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Early evolution of weathering and sulfide depletion of a low-sulfur, granitic, waste rock in an Arctic climate: A laboratory and field site comparison

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ABSTRACT

Leachate from a humidity cell experiment provided a geochemical framework to evaluate the early evolution of weathering of the same waste rock in an Arctic environment. Comparison of laboratory and field results indicates the hydrogeochemical system within the higher sulfide (0.01–0.27 wt.% S) waste rock pile has attained a peak weathering state, indicated by a shift to acid neutralization by weathering products of Al-bearing minerals, stabilization of the pH near 4.5, mobility of metals from sulfide and Al-bearing minerals, and a highly correlated relation between SO_4 release and outflow from the waste rock pile. Further weathering of the waste rock should be driven by external environmental factors of temperature and precipitation/infiltration instead of internal factors of wetting and transient acid-neutralization processes. An evaluation of sulfide depletion indicates that 4% of the sulfide in the <6.3-mm fraction of this waste rock pile, corresponding to 2% of the sulfide in the <50-mm fraction, has been removed through oxidation and leaching as SO_4. Weathering is strongly seasonal because of the Arctic climate, which produced a daily sulfide-depletion rate corresponding 0–0.02% and a peak annual depletion of 100 kg or 1.5% of the remaining sulfide content in the <6.3-mm fraction. Peak sulfide weathering is expected to continue until about 15% of the available sulfide is depleted, similar to an observed decrease in sulfide weathering in the laboratory. Estimates of the reactive surface area of the sulfide minerals and a peak rate constant were used to evaluate the sulfide percent in the fine fraction of rock in the pile undergoing weathering during the annual freeze-thaw cycle, which can be used to estimate a climate rate factor to adjust the weathering mass through the seasonal changes. For modeling of future leachate, laboratory results indicate that an exponential rate limiting factor is necessary to account for slowing of sulfide oxidation after peak weathering because of the formation of secondary minerals that inhibit element release.

1. Introduction

The oxidation of sulfide minerals in waste rock can produce acidic leachate or acid rock drainage (ARD) characterized by low pH, elevated SO_4 concentrations, and greater mobility of associated transition metals such as Co, Cu, Fe, Ni, and Zn (Blowes et al., 2003; Nordstrom and Alpers, 1999; Nordstrom and Southam, 1997). The release of ARD from waste rock can be a significant environmental concern because it can have long-term impacts on local and regional water resources. The amount of ARD produced is affected by microscale properties such as mineral composition and sulfide surface area that influence reactive processes (Blowes et al., 2003; White and Peterson, 1990) and macroscale properties such as precipitation infiltration, flow pathways, and atmospheric conditions that influence transport of the elements from waste rock piles (Kyhn and Elberling, 2001; Ritchie, 1994). The Diavik Waste Rock Project was implemented to examine the potential for and extent of ARD generation from a low-sulfur, granitic, run-of-mine (ROM) waste rock in an Arctic environment. Significant parts of the project included a long-term (years) humidity cell experiment and long-term (years) monitoring of leachate from two mine-site, experimental, waste rock piles (test piles). Geochemical results of leachate from the humidity cells and test piles were used to evaluate the early evolution of leachate geochemistry and rate of sulfide depletion under laboratory and field conditions to better understand the generation of ARD with this waste rock in this Arctic climate.

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The humidity cell and test pile experiments were conducted to evaluate the potential for ARD and mineral weathering rates—the rate at which primary minerals are transformed to secondary minerals or dissolved reaction products, congruently or incongruently, with release of elements (Ardau et al., 2009; Sapsford et al., 2009). Much of the weathering of waste rock occurs in the fine grained material, similar to the grain size fraction used for humidity cell experiments (Stockwell et al., 2006; Strömberg and Banwart, 1999). Predicting field conditions from humidity cell results can be difficult due to the accelerated weathering of the laboratory samples and the coupled and sustained biogeochemical processes in the field that create a more complex environment (Bowell, 2002; Bowell et al., 1999; Lefebvre et al., 2001; Malmström et al., 2000; Nordstrom and Alpers, 1999). Differences in field and laboratory environments must be evaluated to determine appropriate rate and scaling factors for prediction of ARD (Malmström et al., 2000). This study used the evolution of leachate geochemistry from the laboratory results to understand the influence of the Arctic freeze-thaw cycle on the weathering evolution and sulfide depletion within a waste rock pile.

2. Site and sample description

The Diavik Diamond Mine (Diavik) is located on an island in Lac de Gras in the Northwest Territories, Canada (Fig. 1). The site is in the Canadian Arctic: a permafrost, polar climate area with an annual precipitation of less than 300 mm (40% as rain, 60% as snow) and average minimum, mean, and maximum temperatures of −31, −9, and 18 °C, respectively (Environment Canada, 2014). Following a May/June snowmelt, precipitation occurs as rain until a rain-snow transition in October. An active freeze-thaw zone extends about 4 m into the bedrock at the site and deeper into unconsolidated materials (Pham et al., 2013). Wetting of the test piles occurs at a rate of 0.2–0.4 m d^{-1} (matrix pore water velocity) during typical rainfall events, although a rate of 5 m d^{-1} (macropore flow) is possible with intense rainfall (Neuner et al., 2013). The initial wetting of the test piles likely occurred during the first two years after construction of the piles (Fretz, 2013).

The mine's waste (gangue) rock is composed of about 75% granite, 14% pegmatitic granite (pegmatite), 10% biotite schist, and 1% diabase dykes (Blowes and Logsdon, 1998). The Archean granite and pegmatite are massive and moderately to coarsely crystalline, and the metasedimentary schist occurs as xenoliths within the granite. The granites are primarily quartz [SiO_2], K-feldspar [$KAlSi_3O_8$], and albite [$NaAlSi_3O_8$], with greater albite in the granite and greater K-feldspar in the pegmatite (Jambor, 1997). The biotite schist is composed primarily of albite (35–55%), quartz (20–50%), and biotite [$KMg_3AlSi_3O_{10}(OH)_2$] (10–25%) and has an average sulfide content of 0.24 wt.% S (range 0.02–0.42 wt.% S). The sulfide is principally pyrrhotite [Fe_7S_8] with traces of pyrite [FeS_2], chalcopyrite [$CuFeS_2$], and sphalerite [(Zn,Fe)S]. The waste rock is segregated prior to disposal according to total S content: low, <0.04 wt.%; intermediate, 0.04–0.08 wt.%; and relatively high, >0.08 wt.%, which corresponds to mine-site designations of Type I, Type II, or Type III waste rock, respectively.

Differences in grain-size distribution, saturation level, and environmental conditions are problematic for scaling laboratory weathering results to field conditions (Lapakko, 2003; Malmström et al., 2000; Velbel, 1993). Humidity cells typically contain grain-size fractions <6.3 mm in diameter (ASTM, 2013), because most sulfide oxidation is expected to occur with the more available sulfide minerals of the fine-grain fraction. Grain-size and mineral analyses indicated that 18.5 vol.% of the test piles was <6.3 mm, this fraction can hold water by capillarity, and the sulfide minerals are concentrated in this fraction (Neuner et al., 2013; Smith et al., 2013b). Wind-driven advection through the large clast mix of the test piles produces well-oxygenated conditions, and the Arctic climate produces an annual freeze-thaw cycle (Amos et al., 2009b; Chi et al., 2012; Pham et al., 2013). Less than 50% of precipitation typically infiltrates into the test piles (Neuner et al., 2013). A minimum 5-mm rainfall event is necessary for infiltration (Neuner et al., 2013) unless it occurs during the lower temperature months in spring and fall when evaporation is lower (Fretz, 2013). The annual freeze-thaw cycle of the test piles results in fully frozen conditions in the late fall and a gradual thawing through late spring and summer with reverse gradients (Fig. 2) between freezing (cooling inward) and thawing (warming inward) (Pham et al., 2013).

3. Materials and methods

Waste rock samples were collected in 2004 and 2005 from the mine's waste rock streams (differentiated by S content) for laboratory weathering. The samples (Table 1) include four types: A (relatively low sulfide and low carbonate (Type I)), B (intermediate sulfide and low carbonate (Type II)), C (intermediate sulfide and high carbonate (Type III)), and D (relatively high sulfide and low carbonate (Type

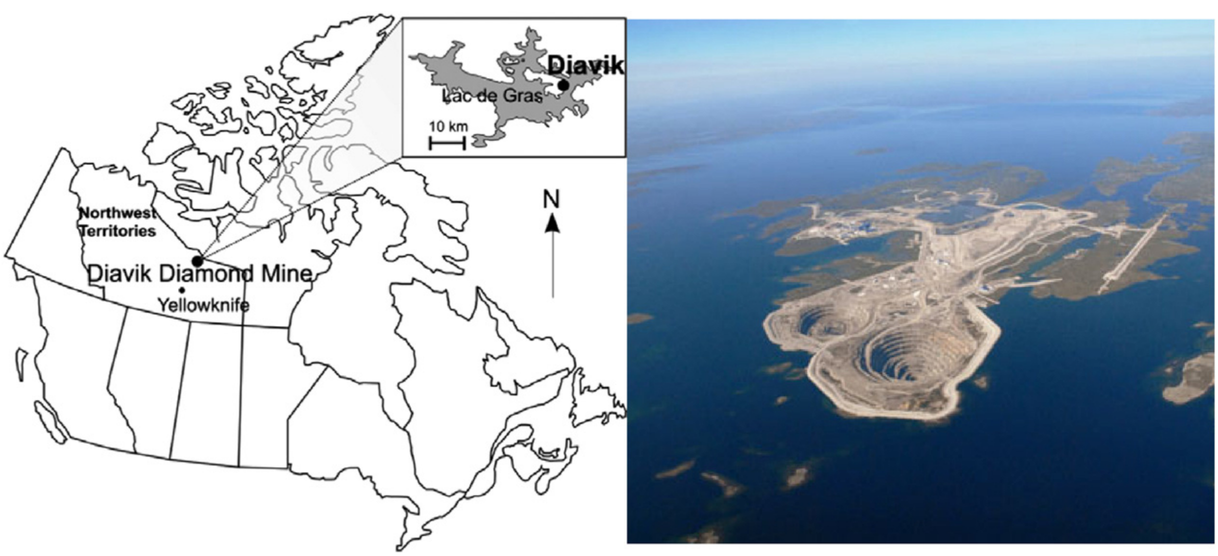

Fig. 1. The Diavik Diamond Mine is located on East Island in Lac de Gras, 300 km northeast of Yellowknife, NT and 220 km south of the Arctic Circle (Smith et al., 2013c; photo courtesy of Diavik Diamond Mines, Inc.).