



Application of a topochemical reaction model to predict leaching behavior of high carbonate uranium ores in alkaline solutions: An experimental case study

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ABSTRACT

A variant of a topochemical reaction model based on the shrinking core mechanism was developed for application to leaching of high carbonate uranium ores using sodium carbonate as leachant and oxygen as oxidant. Since the ground ore was not uniform in size, particle size distribution was incorporated in the model according to the well known Gates–Gaudin–Schumann equation which best represented the size distribution of the ground ore. The model was fitted to leaching data obtained from a uranium ore from a prospective deposit at Gogi in Karnataka, India. The ore comprised pitchblende and coffinite (0.2% as U_3O_8) as principal uranium minerals and a high percentage of pyrite (about 5%), calcite (62%) and quartz (13%) as main gangue minerals. Partial pressure of oxygen (0.15–3 atm), temperature (70–110 °C), concentration of leachant (0.47–0.94 g/L) and stirring speed (573–900 rpm) were tested. The activation energy for dissolution of uranium was determined to be 7 kcal/mol, which implies that leaching was controlled by chemical reaction. Extraction of uranium was found to be faster at partial pressures of oxygen below 1 atm and decreased significantly with increasing pressure due to the increased rate of dissolution of pyrite. Energy Dispersive X-ray spectroscopy of scanning electron micrographs of the ore and leach residue has also showed high extraction of uranium and low dissolution of pyrite at <1 atm partial pressure of oxygen. The dissolution rate of uranium from the ore studied was found to be much higher than that of pure UO_2 under the same leaching conditions.

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1. Introduction

The uranium ores currently being mined in India are mainly from medium and high tonnage deposits in Jaduguda, Bhatin, Narwapahar, Turamdih and Banduhurang in the state of Jharkhand for the two mills operating at Jaduguda and Turamdih (Anon, 2012). These ores contain mainly quartz and chlorite (up to 80% by wt) with U_3O_8 in the range 0.03 to 0.06% (Taraknath V.K. et al., 2009). In addition, the large tonnage uranium in Tummalapalle deposit, where the ore contains about 83% carbonates and 0.05% U_3O_8 (Rai et al., 2009; Suri et al., 2010), has led to commissioning of the third mill in Tummalapalle. Two more uranium mills are planned: one at Lambapur in Andhra Pradesh, with a medium tonnage deposit, containing 96% quartz and 0.1% U_3O_8 (Shrivastava et al., 1992) and another at Kylleng–Pyndengsohiong–Mawthabah (KPM) in Meghalaya, with a large deposit containing 90% quartz and 0.1% U_3O_8 (Padmanabhan et al., 2010a). In addition, a uranium plant is now being designed for a low tonnage, high grade (0.2% U_3O_8) deposit at Gogi, in Karnataka (Chaki et al., 2005). Extraction of uranium from

this deposit is investigated as a case study for the kinetic model developed herein.

Though leaching is a heterogeneous process, the kinetic models reported in literature on leaching of various minerals and ores are derived from the rate equations of both homogeneous and heterogeneous processes. Leaching of ulexite mineral in perchloric acid (Demirkiran and Kunkul, 2007) was found to follow the Avrami equation, a homogeneous kinetic model. Leaching models are reported for ore particles with valuable minerals in pores of the ore rocks using heterogeneous reactions (Bouffard and Dixon, 2007). Bartlett (1972) coupled the pore diffusion limited model with particle size distribution. Most leaching models are governed by shrinking core model equations assuming uniform particle size and spherical shape of the reacting solids (Santos et al., 2010; Vu et al., 2013; Wang and Maroto-Valer, 2011). However, the models with particular reference to leaching of uranium are at large based on either simple homogeneous chemical rate equations or Michaelis–Menten type of kinetic models (De Pablo, 1999; Giammar et al., 2012; Habashi and Thurston, 1967; Schortmann and DeSesa, 1958; Sharma et al., 1996). De-Xin Ding (2013) developed a model incorporating particle size distribution for heap leaching, characterized by advective flow conditions, of uranium using aqueous acid solution. Carbonate leaching of a low grade Indian uranium ore is reported to follow mixed control shrinking core model (Anand Rao et al., 2010). Most of the models reported on

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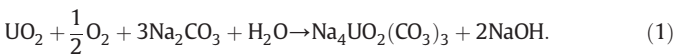
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carbonate uranium leaching are developed for either ores with low pyrite content or pure uranium minerals or for advective flow conditions.

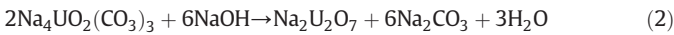
In the present study, a topochemical model for heterogeneous reactions, namely, shrinking core model incorporating particle size distribution effects is developed for leaching under agitated conditions. The model is verified with the alkaline leaching tests using Na_2CO_3 as leachant and industrial oxygen as oxidant on a carbonate uranium ore from Gogi, Karnataka, India, containing low silica and abundant pyrite (about 5%) more than the stoichiometric requirement for uranium leaching. Excess pyrite not only needlessly consumes reagents but also decreases extraction of uranium. Hence, it is usual practice to float out pyrite from ores with high pyrite content. The pyrite rich concentrate and the tails are treated separately by acid and alkaline leaching, respectively, for recovery of uranium (Anon, 1990). In contrary to the general practice, the present study unveiled possibility of single step alkaline leaching of Gogi ore for dissolving maximum uranium values while minimizing undesirable pyrite reaction. Experimental study also included ore characterization using optical microscope, observation of morphological changes due to leaching using a Scanning Electron Microscope.

2. Chemical reactions

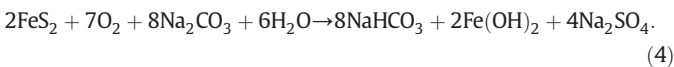
The macroscopic chemical reaction combining both oxidation of uranium present in uraninite/pitchblende and its subsequent dissolution is given by Eq. (1) (Sharma et al., 1996):



The hydroxyl ion precipitates uranium back as $\text{Na}_2\text{U}_2\text{O}_7$ according to Eq. (2), if not neutralized by sodium bicarbonate. The neutralization reaction is given by Eq. (3) (Anon, 1993):



But, sodium bicarbonate may not be required to be added if the ore contains pyrite, which generates sodium bicarbonate in situ upon reaction with sodium carbonate, as given by Eq. (4):



3. Kinetic model

Autoclave batch leaching of uranium ore particles may be represented by the conventional shrinking core model, the governing equation of which depends on one or more of the controlling mechanisms: (1) film diffusion, (2) chemical reaction at the surface and (3) ash diffusion of reactants/products (Levenspiel, 2001). In case of agitation leaching, as is the present case, the film layer on the particles is continuously replaced which leads to fast diffusion of solutes in the film. Hence film diffusion does not control the leaching reaction. Also, since all the reaction products of uranium dissolution Eqs. (1), (2) and (4) are soluble and leave no ash product, ash diffusion controlled mechanism is also ignored. The leaching rate then can be investigated with surface chemical reaction control mechanism governed by Eq. (5) (Ajemba and Onukwuli, 2012).

$$1 - (1 - \alpha)^{\frac{1}{3}} = k t \quad (5)$$

where α = conversion at time t (min), and k = overall rate constant (min^{-1}). k is a function of concentrations of reactants, temperature, stirring speed, size of particles, and many other properties of the system. Particle size distribution is incorporated into Eq. (5) using the procedure adopted by Gbor and Jia (2004). Dixon (1995) also presented the doubly

integrated micromodel for batch leaching for distribution of particle size. Suni et al. (1989) illustrated experimental evidence that leaching models without consideration of the variation of particle size are prone to erroneous conclusions. Nunez and Espiell (1985) have purported that particles possess not only a distribution of size but also shape. However, variation of particle shape is not included in the present study owing to the difficulty in obtaining the shape distribution. Hence, one of the limitations of the model presented in this paper is that particles were assumed to be spherical.

For mono-sized particles of diameter D , undergoing shrinking core dissolution, the rate constant k is given by

$$k = \frac{k_n}{D} \quad (6)$$

where k_n is independent of D . Eqs. (5) and (6) may be combined to get the conversion, $\alpha(D, t)$, as a function of diameter, D and time, t to get

$$\alpha(D, t) = 1 - \left(1 - \frac{k_n t}{D}\right)^3. \quad (7)$$

For a group of particles of various sizes divided into discrete size ranges,

$$\text{Fraction reacted} = \sum_D (\alpha(D, t)) (\text{Mass fraction of size } D). \quad (8)$$

This equation transforms as following for a continuous distribution of particles,

$$\text{Fraction reacted} = \int_0^{D_{max}} (\alpha(D, t)) (p(D)) dD \quad (9)$$

where $p(D)$ is the particle size density function based on mass of particles. D_{max} is the size of largest particle in the system. As there could be always few particles of less than certain size that react fully in a given time, an intermediate diameter, D_t is introduced such that

$$\alpha(D, t) = 1, 0 < D < D_t. \quad (10)$$

Eq. (10) implies that at any time (t), all particles with sizes (D) less than D_t are fully reacted and thus have a conversion value of 1, which when substituted in Eq. (7) yields $D_t = k_n t$. But all the particles greater than D_t are partially reacted with a conversion given by Eq. (7) with the limits of particle size, D , as given in Eq. (11).

$$\alpha(D, t) = 1 - \left(1 - \frac{k_n t}{D}\right)^3, D_t < D < D_{max} \quad (11)$$

Popular Gates Gaudin Schumann (GGs) size distribution function is chosen for $p(D)$ derived from $f(D)$ given in Eq. (12).

$$f(D) = \left(\frac{D}{D_{max}}\right)^m. \quad (12)$$

Differentiation of Eq. (12) yields $p(D)$:

$$p(D) = m \frac{D^{m-1}}{D_{max}^m}. \quad (13)$$

Substituting Eqs. (10), (11) and (13) into Eq. (9) gives

$$\begin{aligned} \text{Fraction reacted} = \alpha &= \int_0^{D_t} (1) \left(m \frac{D^{m-1}}{D_{max}^m}\right) dD \\ &+ \int_{D_t}^{D_{max}} \left(1 - \left(1 - \frac{k_n t}{D}\right)^3\right) \left(m \frac{D^{m-1}}{D_{max}^m}\right) dD \end{aligned} \quad (14)$$

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