



A study of uranium-ore agglomeration parameters and their implications during heap leaching



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ABSTRACT

Agglomeration is commonly used for processing clay-rich ores in order to prevent undesirable effects, especially the risks of clogging or preferential channelling within the heap leaching piles. Several parameters such as agglomeration time, agglomerates water content or agglomerates binder content were identified as having a strong influence on the agglomeration process. In the present study, the impact of water content, sulfuric acid and addition of a polyacrylamide binder on the porous structure and mechanical strength of uranium-ore agglomerates were investigated, before as well as after 10 days of leaching. A multi-analytical approach, combining SEM, MIP, ICP-OES, X-ray tomography analyses and oedometer tests was used for this purpose. Increasing sulfuric acid concentration at agglomeration was found to enhance uranium extraction extent but reducing agglomerate strength during leaching. In addition, the increase of L/S ratio caused a decrease of agglomerate porosity due to the formation of higher amount of primary aluminous silicate matrix. Finally, the use of a polyacrylamide binder improved leaching resistance of agglomerates, allowing an increase of both agglomerate porosity and uranium extraction rates between 24 and 240 h of leaching while keeping a better compressive strength.

1. Introduction

The agglomeration process represents a way used by nickel, copper and uranium industries to enhance acid heap leaching of low grade ores containing large amount of fines and clays (Dhawan et al., 2013). This industrial technology consists in increasing ore particle size distribution by gathering fines to improve the heap permeability and stability and also prevent fine particle migration during leaching (Bartlett, 1997; Dhawan et al., 2013). In some cases, a binder is added to strengthen the agglomerates.

Agglomerate quality is often related to the uniformity of particle size distribution, water content prior and during leaching, by their porosity and their strength (Bouffard, 2005). These two latter properties are critical for heap leaching success, but not easy to combine since a suitable agglomerate has to be both porous enough to be fully leached and strong enough to support the weight of heap above (Liu et al., 2012). Different types of quality tests have been proposed to assess these properties such as visual tests (e.g., glove test or agglomerate size distribution analysis) (Velarde, 2005; Dhawan et al., 2013), or batch

measurements (such as compaction, electric conductivity, permeability or soak measurement) applied to a pack of agglomerates (McClelland, 1988; Velarde, 2005; Lewandowski and Kawatra, 2009; McFarlane et al., 2011; Dhawan et al., 2013; Xu et al., 2013). However, they give only indicative and qualitative properties which mostly cannot be related to agglomerate strength, failing to give a relation between agglomerate quality and process methodology. Several operating parameters such as: ore properties (as mineralogy, particle size distribution, etc...) (Quaicoe et al., 2013), the residence time of the agglomerates within the drum and its speed (Bouffard, 2008; Nosrati et al., 2012), or the water content during agglomeration (i.e. the Liquid/Solid (L/S) ratio) were identified to have an impact on agglomeration (Velarde, 2005; Nosrati et al., 2012; Dhawan et al., 2013). High L/S ratio for instance causes an increase of agglomerate size and a decrease of agglomerate strength, suggesting the existence of an optimal moisture content for each kind of agglomerate (Velarde, 2005; McFarlane et al., 2011; Vethosodsakda et al., 2013). Finally, the presence and nature of the binder can improve the adhesion between fine and coarse particles and reduce fines migration during leaching. A “good” binder is defined

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as an agent allowing the formation of strong bonds between particles without hindering the leaching of the element of interest and being chemically compatible with the lixiviant (Kodali, 2010; Qiu et al., 2003). The choice of the binder depends mainly of the ore mineralogy and the chemistry of the leaching solution. In the case of sulfuric acid leaching, some sulfuric acid is also usually added during agglomeration as a binder for uranium, copper and nickel heap leaching (Bouffard, 2005). Sulfuric acid is interesting as it functions both as a binder and the lixiviant agent. This results in a pre-dissolution of minerals of interest during agglomeration and maturation, before the beginning of leaching (Nosrati et al., 2013; Hoummady et al., 2017). This binder also produces solid bonds between particles during the maturation time. Other inorganic binders such as gypsum or stucco have been tested (Kodali et al., 2011). Analyses concluded that such binders allowed immobilisation of fines. Polymeric binders, such as polyacrylamides have also been investigated (Lewandowski and Kawatra, 2009). These act as flocculating agents causing fine particles to stick together. Polyacrylamide-bound agglomerates tend also to have less dense packing than acid-bound agglomerates during leaching column tests. However, industrial use of these binders remains low, due to their cost and no use for uranium-ores has been reported (Dhawan et al., 2013; Kodali et al., 2011; Lewandowski and Kawatra, 2009; Qiu et al., 2003).

Agglomeration of nickel laterite and copper ores has been intensively investigated, but the only published study on uranium-ore agglomerates is our previous work (Hoummady et al., 2017). That reported that uranium ore agglomerates were composed of coalesced and layered micro agglomerates, themselves composed by the layering of phyllosilicates and a primary aluminous silicate matrix around a nucleus. This aluminous silicate matrix was constituted by new-formed phases coming from phyllosilicates as chlorites, illites-smectites and dissolved uranium mineral products during agglomeration and maturation. During leaching, two stages were observed: (i) during the first 48 h, the primary matrix was leached accompanied by high uranium recovery rates and an increase of agglomerate porosity. Then, (ii) from 48 h to the end of leaching, a secondary aluminous silicate matrix phase was formed, subsequently to the dissolution of residual illites. This caused a slight decrease of agglomerate effective porosity.

Most agglomerate quality tests are qualitative and data obtained reflect only some aspect of macroscopic averaged properties of a batch of agglomerates, while the microstructure which plays a crucial role on agglomerate quality and behaviour during leaching remains poorly understood (Hoummady et al., 2017). The current work presents further results, utilising the same comprehensive multi-analytical approach to assess these agglomerate properties, namely porosity, microstructure and mechanical strength, as in our earlier work (Hoummady et al., 2017), to identify preferential agglomeration conditions. A combination of imaging methods (X-ray Computed Tomography (CT) and SEM), petrophysical (mercury intrusion porosimetry (MIP)) and chemical (ICP OES) analyses was carried out while the mechanical properties are assessed from compressive strength tests. Results are used to characterise the differences induced by a change in agglomeration parameters (influence of sulfuric acid concentration, of the addition of a polyacrylamide binder and of the Liquid/Solid (L/S) ratio), and ultimately to improve agglomeration process.

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 more than 10 wt% of clay minerals (mainly kaolinites, illites, mixed layered illite-smectite phases and chlorites) provided by AREVA and originating from Somair, Niger. A particle size analysis of the ore was conducted with particles ranging from less than 80 μm to more than 10 mm and a P_{80} of about 10 mm. Mineral analyses indicated that uranium, as U oxides, is mostly

Table 1

Agglomeration parameters used in the experimental tests.

Batch	Sulfuric acid content (kg/t of ore)	Nalco binder content (g/t of ore)	Liquid / Solid (L/S) ratio (kg/kg)	Agglomeration time
#1 (RC)	25 kg/t	0 g/t	0.08	3 min
#2	0 kg/t	0 g/t	0.08	3 min
#3	40 kg/t	0 g/t	0.08	3 min
#4	25 kg/t	500 g/t at 1%	0.08	3 min
#5	25 kg/t	0 g/t	0.11	3 min

hosted by clay minerals and especially by chlorites. Due to this feature and to the high clay content, this ore is a good raw material for agglomeration tests.

Different agglomeration conditions have been investigated as detailed in Table 1. Crushed ore has been agglomerated in a cement mixer at 32 rpm with water, sulfuric acid and in some cases with a polyacrylamide binder (Binder Nalco 71760) provided by the Nalco Chemical Company. In the latter case, half of the water was first mixed with the Nalco binder and dry ore and then sulfuric acid mixed with the rest of the water was added and the ore was agglomerated for 3 min. Then, agglomerates were stored for maturation for at least 24 h to harden the bonds between the particles of the agglomerate (Pietsch, 2002). The first set of agglomeration conditions is referred to as reference agglomeration conditions (or RC) and corresponds to the agglomerates studied in Hoummady et al. (2017). Note that these conditions are similar to the conditions used by AREVA during agglomeration and leaching tests. Only agglomeration time was adjusted from 1.5 to 3 min to obtain similar agglomerates size distribution, due to the difference of the agglomeration drum used. Comparison with the other batches was conducted to evaluate the effects of agglomeration parameters such as the sulfuric acid concentration, the L/S ratio and the addition of the Nalco binder on the structure of agglomerates and the resulting heap leaching efficiency.

For each batch, about 120–150 g of 10-mm-diameter agglomerates were leached in a 10 cm high column over 10 days with sulfuric acid at a concentration of 10 g/L and a flowrate of 7.2 mL/h (i.e. about 5.7 L/m²/h). Leaching parameters are taken from the standard protocol designed by AREVA and values are representative of actual process conditions for acid heap leaching (Petersen, 2016). Due to the scale effect between the columns and a 6 m high heap, 10 days of leaching under laboratory conditions correspond to a leaching liquid/solid ratio of 11 equates to, about 22 months of uranium production at the heap scale, which represent 7–8 time the usual leaching time. This helps to perform long-term analysis of uranium recovery and agglomerate behaviour for industrial applications. The leached solution was sampled daily and analysed by ICP-OES (results are given with a measurement uncertainty of 10%) and the extent of uranium extraction has been calculated as moles of uranium present in the leached solution to the total moles present within the feed. At the end of the leaching process, agglomerates were dried in the open air for 1 h for drainage before post-mortem analyses.

2.2. Agglomerate analyses

A similar multi-analysis approach to the one used in Hoummady et al. (2017) was chosen to study agglomerate structure and porosity during leaching.

After drying in open air, some agglomerates were freeze-dried and analysed by scanning electron microscope (Hitachi S-4800) (SEM) and micro energy-dispersive X-ray spectroscopy (EDS), in order to determine agglomerate structure. Mercury intrusion porosimetry (MIP) measurements on freeze-dried agglomerates were performed with a Micromeritics Autopore IV (60,000 psi) porosimeter. Two mercury injections were done: the first one was used to determine the total

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