



Review

A review of acid leaching of uraninite



Suresh K. Bhargava^{a,*}, Rahul Ram^a, Mark Pownceby^b, Stephen Grocott^c, Bob Ring^d,
James Tardio^a, Lathe Jones^a

^a Centre for Advanced Materials and Industrial Chemistry (CAMIC), School of Applied Sciences, RMIT University, GPO Box 2476, Melbourne, VIC 3001, Australia

^b CSIRO Process Science and Engineering, Bayview Avenue, Clayton, Victoria 3168, Australia

^c Rio Tinto Technology and Innovation, Research Avenue, Bundoora, VIC 3083, Australia

^d ANSTO Minerals, Lucas Heights, Sydney, NSW 2032, Australia

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ABSTRACT

Uraninite is mined/processed more than any other uranium mineral for the production of uranium based compounds that are subsequently used to produce nuclear fuel. This review article provides a concise account of the available literature on one of the major processes involved in processing uraninite bearing ores, acid leaching. Improvements in the processes used to leach uraninite are required in order to ensure efficiency in the processing of lower grade uraninite bearing ores with minimal environmental impacts. This in turn requires improvements in our understanding of uraninite leaching. The main topics covered in this review include: uraninite structure, composition and low temperature geochemistry; the chemistry of uraninite leaching; key factors that influence uraninite leaching; and leach process technologies. The research that has been reviewed clearly establishes the influence of parameters such as temperature, acid concentration and particle size. The influence of other parameters however, such as solution Fe^{3+} to Fe^{2+} ratio (solution E_h), total Fe concentration, foreign ions present in the leach slurry and uraninite composition is yet to be established. Based on the literature available on the aforementioned factors the chemistry/processes involved in uraninite leaching are quite complex and require significant further studies. From the literature reviewed it is clear that variations in mineral chemistry in individual ore types across multiple deposits also make it essential that before any extraction process is considered, detailed ore characterisation studies of pre- and post-leach residues are of vital importance in order to fully understand the interrelationship between chemistry, mineralogy (ore and gangue), mineral liberation and potential leaching behaviour of uranium.

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Contents

1.	Introduction	11
2.	Uraninite: occurrence, structure, composition and low temperature geochemistry	11
2.1.	Occurrence (genesis)	11
2.2.	Structure and composition	11
2.3.	Uraninite low temperature geochemistry	13
3.	Acid sulfate leaching of uraninite	13
3.1.	Uraninite redox chemistry	14
3.1.1.	Effect of E_h	14
3.1.2.	Effect of Fe^{3+} , Fe^{2+} , Fe^{3+}/Fe^{2+} and Fe_{TOT}	15
3.2.	Leaching of natural uraninite and synthetic uraninite	17
3.2.1.	Effect of foreign ions released from gangue	17
3.2.2.	Effect of substitutional impurities	18
3.2.3.	Effect of acid concentration	18
3.2.4.	Effect of temperature	18
3.2.5.	Effect of grain size, liberation, metamictisation and pulp density	19
3.2.6.	Effect of other uranium bearing minerals	19

* Corresponding author. Tel.: +61 3 9925 3365.

E-mail address: suresh.bhargava@rmit.edu.au (S.K. Bhargava).

4.	Alternative acid sulfate leach processes	19
4.1.	Pressure leaching	20
4.2.	Microbial leaching	20
4.3.	Heap leaching	20
4.4.	In-situ leaching	21
5.	Conclusions	22
	Acknowledgements	22
	References	22

1. Introduction

Uranium is widely distributed in the Earth's crust with estimates of its abundance varying between 2 and 4 ppm (Clark et al., 2006; Fleischer, 1955). Uranium in the crust is concentrated in over 200 mineral species that contain uranium as a necessary structural constituent in either the reduced U^{4+} or oxidised U^{6+} valence state. The main economic uranium minerals are the tetravalent species uraninite, pitchblende, coffinite and brannerite and to a lesser extent the more oxidised hexavalent species carnotite (Table 1). There are also approximately another sixty mineral species that contain minor amounts of uranium, or that contain uranium as an impurity (Burns and Finch, 1999; Finch and Murakami, 1999; Frondel et al., 1967; Krivovichev et al., 2006; Plant et al., 1999).

The most important uranium mineral is the simple oxide species uraninite (nominally UO_2), found in many deposits throughout the world (Fronde, 1958; Frondel et al., 1967; Laxen and Atmore, 1957; Macnaughton et al., 1999, 2000). Uraninite accounts for a significant proportion of the world's uranium supply (Burns et al., 1997; Clark et al., 2006; Morss et al., 2011; Peehs et al., 2007) and processing of uraninite has been the mainstay of uranium production as a result of its relative ease of extraction compared to other, more refractory uranium minerals (Merritt, 1971).

The focus of this article is to provide a concise review of available literature on the acid leaching of uraninite. The review focuses on the key parameters of the leach process under conditions typically used in commercial leaching operations. Other acid-based leach process technologies such as heap leaching, in-situ leaching and pressure leaching are only briefly considered. The chemistry behind acid leaching of uraninite applies in all these technologies and only specific factors that could potentially impact leaching are discussed.

Table 1

Economically important uranium minerals.
Adapted after Pownceby and Johnson (2013).

Mineral type	Mineral name	Formula	Typical %U
<i>Tetravalent uranium minerals (U^{4+})</i>			
Oxides	Uraninite	$(U^{4+}_x U^{6+}_{1-x})O_{2+x}$ [ideally UO_2]	46–88
	Pitchblende	UO_2 amorphous	86–88
REE ± Ti ± Fe-oxides	Brannerite	$(U,Ca,Y,Ce,La)(Ti,Fe)_2O_6$	26–44
	Betafite	$(Ca,U)_2(Ti,Nb,Ta)_2O_6(OH)$	15–24
	Davidite	$(La,Ce)(Y,U,Fe)(Ti,Fe)_{20}(O,OH)_{38}$	1–6
	Orthobrannerite	$U_2Ti_4O_{12}(OH)_2$	~53
Silicates	Coffinite	$U(SiO_4)_{1-x}(OH)_{4x}$	40–60
	Uranothorite	$(U,Th)SiO_4$	<10
<i>Hexavalent uranium minerals (U^{6+})</i>			
Phosphates	Autunite	$Ca(UO_2)_2(PO_4)_2 \cdot 10-12H_2O$	48–50
	Meta-autunite	$KCa(H_3O)_3(UO_2)_7(PO_4)_4O_4 \cdot 6-8H_2O$	~57
	Torbernite	$Cu(UO_2)_2(PO_4)_2 \cdot 8-12H_2O$	47
Vanadates	Carnotite	$K_2(UO_2)_2(VO_4)_2 \cdot 1-3H_2O$	53–55
	Tyuyamunite	$Ca(UO_2)_2(VO_4)_2 \cdot 5-8H_2O$	~52
Silicates	Boltwoodite	$HK(UO_2)_2SiO_4 \cdot 1.5H_2O$	~55
	Sklodowskite	$(H_3O)_2 Mg(UO_2)_2(SiO_4)_2 \cdot 4(H_2O)$	49–54
	Uranophane	$Ca(UO_2)_2(SiO_3)(OH)_2 \cdot 5H_2O$	~41

2. Uraninite: occurrence, structure, composition and low temperature geochemistry

2.1. Occurrence (genesis)

Uranium resources are typically classified into fifteen main categories of uranium ore deposit types. Arranged, according to their approximate worldwide economic significance, they are as follows: (1) unconformity-related deposits; (2) sandstone deposits; (3) hematite breccia complex deposits; (4) quartz-pebble conglomerate deposits; (5) vein deposits; (6) intrusive deposits; (7) collapse breccia pipe deposits; (8) volcanic deposits; (9) metasomatite deposits; (10) surficial deposits; (11) phosphorite deposits; (12) metamorphic deposits; (13) lignite; (14) black shale deposits; and (15) other deposits (Bowell et al., 2011; Burns and Finch, 1999; Ceyhan, 2009; Cuney, 2009; Finch and Murakami, 1999; Frondel, 1958; Hazen et al., 2009; Seidel et al., 1993; I.A.E.A. et al., 2006; Kyser and Cuney, 2008; Morss et al., 2011; Peehs et al., 2005; Plant et al., 1999).

The chief sources of uraninite have historically been hydrothermal vein deposits, accounting for most of the world's historical uranium supply (Sharpe and Fayek, 2011). In such deposits, the uraninite usually occurs as fine-grained material commonly known as pitchblende (Ford, 1993). In more recent times, uraninite has been found to be associated with almost all deposit types (except the highly oxidised surficial deposit types) with most modern production coming from unconformity-related (Alexandre et al., 2009; Kyser and Cuney, 2008; Raffensperger and Garven, 1995), sandstone-hosted (Adams, 1991; Lovley and Phillips, 1992), quartz-pebble conglomerate (Robb and Meyer, 1995; Vennemann et al., 1992), vein (Kyser and Cuney, 2008) and breccia complex deposits (Reeve et al., 1990; Reynolds, 2000). Table 2 summarises a range of data including location, grade, deposit classification, and mineralogy from uraninite-containing deposits. Although dominated by uraninite, the data shows a variety of uranium-bearing minerals and gangue minerals associated with different deposit types. All have the potential to impact acid leaching of uraninite through increased consumption of reagent (gangue) or through reduced uranium extraction rates (other more refractory uranium minerals).

2.2. Structure and composition

Uraninite is commonly depicted using the chemical formula UO_2 . It is isometric (fluorite structure, space group $Fm\bar{3}m$, cell parameter $a = 5.4682 \text{ \AA}$, unit cell volume $V = 163.51 \text{ \AA}^3$) with the U^{4+} coordinated by eight O atoms in a face centred cubic arrangement with each O atom bonded to four U^{4+} cations at the corner of a regular tetrahedron (Fig. 1) (Belle, 1969; Fayek et al., 2000; Forster, 1999; Frondel, 1957; He et al., 2010; Kotzer and Kyser, 1993; Krivovichev and Plášil, 2013; Pouchet and Gauthier-Lafaye, 1998; Wilson et al., 1961). Uraninite that contains exclusively tetravalent U^{4+} is unknown in nature. Naturally occurring uraninite always has a portion of the uranium partially oxidised giving the general formula UO_{2+x} , with x typically in the range of 0.0 to 0.25–0.30 (Belle, 1969; Finch and Murakami, 1999; Janeczek and Ewing, 1991).

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