THE FUNDAMENTAL RESEARCH AND INDUSTRIAL APPLICATION OF THE CO2 AND O2 IN SITU LEACHING PROCESS IN CHINA

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1. INTRODUCTION

Owing to the advantages of reduced chemical reagent consumption and groundwater pollution, the CO2 and O2 in situ leaching (ISL) process became one of the important research fields in uranium mining. China was the second country in world to apply the CO2 and O2 ISL process in uranium production. This extended abstract describes the development and characteristics of the CO2 and O2 ISL process in China, including the main principles, technological processes, well-field design, production well construction and uranium processing. Finally, the industrial application status and development potential of the CO2 and O2 ISL process in China is summarized.

2. DISCOVERED URANIUM RESOURCES STATUS AND DISTRIBUTION

There are 21 known uranium ore deposits in China, distributed in throughout 13 provinces, including Inner Mongolia, Xinjiang, Jiangxi and Guangdong. Since 1994, the exploration targets changed from those deposits suited to conventional mining in southern China to deposits in northern China which are amenable to in situ leaching. A number of medium–large scale sandstone uranium deposits have been discovered. As a result, the U resources/reserves have increased rapidly since 2000.

China’s identified resources (reasonably assured and inferred) amount to 370 900 tU, although the potential for more than 2 million tU is predicted. Uranium deposit types and their share of uranium resources in 2016 were listed as follows: sandstone (56.9%: 210 000 tU), granite (23.6%: 87 000 tU), volcanics (11.1%: 41 000 tU), carbonaceous–siliceous–pelitic (7.5%: 28 000 tU) and others (0.9%: 3500 tU). The sandstone hosted uranium resources mainly come from six basins located in northern China: Yili, Turpan-Hami, Ordos, Erlian, Songliao and Bayin Gobi. The Yili and Songliao Basins are the key and potential areas for future development.

Most sandstone uranium deposits in China are complex and about 70% of these are have adverse factors such as high carbonate content (>2.0% CO2), low permeability (<0.17 m/d), low uranium grades (~0.03%) and high salinity groundwater (total dissolved solids 5–10 g/L).

3. BRIEF DEVELOPMENT HISTORY OF CO2 AND O2 ISL

The CO2 and O2 in situ leaching process for uranium has been under development in China since 2000. Some CO2 and O2 leaching experiments have been carried out to simulate CO2 and O2 leaching characteristics and some technical parameters have been obtained.

Since 2006, a field test and industrial tests have been implemented at the Qianjiadian uranium deposit in the Songliao Basin, Inner Mongolia. The depth of orebody is 251.8–298.31 m and it has a thickness of 6.46–15.75 m, a mean grade of 0.025% U and a mean uranium content of 3.95 kg/m². The modes of occurrence of uranium in the Qianjiadian deposit include absorbed uranium, uranium minerals and uranium-bearing minerals. The ratio of U(VI):U(IV) is 0.266:1.116 with an average value of 0.761.
The permeability coefficient of the ore-bearing aquifer is 0.025–0.223 m/d and the depth of confined water is 5.29–7.06 m. The type of water (chemistry) is a combination of HCO$_3$–Na and HCO$_3$–Cl–Na with a salinity range of 3.10–5.7 g/L, a pH of 7.2–8.4, and an Eh of 100–200 mV.

An industrial scale well field had been established, including 10 production wells and 32 injection wells. Depending on orebody geometry and surface topography, 7 spot well patterns and a 35 m well spacing were used. For injection wells, the average flow rate is 2.8 m$^3$/h, the equivalent average flow rate for recovery wells is 8.1 m$^3$/h.

4. CO$_2$ AND O$_2$ LEACHING PROCESS

Both gaseous oxygen and carbon dioxide are added to groundwater to produce lixiviant. Oxygen is typically added to maintain the strongly oxidizing conditions required to oxidize tetravalent uranium in ore minerals to the hexavalent stage. The oxygen concentrations vary from 150 mg/L to 500 mg/L for different leaching stages, depending on uranium concentration and the dissolved oxygen of the lixiviant. Carbon dioxide is added for pH control and increasing bicarbonate concentration. Carbon dioxide concentrations vary from 100 mg/L to 300 mg/L.

After 2 years of operation, uranium recovery has been up to 53.1% with a liquid:solid ratio of 2.64, while average uranium concentration was about 32 mg/L. Specific consumption of CO$_2$ was 10.8 t/tU and specific consumption of O$_2$ was 12.0 t/tU. The flow rate of the recovery well and injection well remained stable, which indicated that calcium carbonate scaling was not generated, which may otherwise adversely impact the field test.

Uranium mobilization and processing excess water must be properly managed. The production wells extract slightly more water than is re-injected into host aquifer. The production bleed is more than 0.3–1.0% of the circulation rate. The main purpose is to maintain the negative balance and help to minimize the potential movement of lixiviant.

Some technical parameters include the following:

- Well pattern: 5 spot and 7 spot;
- Well-spacing: 30–35 m;
- Drilling hole structure: gravel filling type;
- Depth: 240–320 m;
- Lixiviant: 100–300 mg/L CO$_2$ + 150–500 mg/L O$_2$;
- Recovery rate: >75%;
- Uranium extraction: ion exchange process with fixed bed column;
- Water waste treatment: reverse osmosis and evaporation ponds.

5. URANIUM PROCESSING

The ion exchange resin D261 is widely used in ISL projects in China and the ion exchange circuit is accomplished in two fixed bed columns in series. On the basis of average uranium concentrations (about 32 mg/L), more than 97% of the uranium is extracted during the ion exchange process. The stream exiting the lixiviant columns normally contains less than 0.1 mg/L. Before entering the ion exchange columns, CO$_2$ was added into the pregnant solution with the concentration varying from 100 to 300 mg/L. The purpose is to maintain the pH in the range 6.8–7.2 and increase the saturated resin load.

The elution process is accomplished in columns arranged in series by contact of the resin with a mixed solution of sodium chloride and sodium bicarbonate, thus obtaining a pregnant eluant solution with about 35–50 g/L. Typical operational parameters are 80–120 g/L of sodium chloride and 10–20 g/L sodium...
bicarbonate. The recovery rate of the elution process is normally around 99.9%. After enough pregnant eluant solution is obtained, it is moved to the precipitation circuit.

In the precipitation circuit, the pregnant eluant is typically acidified using hydrochloric acid to destroy the uranyl peroxide. The pH of the pregnant eluant decreased to about 3.0–4.0 and was maintained for 4 h, accompanied by stirring. Caustic soda is then added to precipitate the uranium as sodium diuranate at a pH of 6.0–7.0. Finally, the resulting slurry is sent to a plate-and-frame filter press where it is filtered and washed. According to the natural uranium product quality standard, the uranium content is required to be equal to or exceed 60% in solid material and the water content is required to be equal to or less than 30%.

During the process, some liquid waste is generated which may contain elevated concentrations of radioactive and chemical constituents. Reverse osmosis is commonly used to segregate contaminants. Through the reverse osmosis process, two fluids were yielded: clean water (about 70% Cl <350 mg/L) that can be reinjected into the aquifer and brine (about 30%) which is sent to the evaporation pond.

6. **ISL MINES PRODUCTION STATUS AND POTENTIAL APPLICATION**

Owing to the advantages of low operating cost, shortened loading period and reduced environment pollution, the proportion of uranium produced by in situ leaching mines has increased rapidly. In China, in situ leaching production dominates uranium production, accounting for 65.6% of production in 2016, exceeding both heap leaching and conventional mining, the second and third principal modes of extraction.

In the period 2006–2009, the first CO$_2$ and O$_2$ ISL project came into commercial operation at the Qianjiadian uranium deposit in Inner Mongolia. As regards the ongoing discovery of ISL amenable sandstone hosted uranium resources, two CO$_2$ and O$_2$ ISL mines have now been put into operation respectively, in the Songliao and Yili Basins. Other two mines are undergoing pilot scale tests in the Erdos and Erlian Basins. These will be put into production soon.

China National Nuclear Corporation has shut down some high cost underground and open pit uranium mines in southern China, and instead is focusing on the development of ISL amenable sandstone uranium resources in northern China and on plans to build three 1000 tU/year ISL mines by 2020.

7. **CONCLUSION**

Owing to successful application results and strict environmental requirements, CO$_2$+O$_2$ ISL has become a priority option and indeed the only option for sandstone type uranium deposits with high carbonate contents and high salinity groundwater. According to the development plan, about 90% of natural uranium production will be provided by ISL, especially by the CO$_2$ and O$_2$ method. Some large scale and ‘green’ mines are undergoing planning and implementation.