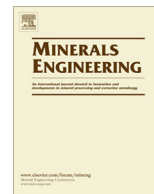


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Gangue–reagent interactions during acid leaching of uranium

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ABSTRACT

During the acid leaching of uranium, gangue–reagent interactions have both negative and positive consequences. Gangue dissolution increases reagent costs, and in some cases can prevent the economic acid leaching of an ore, but can also increase uranium mineral exposure and improve recoveries. Due to rapid dissolution kinetics, the acid consumption characteristics of the various carbonate species are readily predicted, however the same is not true of silicate gangue. Due to factors including slower leach rates, incongruent dissolution, parabolic kinetics, and surface area, pH and temperature dependence, the gangue acid consumption characteristics of silicate minerals are significantly more complex. A detailed mineralogical investigation and acid leach tests were conducted on sandstone-hosted uranium ore samples. The dissolution characteristics of the more common gangue phases were determined. The study demonstrated that gangue–reagent interactions can be predicted from mineralogical data, thus reducing technical risk during processing.

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1. Introduction

Uranium ores are processed by either an acid or an alkali leach (Abhilash et al., 2009). Acid leaches are typically preferred to alkali methods, as they frequently offer more rapid dissolution kinetics and are able to leach more efficiently at coarser grinds (Lunt et al., 2007). Alkali leaches are used where the reactivity of the gangue prevents acid leaching (Merritt, 1971; Lunt et al., 2007). However, under certain conditions it may not be possible to process an ore by alkali leach, for example if gypsum is present (IAEA, 1980). Therefore the nature of the gangue mineralogy can prevent the economic leaching of U. It would therefore be desirable to better understand the factors that influence gangue acid consumption and develop a method of quantitatively predicting gangue acid consumption from mineralogical data.

The major advantage of being able to model gangue acid consumptions from mineralogy (rather than simply performing a leach test), is that a model can be used to generate data for a range of possible conditions, while separate leach tests would be necessary for each condition of interest. The availability of mineralogical data also makes it possible to identify other potential mineralogical problems, such as clays.

In addition to contributing to acid consumptions, the dissolution of gangue minerals can aid in the recovery of valuable metals

(Lottering et al., 2008). As the gangue dissolves, so the proportion of the mineral of interest that is exposed to the leach liquor increases, resulting in improved recoveries.

The gangue–reagent interactions that occur during the acid leaching of U, were investigated in order to understand their effect on reagent consumption and U dissolution. Samples were obtained from two Karoo-aged U occurrences. One sample was obtained from a U prospect within the main Karoo basin (sample MKB1) and three others (samples RAO, OAO and MSO) were obtained from the Kayelekera mine in Malawi.

1.1. Gangue mineral dissolution

Due to the rapid dissolution of carbonates, these minerals are typically considered to consume their stoichiometrically equivalent amount of acid during U leaching (IAEA, 1980). The dissolution of silicate minerals is more complex than that of carbonates. Due to the complex nature of silicate dissolution, it has been suggested that the dissolution of these phases cannot be predicted (IAEA, 1980). However, large amounts of research have been performed on the acid dissolution characteristics of silicate minerals since 1980. Much of this research (for example Lawson et al., 2005) was aimed at understanding the behaviour of gangue minerals during acid mine drainage, however due to similarities in pH and acid type, the information is readily transferred to U leaching.

Several factors influence the dissolution characteristics of minerals. The following factors influence the rate of mineral dissolution: mineral composition, time, temperature, pH, surface area

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and incongruent dissolution (Ross, 1969; Brantley and Conrad, 2008; Lüttge and Arvidson, 2008; Brantley, 2008).

Since minerals dissolve at a finite rate, the extent of dissolution is a function of time. However, the rate of dissolution can also vary with time. Generally two stages are seen during dissolution, (1) an initial more rapid rate which gradually slows to (2) steady-state dissolution (Brandt et al., 2003 and Brantley, 2008). The U leach tests typically include the initial phase of rapid dissolution but, (depending on the constituent minerals and conditions of the leach, may not necessarily reach steady-state dissolution. This decay from an initially rapid rate to steady state conditions is typically referred to as parabolic kinetics (Brantley, 2008). However, in practice, the change in rate as a function of time is more readily modelled by an exponential decay function, than a parabola.

The rate of mineral dissolution is considered to vary with temperature according to the Arrhenius equation (Brantley and Conrad, 2008).

$$k = A \exp(-E_a/RT) \quad (1)$$

where k is the rate constant at a given temperature, A is the pre-exponential factor, E_a is the activation energy, R is the ideal gas constant and T is the absolute temperature (Brantley and Conrad, 2008). Using the Arrhenius equation it is possible to adapt kinetic data collected at one temperature to a more desirable temperature (Brandt et al., 2003).

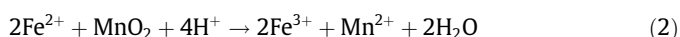
Dissolution rates are typically normalised to the surface area of the mineral (Lüttge and Arvidson, 2008). Many workers (including Lawson et al. (2005); Brandt et al. (2003); Knauss and Wolery (1989)) favour the BET (Brunauer et al., 1938) method of surface area determination. This method allows for surface area determination by measuring the adsorption of an inert gas onto the surface of the sample. However, this is not as readily adapted to complex samples consisting of several minerals in various concentrations, such as is typically the case with U ores.

In addition to temperature and mineral surface area dependence, the release rate of an element from a dissolving mineral is influenced by incongruent dissolution. Incongruent dissolution describes the case where elemental release rates are not proportional to the stoichiometry of the mineral (Brantley, 2008).

1.2. Reagents used in uranium leaching

Sulphuric acid is typically used to achieve the low pH required for acid U leaching (Ho and Quan, 2007). This acid has two acidic protons. The dissociation of the first proton has a sufficiently high equilibrium constant that it can be considered to proceed to completion, while the second dissociation has an equilibrium constant of 1.2×10^{-2} (Brown et al., 2003).

During acid leaching hexavalent U minerals tend to dissolve readily, while tetravalent U minerals require oxidation to the hexavalent state prior to dissolution (Lunt et al., 2007). In the processing of the Witwatersrand ores of South Africa, MnO_2 in the form of pyrolusite, is the preferred oxidant (Lottering et al., 2008). At Olympic Dam sodium chlorate is used (Miki and Nicol, 2009). Other oxidants include SO_2/O_2 (Ho and Quan, 2007), hydrogen peroxide and oxygen (Venter and Boylett, 2009). In this study only MnO_2 was considered. It has been found that these oxidising agents do not act on the U minerals directly, but instead serve to oxidise iron to the Fe^{3+} state, which in turn oxidises the U according to Eq. (2) (Lottering et al., 2008). This equation shows that the dissolution of MnO_2 consumes two moles of sulphuric acid for each mole of MnO_2 dissolved.



2. Methodology

In order to understand the response of the gangue minerals during leaching, the head samples underwent detailed mineralogical characterisation prior to acid leaching.

2.1. Gangue mineral characterisation

The samples were milled to the desired grind using a laboratory scale rod mill. All of the samples were milled to a P_{80} of 212 μm . Due to the large amount of MKB1 material, a second aliquot of this sample was milled to $-75 \mu\text{m}$ and treated as a separate sample for the textural analyses and leach tests.

Both the head and leach residue samples were analysed by XRD and QEMScan bulk mineralogical analysis (BMA), to identify and quantify the gangue minerals present, and determine the mean grain size and degree of exposure of each gangue phase.

The XRD analyses were performed using the guidelines of McCusker et al. (1999). The powder samples were micronised and then analysed using a Panalytical X'pert Pro diffractometer and $\text{Co K}\alpha$ radiation. The resulting data was processed using HighScore Plus software and the PanICSD database. Rietveld refinement was used to determine the quantities of each gangue phase.

QEMScan BMA analyses, conducted according to the methods described by Gottlieb et al. (2000) and Coetzee et al. (2011), were also used in the gangue mineral quantification. The BMA analyses were performed on transverse cut polished sections, using a QEMScan based on a Carl Zeiss Evo 50 scanning electron microscope with four SiLi EDX detectors. The BMA and XRD data were validated against each other and against major element XRF data. In addition to XRF analyses, the samples were also analysed by titration to determine their Fe^{2+} content.

The BMA data was also used to determine the textural characteristics of the gangue. BMA analyses are more rapid and cost effective than other QEMScan techniques and therefore more readily applied to variability studies. For this reason the BMA was chosen over particle map analysis (PMA). However, BMA analyses are more prone to stereological bias than two dimensional scans, such as PMA (Sutherland et al., 1988), and it was necessary to stereologically correct the grain size data.

2.2. Uranium mineral characterisation

The U mineral characterisation was performed by QEMScan trace mineral search (TMS) using the same instruments used for the BMA analyses. The mineral identification was validated by manual scanning electron microscopy with associated energy dispersive X-ray analysis.

2.3. Leach tests

The batch acid leach tests were conducted using the guidelines given by the IAEA (1980 and 1990). Approximately 2 kg split aliquots of each sample were used. Due to the large amount of MKB1 material, it was possible to leach this sample at a number of different pH and Eh conditions and at two different grinds (P_{80} 212 and 75 μm). The limited amount of material in the Kayelekera samples limited these leach tests to one test per sample (P_{80} 212 μm).

The leaches were performed at a solid to liquid ratio of 1:1, in 5 L plastic beakers. Agitation was achieved by overhead stirrers. Temperature was controlled to between 35 and 40 °C. The pH was controlled using concentrated sulphuric acid and the redox potential was controlled using MnO_2 , both Associated Chemical Enterprises, Platinum Line, analytical reagent grade.

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