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An integrated multiscale approach to heap leaching of uranium-ore agglomerates

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ABSTRACT

In heap leaching, agglomeration of clay rich ores is commonly used and may prevent plugging phenomena within heaps, but is not always successful. In order to better understand the heap leaching process of uranium ore agglomerates, a multi-scale approach, based on 10-cm and meter-scale columns, was used to achieve a Representative Elementary Volume (REV) of packed agglomerates. Flow rate and agglomerate size distribution were found to have no impact on the ore leaching kinetics. An increase of the sulfuric acid content of the leaching solution caused a slight increase of uranium extraction. Meter-scale tests indicated that scaling up had no significant influence on extraction kinetics. Some tests have also shown that column porosity and permeability decreased during irrigation and with the heap depth. This led to the occurrence of plugging. X-ray tomography analyses on clogged sections of the column revealed that this phenomenon arises from agglomerate mechanical degradation. In addition, an integrative numerical model of the leaching of the meter scale column of agglomerates was built, combining flow and reactive transport equations where the effective reaction term was directly inferred from the REV experiments. An excellent agreement was observed with the column tests.

1. Introduction

Heap leaching is an industrial method to process low grade ores, consisting of percolating an acid or alkaline leaching solution through a 4 to 10 meter high heap of crushed ore (Ghorbani et al., 2016). This technique is used to process several ores such as uranium, copper or nickel laterites and is based on fluid-rocks interactions at the heap scale and on the percolation of the leaching solution through ore porosity to dissolve minerals of interest at the micro-scale (Bartlett, 1997). In this way, the accessibility of these minerals by the leaching solution is a limiting factor (Ghorbani et al., 2011).

Due to the significant size of heaps and the long leaching time (about 1 year for a 6 to 9 m high heap), heap leaching processes may be difficult to investigate. Smaller size experimental models and numerical models are used instead. Experimentally, columns from 0.5 to 6 m high are used to reproduce leaching conditions (Bennett et al., 2012; Robertson, 2017). Such tests allow analysing the influence of the different leaching parameters. However, they may contain a bias: Bouffard and West-Sells (2009) noted that the leached ore density decreased with the decrease of column diameter. This scale reduction decreases the ore packing density, due to wall effects, which subtly alters mechanical behaviour in the case of column leaching. In addition, heterogeneities present in the heap, such as a wide particle size distribution or the presence of inert materials, may cause an overestimation of the leaching rate measured in columns compared to that measured in situ. Numerical models (Dixon and Hendrix, 1993; Mellado et al., 2009) using parameters fitted to experimental data were also developed, in order to study the impact of the different leaching parameters (Ghorbani et al., 2016) from very simplified analytical models to more complex mechanistic approaches. They classically describe fluid flow and transport of leached solution through the heap and reaction kinetics with the minerals of interest (Dixon, 2003; Dixon and Petersen, 2003). Depending on the operating conditions (evolution of heap packing or of the heat within the heap), additional couplings may be required (Dixon, 2000; Valencia et al., 2008). Note that several models were proposed in the literature but few are used in industrial applications (Ghorbani et al., 2016; Robertson, 2017). The main difference between these models lies in the governing assumptions and the related mass balance equations used to represent dissolved species transport, leaching solution flow and the reactional model. The Richards equation

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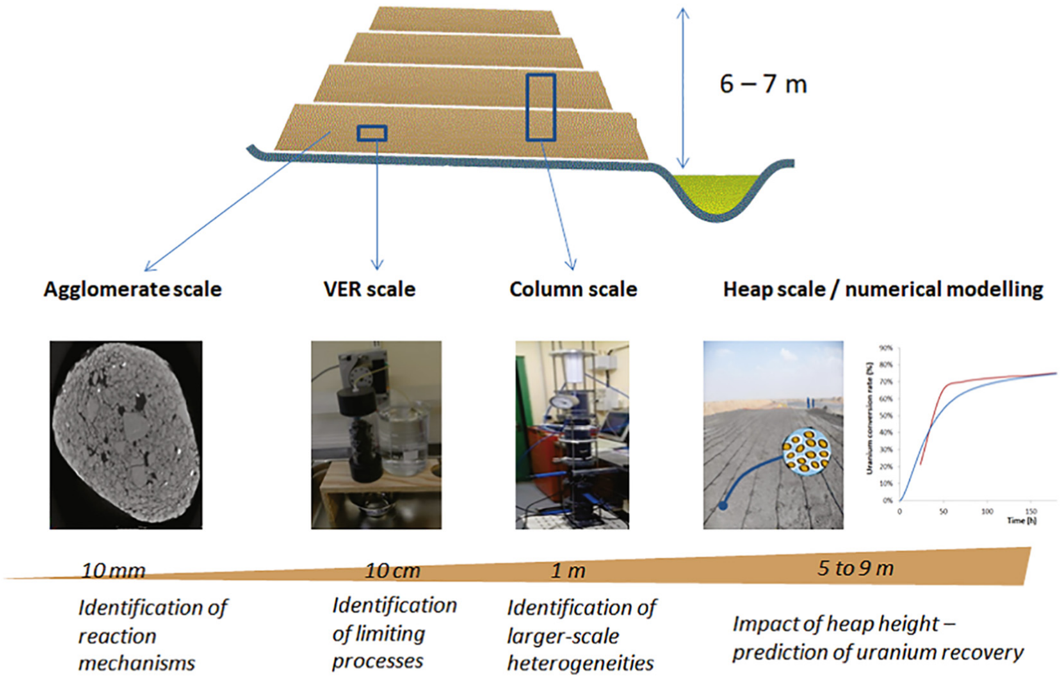

Fig. 1. Multi-scale analysis of uranium ore agglomerates leaching.

in combination with the traditional Mualem-Van Genuchten model is usually chosen to model solution flow through the heap (Robertson, 2017) as leaching are often occurs under partially saturated conditions. However a generalised Darcy model may also be used when the gas phase is considered (Bennett et al., 2012). Moreover, two types of leaching transport models may be distinguished depending on the level of column heterogeneity (Golfier et al., 2007): (i) models considering the heap as a homogeneous medium controlled by diffusion or advection (Bennett et al., 2012) and (ii) dual porosity models, considering the heap as two distinct regions: a stagnant region and a mobile one (Bouffard and Dixon, 2001; Dixon and Petersen, 2003; Robertson, 2017). Such models can closely emulate heap behaviour in case of channelling (Ogbonna et al., 2006). At the mineral particle scale, the easiest approach is to consider a uniform reaction model ("uniform- or homogeneous-reaction model"), meaning a uniform penetration of leaching solution within particles, independently of their size. However, when diffusion is limiting (Sheikhzadeh and Mehrabian, 2007), most models use the shrinking core model to describe reactions between minerals and leaching solution (Bennett et al., 2012; Robertson, 2017). Although the shrinking core model is easy to apply, it supposes that the particles to be leached are spherical and that minerals distribution is homogeneous, which is not the case in experimental tests (Bennett et al., 2012). All of these models led to identify and assess parameters that influence heap leaching efficiency such as heap temperature or leaching reagent concentration which both impact the dissolution kinetics (Bartlett, 1997; Petersen, 2016). Ghorbani et al. (2011) also point out heap permeability as a key factor of heap leaching. If percolation of leaching solution is too slow, chemical reactions between reagent and minerals cannot occur and on the other hand, if this characteristic time is too long, it might result in the flooding of the ore (Ghorbani et al., 2016). In addition, a low porosity may enhance plugging, causing a decrease of leaching efficiency and heap collapsing. An uneven permeability is also reported to induce channelling and to prevent irrigation of some parts of the heap. Such permeability difficulties are mostly caused by the nature of the ore and especially high clay and fine particles content.

To solve this problem, the nickel, copper and uranium industries use agglomeration, consisting of increasing ore particle size distribution by binding fines and clays together into larger particles. This improves heap permeability and stability and also prevents fines migration during leaching (Bartlett, 1997; Bouffard, 2005; Yijun et al., 2004). Note that most studies of agglomerate heap leaching focus on copper or nickel laterite ore (Dhawan et al., 2013; Kodali et al., 2011; Nosrati et al., 2012a,b) but up to now, the behaviour of uranium-ore agglomerates remains poorly understood. The only exception is the recent study of Hoummady et al. (2017) who have identified changes in the structure of uranium-ore agglomerates during leaching.

Here, we focus on leaching effects at the agglomerate scale and at the Representative Elementary Volume (REV) scale (by the use of small column leaching tests). The objective of the current work is to upscale these results to the heap scale, by using both REV leaching experiments, meter scale columns leaching tests and numerical modelling. An integrative multiscale approach, illustrated in Fig. 1, is conducted for this purpose by (i) identifying the limiting processes at the smaller scale, (ii) stating the governing assumptions at the heap scale in terms of these latter processes and (iii) predicting the uranium extraction. In addition, this study also intends to characterize and understand plugging.

2. Materials and methods

2.1. Ore, agglomeration and leaching processes

The agglomerates used in this study were produced from clay-rich sandstone containing about 900 ppm of uranium and > 10 wt% of clay minerals (mainly kaolinites, illites, mixed layered illite-smectite phases and chlorites) provided by AREVA and originating from Somaïr, Niger. Most of the uranium in this ore is hosted by clay minerals and especially by chlorites. In addition, this ore contains a low amount of carbonate (0.4 wt%) and calcium (0.29 wt%), compared to the aluminium content (8.32 wt%). Due to this feature and to the high clay content, this ore is ideally suited for a study of the agglomeration process.

Crushed ore was agglomerated with water and sulfuric acid, at a ratio of 25 kg of acid per 1000 kg of ore for a liquid/solid ratio of 0.08 kg/kg, within a cement mixer at a speed of 32 rpm. Half of the water was first mixed with the dry ore. Sulfuric acid mixed with the rest of the water was then added to the ore and the agglomeration carried on for 3 min. Then, agglomerates were stored for curing during at least 24 h to harden the bonds between particles (Pietsch, 2002).