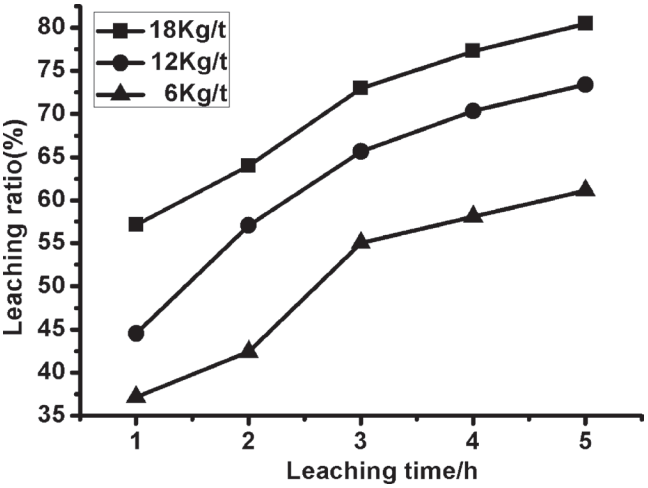


Fig. 7. Effect of the NaCN concentration on the ultrasound leaching ratio.

leaching time is less than 2 h. However, the leaching ratio of gold is lower under 1 kW than these under 120 and 180 W when the ultrasound leaching time is more than 2 h. The more plausible explanation is that the increased acoustic power results in decomposition of CN^-. Yazıcı demonstrated that a high ultrasonic power input was required for the degradation of cyanide.[23] Because ultrasonic irradiation of aqueous solutions leads to the formation of powerful oxidants such as $HO\cdot$ radicals and H_2O_2 and H_2O_2 is an effective oxidant industrially used for the destruction of cyanide species [Eq. (1)] present in the effluents of gold/silver leaching.

$$CN^- + H_2O_2 \rightarrow CNO^- + H_2O \quad \cdots\cdots\cdots\cdots (1)$$

Therefore, the appropriate acoustic power is a requirement during the ultrasound leaching.

3.2.2. Effect of NaCN Concentration on the Leaching Ratio

The effect of the NaCN concentration on the leaching ratio of gold from the gold ore pretreated with 15 wt.% NaOH and 3 h was examined. The results are presented in **Fig. 7**. It implies that the initial rate of gold leaching is fast and almost sensitive to NaCN concentration. The leaching ratio increases with an increase of the NaCN concentration from 6 to 18 kg/t. Moreover, the leaching rate decreases with an increase in the leaching time. It is in agreement with the results reported earlier.[24,25] When the NaCN concentration is 18 kg/t, the obtained highest leaching ratio is 80.7% after ultrasound leached for 5 h.

4. Conclusions

We investigated the effect of ultrasonic energy on the pretreatment and leaching of the refractory gold ore. These results showed that the increase of the ultrasound pretreatment time and the concentration of the NaOH had a negative effect on the leaching ratio of gold. Te smaller amounts of NaOH and the short ultrasound pretreatment time could improve the leaching ratio of gold. We found that the ultrasound intensified leaching increase remarkably the leaching ratio of gold, the appropriate acoustic power is a requirement during leaching and the leaching ratio increases with an increase of the NaCN concentration from 6 to 18 kg/t. Therefore, the ultrasound has a broad application prospect in pretreatment and leaching processing of refractory gold ores.

Acknowledgements

The authors are grateful for the financial support from the National Natural Science Foundation (No. 51104073).

REFERENCES

1) B. Nanthakumar, C. A. Pickles and S. Kelebek: *Miner. Eng.*, **20** (2007), 1109.
2) D. M. Muir: *Miner. Eng.*, **24** (2011), 576.
3) L. R. P. Andrade Lima, L. A. Bernardez and L. A. D. Barbosa: *J. Hazard. Mater.*, **150** (2008), 747.
4) M. Benzaazoua, P. Marion, F. Robaut and A. Pinto: *Mineral. Mag.*, **71** (2007), 123.
5) J. A. Brierley: *Hydrometallurgy*, **71** (2003), 13.
6) G. Xue and W. Ren: *Chin. J. Nonferrous Met.*, **3** (2007), 44.
7) F. P. Gudyanga, T. Mahlangu, R. J. Roman, J. Mungoshi and K. Mbeve: *Miner. Eng.*, **12** (1999), 863.
8) G. Ofori-Sarpong, K. Osseo-Asare and M. Tien: *Miner. Eng.*, **24** (2011), 499.
9) R. E. Browner and K. H. Lee: *Miner. Eng.*, **11** (1998), 813.
10) S. Ubaldini, R. Massidda and C. Abbruzzese: Proc. 5th Int. Mineral Processing Symp., Turkish Mining Development Foundation, Istanbul, Turkey, (1994), 403.
11) T. Qiu, S. Xiong and Q. Xia: *Metal Mine.*, **342** (2004), 32.
12) Y. Yang, T. Jiang, Y. Guo, G. Li and B. Xu: *Trans. Nonferrous Met. Soc. China*, **20** (2010), 903.
13) K. M. Swamy, K. Sarveswara Rao, K. L. Narayana, J. S. Murty and H. S. Ray: *Miner. Process. Extr. M.*, **14** (1995), 179.
14) K. L. Narayana, K. M. Swamy, K. J. Sarveswara Rao and S. Murty: *Miner. Process. Extr. M.*, **16** (1997), 239.
15) A. V. Bese: *Ultrason. Sonochem.*, **14** (2007), 790.
16) A. S. Slaczka: *Ultrasonics*, **24** (1986), 53.
17) H. Okur, T. Tekin, A. K. Ozer and M. Bayramoglu: *Hydrometallurgy*, **67** (2002), 79.
18) S. Mohammadnejad, J. L. Provis and J. S. Van: *J. Int. J. Miner. Process.*, **100** (2011), 149.
19) O. Celep, I. Alp, D. Paktunç and Y. Thibault: *Hydrometallurgy*, **108** (2011), 109.
20) I. De La Calle, N. Cabaleiro, M. Costas, F. Pena, S. Gil, I. Lavilla and C. Bendicho: *Microchem. J.*, **97** (2011), 93.
21) F. Xie, H. Li, Y. Ma, C. Li, T. Caia, Z. Huang and G. Yuan: *J. Hazard. Mater.*, **170** (2009), 430.
22) R. Penn, E. Yeager and F. Hovorka: *J. Acoust. Soc. Am.*, **10** (1959), 1372.
23) E. Y. Yazıcı, H. Deveci, I. Alp and T. Uslu: *Desalination*, **216** (2007), 209.
24) H. Kasaini, K. Kasongo, N. Naude and J. Katabua: *Miner. Eng.*, **21** (2008), 1075.
25) X. Dai and M. I. Jeffrey: *Hydrometallurgy*, **82** (2006), 118.