Hydrometallurgy 150 (2014) 99–106

Contents lists available at ScienceDirect

Hydrometallurgy

journal homepage: www.elsevier.com/locate/hydromet

A novel method using a complex surfactant for in-situ leaching of low permeable sandstone uranium deposits

Kaixuan Tan *, Chunguang Li, Jiang Liu, Huiqiong Qu, Liangshu Xia, Yang Hu, Yongmei Li

School of Nuclear Resources Engineering, University of South China, Hengyang 421001, China

ARTICLE INFO

Article history:
Received 28 January 2014
Received in revised form 29 September 2014
Accepted 2 October 2014
Available online 21 October 2014

Keywords:
Complex surfactant
Leaching kinetics
Resin adsorption and elution
Low permeable sandstone uranium deposit
In-situ leaching of uranium mining

ABSTRACT

Applications of a complex surfactant developed in-house to in-situ leaching of low permeable sandstone uranium deposits are described based on results from agitation leaching, column leaching, resin adsorption, and elution experiments using uranium containing solution from the in-situ leaching site. The results of agitation leaching experiments show that adding surfactant with different concentrations into leaching solution improves the leaching rate of uranium. The maximum leaching rate of uranium from agitation leaching reached 92.6% at an added surfactant concentration of 10 mg/l. Result of column leaching experiment shows that adding surfactant with varying concentrations into leaching solutions increased the permeability coefficient of ore-bearing layer by 42.7–86.8%. The leaching rate of uranium from column leaching increased by 58.0% and reached 85.8%. The result of kinetic analysis shows that for the extraction of uranium controlled by diffusion without surfactant the apparent rate constant 0.0023/d changed to 0.0077/d for the extraction with surfactant controlled by both diffusion and surface chemical reactions. Results from resin adsorption and elution experiments show that there was no influence on resin adsorption and elution of uranium with an addition of 50 mg/l surfactant to production solution from in-situ leaching. The adsorption curve, sorption capacity of resin, recycling of resin remained the same as without adding any surfactant. Introducing complex surfactant to leaching solution increased the peak concentration of uranium in eluents, reduced the residual uranium content in resin, and promoted the elution efficiency. The method of using a complex surfactant for in-situ leaching is useful for low permeable sandstone uranium deposits.

1. Introduction

Uranium is the key element and the raw material to fuel nuclear reactors for the use of nuclear energy. It is a challenge for the uranium mining and processing industry to provide an adequate supply of nuclear fuel to sustain the nominal growth rate for nuclear power (IAEA, 2009). An unconventional mining technique, called in-situ leaching mining or solution mining, has been developed in the last few decades. With this technique, the leaching solutions are directly injected into underground ore bed so the metal or the compound can react between solutions and minerals. The in-situ leaching process provides an effective and low cost method for ore production (IAEA, 2004). Low grade ore bodies that are uneconomical by conventional means may become economically viable using in-situ leaching method. Thus, the in-situ leaching technology has become a major production method of uranium and has been employed by the mining industry in many countries such as in the USA, former USSR, Bulgaria, Czechoslovakia, German Democratic Republic, and Australia as well as in northwest China (Li, 2005; Tan et al., 2004; Wang et al., 1998).

In recent years, abundant sandstone uranium deposits have been explored in Xinjiang, Yunnan, Shanxi and Inner Mongolia (Chen, 2006). Nevertheless, many of them are less suitable for employ in-situ leaching because of the low permeability. The permeability in porous rock is structurally characterized by porosity, pore size, and pore connectivity. The permeability of leaching solution migration in-situ leaching process is related to surface tension. A majority of leaching solution migrates along the well-connected pore channels. Some penetrate through the hairline cracks and the capillaries at a lower rate. A minority of leaching solution moves in the tiny pore or poorly connected pores at very low rate or in dead zone at zero flow rate. The permeable ore-bearing layer could be damaged severely in the production process of in-situ leaching mining such as drilling, washing-hole supplying, and chemical leaching. These factors result in decrease of both the injection and the extraction, slow down significantly the normal operation of the in-situ leaching mining (Fan et al., 2004; Tan et al., 2006). Apparently, it is critical to enhance the permeability of the ore-bearing layer in the leaching of uranium.

Surfactants have been widely applied in many fields such as mining, petroleum, industrial, agriculture, food, cosmetics, and pharmaceutical (Li and Fang, 2002) because they can reduce the surface or interface tension of the materials significantly at a very low concentration. Owusu

* Corresponding author. Tel.: +86 734 8282634.
E-mail address: nhtkx@126.com (K. Tan).

http://dx.doi.org/10.1016/j.hydromet.2014.10.001
0304-386X/

et al. (1995) used surfactant as dispersants to increase extraction rates of zinc and iron from liquid sulfur. Karavasteva (2001) reported a surfactant mixture improved deposition of impurities during the neutral leaching stage due to surface tension reducing of agents and complexing agents. Li (2005) showed that N-alkylcaprolactams have a stronger affinity to U (VI) than to Fe (III) and the system can attain a high separation factor to U (VI) in the uranium purification process. Marek et al. (2009) removed Polychlorinated Biphenyls (PCBs) from real contaminated soil with a surfactant solution flush. Zhuo et al. (2009) reported that the elemental sulfur produced from the leaching reaction was not completely oxidized and was dissolved only partially even in the presence of bacteria with a limited dissolution rate of marmatite. However, the addition of Ortho-Phenylene-Diamine (OPD) accelerated the oxidation of elemental sulfur in the presence of bacteria, thus enhancing the bioleaching rate of marmatite. Myroslav et al. (2010) modified the adsorption capacities of diatomite by adding a surfactant that improved the extraction efficiency of uranium from its aqueous solution. Recently, continuous applications of surfactants have been reported (Mohammad et al., 2012; Okoliegbe and Agarry, 2012; Samanta et al., 2013).

Although many applications of surfactant were discussed in literatures, little information on the application of surfactant for extraction of uranium from low permeable sandstone uranium deposits is available in detail. Surfactant influence on the resin adsorption and elution of uranium has not been studied and reported even though it is an important aspect of the production process. This paper reports the findings of using a compound surfactant for increasing the permeability and the leaching rate of uranium in low permeability sandstone in in-situ leaching at a mining field of Xinjiang, China, by agitation leaching and column leaching experiment.

2. Basic feature of uranium deposits and samples

The uranium deposit, located west of the southern margin of Yili basin, Xinjiang, China, is a sandstone roll-type deposit associated with an interlayer oxidation zone. The ore-containing stratum in the deposit is a Shuixigou Group of lower-middle Jurassic. The ore belt in east–west extension is 2800 m and stretches 10–200 m along the tendency with "volume" shaped, buried between a depth of 220–250 m. The major body of the roll-type deposit is thick, great grade, and high permeability. It is in good condition for in-situ production and it is a site currently in production. The wings of ore body are relatively thin, poor grade, and low permeability, but with abundant resources. They are excellent locations for testing in-situ leaching with complex surfactant and that is a great potential for uranium production.

All samples were collected from the drilling at the wings of the ore body. They are medium-fine sandstone in color of gray to dark gray with inequigranular texture and massive structure. The uranium minerals are mainly pitchblende (98%) and with minor coffinite and brannerite in clay and silty cement, in the forms of scattered or small disseminated deposits. Other metal minerals are pyrite, hematite, goethite, and sphalerite. Nonmetallic minerals with the average volume fraction are quartz 70%, feldspar 12%, kaolinite 8%, illite 5%, and montmorillonite 3%. Carbonate mineral content is generally low except in a few locations is relatively higher. Table 1 is a summary of chemical composition of three sample groups. The majority of the composition is SiO_2 and the CaO is pretty low (0.15–0.16%) with the exception of sample T-206 (2.13%). Ore mineral and chemical compositions are suitable for using the acid leaching method.

Table 2 shows data for pore structure of ores from the mercury intrusion method and based on the Washburn equation,

$$Pr = -2\sigma\cos\theta \quad (1)$$

where P is the pressure required to force mercury into the pore (additional pressure imposed to mercury) in MPa; r is the radius of pore in μm; σ is the surface tension of the mercury in N/m; θ is the contact angle mercury to the material.

For this experiment $\sigma = 480$ N/m and $\theta = 130°$, the Eq. (1) becomes

$$P = \frac{0.617}{r} \quad (2)$$

The pore volume and surface area of different pore size were calculated from measuring the mercury content of uranium ore pore space after degassing treatment under external pressure. The experimental apparatus is fully automatic mercury porosimeter Autopore IV 9510. Three sandstone samples from a leaching uranium mine in Xinjiang are numbered as T-202, T-1406, and T-206. Although the porosity of the uranium ore is not very low (between 19.47% and 23.40%), the average pore size is small (33.0–104.9 μm). The percent of the tiny pore (diameter smaller than 0.1 μm) is large, 24.1–70.7% of the total pore volume. The percent of the larger pore (diameter larger than 1 μm) is smaller, 22–41.9% of the total pore volume. These implicate the leaching rate of uranium and permeability will be significantly increased if the leaching solution infiltrates to the tiny pore.

3. Experimental methods

3.1. Complex surfactant

A complex surfactant (CS) was prepared in house for experiments. It was a product mixing from octyl-phenyl polyoxyethylene ether (OP-10) and perfluoroalkyl sulfuryl fluoride (FSO) in a certain proportion. Surface tension of solution with different concentrations were measured by a JZHY-180 interfacial tensiometer using circular ring heave liquid membrane method at a temperature of 20 ± 0.2 °C. Fig. 1 shows that the surface tension approaches to a stable value 16.5 mN/m with a high surface activity at 0.1% aqueous solution. The complex surfactant exhibits more synergistic effects, compared to the single surfactant.

3.2. Agitation leaching experiment

In the agitation leaching experiment, the sample T-202 was used to investigate the effect of the complex surfactant with various concentrations on the leaching of uranium. The surfactant with varied concentrations from 0 to 300 mg/l was added to dilute sulfuric acid leaching solution. The concentration of sulfuric acid of the solution was 10 g/l and solid-to-liquid ratio was 1 to 3. The ore sample placed in a container weighted 40 g and the volume of leaching liquid was 120 ml. The full rotation time for the sample was 24 hours with the temperature at 30 °C for the shaking table. After extraction and filtration, ore sample was washed with 5 g/l dilute sulphuric solution. The volume, content of uranium, pH and redox potential (Eh) of extracting solution and

Table 1
Chemical composition of ores (%).

Sample ID	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	LOI	U
T-202	85.59	6.25	1.28	0.07	0.28	0.15	1.02	1.99	0.04	2.04	0.036
T-1406	81.89	6.63	1.71	0.05	0.34	0.16	1.02	2.42	0.06	3.22	0.029
T-206	79.71	6.69	2.27	0.05	0.66	2.13	1.18	2.04	0.10	5.53	0.054