



Modeling the Key Lake uranium mill's bulk neutralization process using a pilot-scale model



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ABSTRACT

A pilot-scale model of the Key Lake bulk neutralization process (KLBN), termed the synthetic raffinate neutralization system (SRNS), was constructed to study the geochemical controls and the secondary mineralogy of uranium mine tailings deposited into the Deilmann Tailings Management Facility in northern Saskatchewan, Canada. Comparison of the synthetic raffinate with samples collected from the KLBN indicates that the pilot-scale model successfully simulated the full-scale process. Elemental analyses show that the precipitation trends of major ions and trace elements in the SRNS are consistent with what occurs in the KLBN. X-ray diffraction patterns, attenuated total reflectance-infrared and Raman spectra show that the bulk mineralogy of SRNS precipitates is consistent with KLBN precipitates, namely 2-line ferrihydrite with adsorbed arsenate, amorphous Al(OH)₃, and a Mg–Al hydrotalcite-like layered double hydroxide. The mineral phases precipitated in both systems are supported by thermodynamic modeling. Minor mineralogical differences between the SRNS and KLBN were attributed to low aqueous Fe(II) and SO₄²⁻ concentrations in the synthetic raffinate.

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1. Introduction

The Key Lake mill, located in northern Saskatchewan, Canada (57°13'N, 105°38'W), is the largest uranium mill in the world, producing 8.85 Mkg U₃O₈ per year from an average mill head grade of 4.61% U₃O₈ in 2012 (Cameco, 2013). Since 1999, the mill has been processing ore from the McArthur River mine, located 80 km to the north (Bharadwaj and Moldovan, 2005). The U is extracted from the ore using a sulfuric acid leach followed by solvent extraction and refining. The milling process can be divided into six steps: crushing/grinding, ore receiving and blending, leaching, counter current decantation, solution pre-treatment and solvent extraction, yellowcake (U₃O₈) precipitation, and calcining and packaging (Cameco, 2010). A bulk neutralization process (hereafter termed the Key Lake bulk neutralization process; KLBN) is used to treat the U-barren waste streams (termed raffinate) before their release into the environment. Precipitates formed during raffinate neutralization are mixed with leach residue and have been discharged as tailings to the in-pit Deilmann Tailings Management Facility (DTMF) since 1996 (Bharadwaj and Moldovan, 2005).

A goal of the KLBN is to sequester elements that can cause adverse health effects to biological systems into geochemically stable tailings solids, mitigating their release to the receiving environment. In the case of the Key Lake mill tailings, elements of concern (EOCs) include

As, Mo, Ni, and Se dissolved from the ore during the leaching process (Shaw et al., 2011). Due to the potential environmental impacts of EOCs, research efforts have focused on defining the mineralogical controls in the tailings body. The mineralogical controls on tailings deposited in the DTMF, and in other U tailings management facilities in Saskatchewan, are dominated by gypsum (CaSO₄·H₂O), 2-line ferrihydrite (FH), Mg–Al hydrotalcite, Al oxides, and poorly crystalline scorodite, among other minor secondary mineral phases that adsorb and/or precipitate EOCs (De Klerk et al., 2012; Essilfie-Dughan et al., 2013; Gomez et al., 2013; Mahoney et al., 2007; Moldovan and Hendry, 2005; Moldovan et al., 2003; Shaw et al., 2011). While several of these mineralogical controls are well understood, minor changes to the ore and milling process could affect EOC sequestration in the tailings. To improve our understanding of the current mineralogical controls on EOCs in the Key Lake U mill tailings and to characterize the effects of potential changes to the mill process and/or ore bodies on these mineralogical controls, a lab-scale bulk neutralization pilot plant (hereafter termed the synthetic raffinate neutralization system; SRNS) was constructed to simulate the present-day KLBN and the tailings it generates for storage in the DTMF.

Prior to its use in experiments to define mineralogical controls or the impact of future changes to the mill feed or process, the SRNS must be demonstrated to accurately reproduce the aqueous and solid phases produced during operation of the KLBN. As such, the main objective of this study was to evaluate if the SRNS can replicate the secondary mineralogy and geochemical controls of the present-day KLBN. Synthetic

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raffinate was tailored to approximate the chemistry observed in the Key Lake raffinate, and the SRNS operated at pH values representative of target and actual concentrations measured in the KLBN. The resulting composition of elemental aqueous and solids was determined by inductively coupled plasma mass spectrometry (ICP-MS). Mineralogical characteristics of SRNS and KLBN solids were determined by X-ray diffraction (XRD), attenuated total reflectance infrared (ATR-IR) and Raman spectroscopy. Thermodynamic modeling was performed to support observations from the characterization techniques.

2. Materials and methods

2.1. Key Lake bulk neutralization process

The purpose of the KLBN is to treat contaminated solutions originating from several mill processes including raffinate (i.e., U-barren effluent) from the counter current decantation and solvent extraction processes, reverse osmosis reject water, porewater and seepage from the tailings management facilities, tailings thickener overflow water, waste water used in mill areas, and runoff from operational areas. The scope of this study limited these treatment streams to solely the raffinate because it is the main source of EOCs (Lieu et al., 2010). The raffinate is acidic (nominally pH 1) and oxic ($\sim E_h + 600$ mV) and contains elevated concentrations of EOCs (Liu and Hendry, 2011). The majority of the EOCs are precipitated as secondary minerals in the KLBN via the addition of slaked lime ($\text{Ca}(\text{OH})_2$). These solids are mixed with leach residue and discharged into the DTMF for long-term storage.

In the KLBN (Fig. 1), the raffinate flows by gravity through a series of air agitated mixing cells (termed pachucas) and thickeners. The raffinate at pH 1.0 is fed to pachuca 1 (P1) for mixing. From P1, the raffinate flows by gravity to P2 where lime is added to increase the pH to 3.5–4.2. The solution is then pumped to the Se–Mo Thickener and flocculent is added to enhance solid agglomeration and settling properties. The aqueous overflow from the Se–Mo Thickener is then pumped to P3 where lime is added to increase the pH to 6.5. From P3, the solution is pumped to P4 where more lime is added to increase the pH to 9.2. The resulting slurry is pumped to the Lamella Thickener, which serves to primarily collect the Al oxide mineral phases. The underflow

containing the secondary precipitates from both the Se–Mo and Lamella Thickeners are combined and pumped to the DTMF Thickener and raised (via $\text{Ca}(\text{OH})_2$) to a terminal pH of 11 prior to discharge into the DTMF. The overflow from the Lamella Thickener reports to the Radium Removal Circuit where H_2SO_4 is used to incrementally decrease the pH to 6.0 via three reactors (pH 7.5, 6.5, and 6.0). This solution flows by gravity to the Radium Removal Thickener where flocculent is added. The overflow from the Radium Removal Thickener is the final effluent and is released to monitoring ponds on site prior to final release to the environment. The underflow from the Radium Removal Thickener is pumped to the DTMF Thickener. Barium chloride (BaCl_2) is added at P2, P4, and in the Radium Removal Circuit to reduce ^{226}Ra activity in the final effluent via precipitation (Liu and Hendry, 2011).

2.2. Synthetic raffinate neutralization system

The SRNS (Fig. 2) was designed to operate at ambient temperature (approximately 22 °C) and process approximately 200 L of synthetic raffinate solution in 5.5 days at a flow rate of 25 mL/min. The scale of each unit in the SRNS was based on reproducing retention times equal to corresponding units of the KLBN at an overall flow rate of 25 mL/min. To achieve this, the scale of the SRNS to the KLBN ranged between 1:130,000 and 1:245,000. Tracer tests using deuterium oxide (data not presented) demonstrated that the total retention time of the SRNS (19.8 h) matched that of the KLBN (19.6 h). The SRNS consisted of four reactors (to represent the pachucas) and three thickeners.

The reactors were constructed from 127 mm ID opaque PVC pipe, with caps, fittings, and baffles (all PVC; Fig. 3a,b). The dimensions of the baffles were constructed as specified by Green and Perry (2008). The reactors were agitated at 500–600 rpm using IKA® RW 20 Digital laboratory mixers with 316 SS impellers and ring guards (Fig. 3b). The thickeners (Fig. 4a,b) were constructed from 76 and 102 mm internal diameter clear PVC pipe and opaque PVC caps and fittings. A glass wool filter was fitted between two HDPE mesh screens (approximately 9 mesh) near the top to minimize the carry-over of solids into subsequent reactors. All reactor and thickener components were fitted and sealed using Devcon® Plastic Welder™ II due to its resistance to extreme acidic and alkaline conditions. The pH of each reactor was controlled using a Cole-Parmer double junction pH electrode (EW-27001-

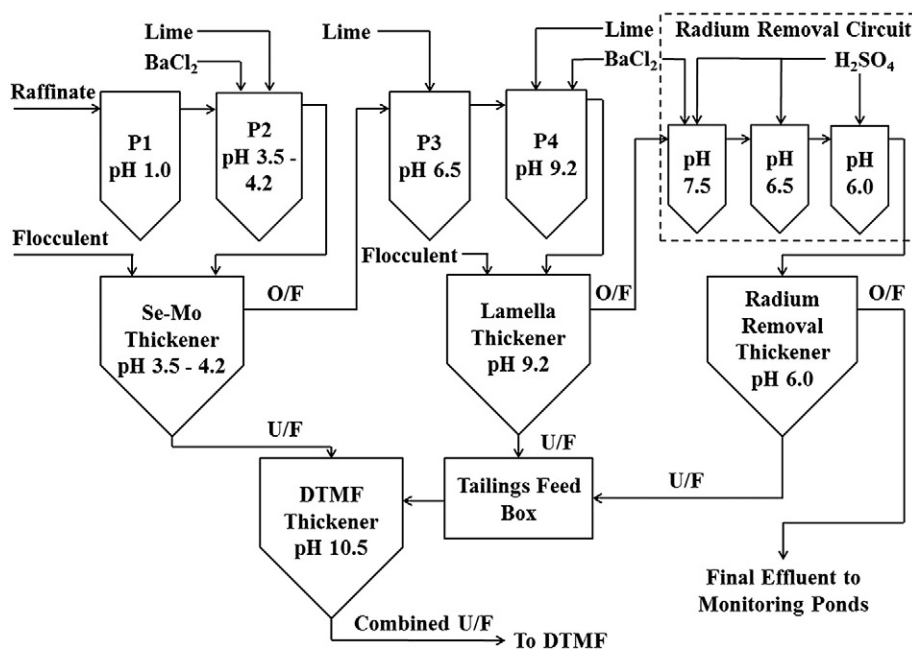


Fig. 1. Block flow diagram of the Key Lake bulk neutralization process. Only major units of the process are displayed. Overflow and underflow of thickeners are indicated by O/F and U/F, respectively.

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