Hydrometallurgy 146 (2014) 119–127

Contents lists available at ScienceDirect

Hydrometallurgy

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

Continuous leaching of uranium from an Indian ore: Residence time scale up and heat effects

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ARTICLE INFO

Article history:
Received 11 October 2013
Received in revised form 6 March 2014
Accepted 14 March 2014
Available online 2 April 2014

Keywords:
Continuous leaching
Uranium
Pyrite
Residence time distribution
Heat effects

ABSTRACT

Commercial level operation of uranium leaching from the ores is carried out as a continuous process because of techno-economical benefits. Batch kinetic data of uranium leaching from a lean ore, is scaled to multiple continuous stirred tank reactors using the Residence Time Distribution (RTD) model. A graphical approach reported earlier was improved for better predictions of the observed experimental results. Batch data was obtained from a laboratory scale reactor. The scaling is used to predict the temperature rise in a continuous reactor, which happens mostly due to conversion of pyrites present in the ore. The scaling obtained with the modified model is compared with measurements of uranium conversions and reactor temperatures in a pilot scale continuous reactor having three stirred tanks in series. A good comparison is obtained for several different operating conditions of the pilot and commercial scale reactors. Hence, RTD is a good tool for scale-up and design of commercial scale leaching of uranium from lean ores.

1. Introduction

Commercial exploitation of a uranium resource is assessed on the basis of maximum extractable uranium from it by leaching. Extraction of uranium is first carried out on a laboratory scale. After identifying the optimum leaching parameters at this scale, they are scaled up to a pilot level and then to a commercial level.

One of the important steps in designing the commercial scale leaching process is finding relation between operating conditions of laboratory batch process and continuous process; the latter offers superior operational convenience and savings on account of no shutdown time for loading and unloading of materials (Levenspiel, 2001). Better product quality control is possible through automation in continuous production as there are fewer opportunities for human error. Use of continuous leaching reactor is indispensable and highly beneficial for commercial uranium mills where high throughputs (few thousand tons per day) of ores are processed.

Residence time in a batch leach reactor is same for all the particles participating in chemical reactions. However, this time is distributed in a continuous reactor due to non-ideal flow patterns that include short circuits and dead zones (Fogler, 2004). Perry (1999) discussed two ways of mitigating these detrimental effects: (1) increasing the residence time in continuous leaching to exceed that of batch leaching and (2) decreasing the possibility of early discharge, caused by short-circuiting, by increasing the number of leaching stages. This increase is achieved by having multiple identical tanks in series, maintaining the total volume of all the tanks constant. The reactor contents flow from one tank to the next in series; in each tank the contents are perfectly mixed.

There are several theoretical scale up methods for designing continuous leaching process from laboratory batch kinetic tests. Nikkhah Khosrow (1998) outlined methods for homogeneous or heterogeneous leaching systems. He assumed the reaction rate in continuous reactor to be equal to the minimum that prevails at the end of a corresponding batch leaching cycle in the case of homogeneous systems. The heterogeneous method is based on shrinking core model described by Levenspiel (2001). Henein and Biegler (1988) reported the design equations for scale up of heterogeneous systems to predict residence time in continuous leaching using a dimensionless residence time, Damkohler number. They provided fairly accurate assessment of scale-up factor required for sizing a continuous leach reactor for same feed size distribution for batch and continuous cases. Pritzker (1993) extended steady-state design equations for continuous leaching in plug-flow and continuous stirred-tank reactors taking into account of the simultaneous depletion of solid and aqueous reactants. Crundwell et al. (2013) showed the population balance approach, incorporating particle size distribution, on performance of continuous reactors with the help of leaching

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http://dx.doi.org/10.1016/j.hydromet.2014.03.014
0304-386X/

Nomenclature

M_t	Reactor discharge at time t, %
t	Time, h
N	Number of tanks in series in continuous reactor
t_{av}	Average retention time per tank, h
Y_{batch}	Percent conversion of pitchblende in batch leaching
Y_{UO2}	Percent conversion of pitchblende in continuous leaching
Y_{UO3}	Percent conversion of UO_3 in continuous leaching
Y_{FeS2}	Percent conversion of pyrite in continuous leaching
Y_{NaHCO3}	Percent decomposition of $NaHCO_3$ in continuous leaching
$\dot{M}_{solids}$	Mass flow rate of solids into/out of the continuous leach reactor (kg/h)
$\dot{M}_{liquid}$	Mass flow rate of liquid into/out of the continuous leach reactor (kg/h)
$\dot{m}_{UO_2}$	Mass flow rate of UO_2 into the continuous leach reactor (kg/h)
$\dot{m}_{UO_3}$	Mass flow rate of UO_3 in the continuous leach reactor (kg/h)
$\dot{m}_{FeS_2}$	Mass flow rate of FeS_2 into the continuous leach reactor (kg/h)
$\dot{m}_{NaHCO_3}$	Mass flow rate of $NaHCO_3$ in the continuous leach reactor (kg/h)
$C_{p,\ solids}$	Heat capacity of solids in continuous reactor (kJ/kg-°C)
$C_{p,\ liquid}$	Heat capacity of liquid in continuous reactor (kJ/kg-°C)
T_{in}	Temperature of inlet slurry to the continuous reactor (°C)
T_{out}	Temperature of outlet slurry from the continuous reactor (°C)
ΔT	Rise in temperature of slurry in the continuous reactor (°C)
ΔH_{UO2}	Heat of reaction of oxidation of pitchblende/kg of pitchblende (kJ/kg)
ΔH_{UO3}	Heat of reaction of dissolution of UO_3/kg of UO_3(kJ/kg)
ΔH_{FeS2}	Heat of reaction of oxidation of pyrite/kg of pyrite (kJ/kg)
ΔH_{NaHCO3}	Heat of reaction of decomposition of $NaHCO_3$/kg of $NaHCO_3$ (kJ/kg)
p	Pyrite content of feed solids to the continuous reactor (%)

number, defined as linear rate of shrinkage times residence time divided by mean particle size, assuming unreacted shrinking-core model. Rubisov and Papengelakis (1995) developed a mathematical model to analyze the performance of continuous hydrometallurgical reactors when process upsets occur due to the sudden changes in oxygen mass transfer, oxygen feed rate, solids feed rate, cooling water injection, etc. Sarkar (1985) developed a graphical method for scale-up without assuming a rate equation for batch leaching. The method involves dividing batch kinetic curve, obtained from experiments, into small intervals of dimensionless time and applying residence time distribution equation for N stirred tanks in series. This method is adopted in the present study, with modification, to find residence time for continuous leach reactor from batch kinetic data generated on leaching of uranium using sodium carbonate and bicarbonate solution. The ore used is from the deposit at Tummalapalle (a small town in the district of Cuddapah located in the state of Andhra Pradesh, India) where India's first ever uranium mill based on alkaline leaching is commissioned in 2012. The simulated residence times are verified with the experimental data generated on three-stage pilot scale continuous stirred tank reactor.

Heat effects play a vital role in the operation of a commercial scale continuous leach reactor. The temperature of all the exiting streams from the reactor is influenced by inlet temperature of entering streams and the heats of reactions (Rubisov and Papengelakis, 1995). The temperature of the reactor contents is essential to calculate thermal stresses for mechanical design of the reactor. Here the heats of reactions of the following reactions are considered: (1) dissolution of pitchblende, the uranium mineral, and pyrite (exothermic) and (2) decomposition of sodium bicarbonate (endothermic). The rate of generation of heat in continuous leach reactor was calculated by combining the simulated conversions of pitchblende and pyrite with their heats of reactions in carbonate medium. The sensitivity analysis of temperature of autoclave with respect to pyrite content of the ore is also included. The experimental and predicted temperatures in a three stage continuous autoclave of 850 L capacity are found to be close.

The heat effects are not crucial if the ores contain small quantities of pyrites. For ores having large amounts of pyrites, they are separated out before the uranium is extracted and hence the heat effects due to pyrites are negligible. However, for ores containing intermediate quantities of pyrites, as in this paper, the effects cannot be ignored.

The results of the present study serve as a reference for predicting the leaching efficiency as a function of time, inlet stream temperature and pyrite content of the ore. The simulation of residence time and heat absorption/evolution for continuous leaching from the laboratory data is useful in operation of the recently commissioned alkaline leaching plant at Tummalapalle in India.

2. Experimental

2.1. Materials

The Tummalapalle uranium ore was crushed using a jaw crusher followed by a roll crusher. The crushed product was ground in a laboratory ball mill to a fine powder and sieved. Each sieve fraction was separated according to specific gravity as bromoform lights, methylene iodide lights and methylene iodide heavies. Minerals in each fraction were estimated under transmitted and reflected light by microscopic grain mounting. Cellulose nitrate film auto-radiography was carried out on all fractions to estimate the distribution of radioactive phases. The complete mineralogical composition of feed ore sample is given in Table 1. The optical micrographs shown in Fig. 1 indicate that uranium occurs as pitchblende both as discrete grains and in association with pyrite, goethite, magnetite and quartz in various fractions. Since uranium ores containing >15% carbonates cannot be subjected to acid leaching (Merrit, 1970), the Tummalapalle ore (containing 83% carbonates by wt.) was treated by alkaline leaching using a mixture of sodium carbonate and sodium bicarbonate of L.R. grade (99.5% pure). Pressurized oxygen gas of commercial grade (99.6% pure) was used as an oxidant.

2.2. The chemical reactions

During alkaline leaching of uranium ores, the uranium is oxidized from the +4 to +6 oxidation state by dissolved oxygen (reaction

Table 1
Mineralogical composition of the Tummalapalle[a] uranium ore.

Mineral	% Weight
Carbonate minerals (calcite + dolomite)	83.2
Quartz + feldspars	11.3
Collophane	4.3
Pyrite	0.47
Chalcopyrite	0.05
Magnetite	0.15
Ilmenite + leucoxene	0.25
Iron hydroxides (goethite)	0.27
Pitchblende	**0.01**

[a] A small town in the district of Cuddapah located in the state of Andhra Pradesh, India.