



Physicochemical gradients, diffusive flux, and sediment oxygen demand within oil sands tailings materials from Alberta, Canada[☆]



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ABSTRACT

The Athabasca Oil Sands contain one of the world's largest oil reserves consisting of approximately 168 billion barrels of currently recoverable bitumen. With 20% recoverable through open pit mining methods, this extraction process produces a considerable amount of fluid fine tailings (FFT) waste material, which must be deposited on site in tailings ponds. These ponds allow the waste sand, clay and residual bitumen to settle out of the water column, allowing for the water to be recycled for use again in the extraction process. It is vital to gain a better understanding of the processes contributing to the development of physicochemical gradients (pH, Eh, Oxygen etc...) that form in these tailings ponds over time, with the goal of remediation and subsequent construction of end-pit lake systems once oil extraction has ceased. To differentiate between the impacts of biotic and abiotic processes in fresh (newly processed material) and mature FFT (~38 year old tailings) over a 52-week study, a specific experimental design was utilized in accordance with novel microsensor profiling techniques. The sulfide diffusive fluxes within mature biotic systems measured $37.6 \mu\text{mol m}^{-2} \text{day}^{-1}$ at the onset of the experiment, decreasing over time, as FeS mineralization progressed. In addition, DO fluxes also showed strong correlation to the physical affects of consolidation, and overall biological consumption of O_2 at the FFT-water interface. This holistic study comparing different tailings pond materials provides insight regarding biotransformation and physicochemical controls effecting sediment oxygen demand associated with reclaimed wetlands and end pit lake development.

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1. Introduction

Producing approximately 1.9 million barrels of bitumen a day, the Athabasca oil sands in Alberta, Canada continues to increase production rates as the conventional oil reservoirs decline globally and more efficient, less costly extraction techniques are developed (Alberta Energy, 2013; Williams, 2003; CAPP, 2014). During open-pit mining of bitumen, separation of the bitumen from raw ore through hot water extraction, results in large amounts of oil sands process-affected materials (OSPM) (CAPP, 2014). This initial tailings slurry consists of 55 wt% solids (silts, clays and sands), and contain ~1% residual bitumen, and alkaline process-affected waters (Chalaturnyk et al., 2002), which are pumped into large settling basins known as tailings ponds, where the solids are allowed to

settle out, forming semi-consolidated fluid fine tailings (FFT). Compliance with a zero-discharge policy is mandatory, requiring oil sands companies to provide suitable containment for OSPM. The basins themselves allow for the recycling of process waters to reduce consumption of fresh water sources (Alberta Energy, 2013). Reclamation strategies will result in the formation of either reclaimed wetlands and/or end-pit lake (EPL) environments within the reclaimed landscape. Vacant mine pits will be filled with both highly consolidated tailings and likely an addition of natural wetland or lake sediments into the mix of tailings sediments aiding in the efficiency of natural reclamation, and ecosystem sustainability. Given the scale of these tailings environments however, it is vital to understand the chemical and biological processes at the tailings-water interface, and how these interactions dictate or influence the long-term fate of such EPLs or remediated wetland environments, prior to the addition of natural or uncontaminated sediments to the ecosystem. To date these comparative, baseline and fundamental geochemical studies are lacking with respect to different tailings environments within the Athabasca Oil Sands.

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The geochemical gradients and corresponding chemical diffusive fluxes within the FFT, as with any aquatic or marine sediment, can change significantly during the consolidation process and aging. What is becoming more apparent is that the FFT material may be chemically complex and dependent on operator, therefore harboring a complex microbial consortium (Fedorak et al., 2002; Penner and Foght, 2010; Chi Fru et al., 2013). Further the biogeochemical activity within the FFT will have a direct impact on the quality and composition of the cap water. Depletion of dissolved oxygen (DO), along with other nutrient cycles such as iron cycling, denitrification and the production of both hydrogen sulfide ($\text{H}_2\text{S}_{(g)}$ or HS^-) and methane (CH_4) are common indicators of reducing conditions that can directly or indirectly influence aquatic organisms inhabiting these environments (ex. Broderius et al., 1977; US National Research Council, 1979; DiToro et al., 1990). These redox governing processes are commonly observed not only in saturated tailings materials, but also observed in natural marine and wetland settings where organic matter is prone to degradation, and other terminal electron acceptors will dominate in the absence of O_2 (Froelich et al., 1979). These standard redox ladders beginning with aerobic respiration and ending with methanogenesis ordered from most to least favorable energy yield are common formulas found throughout the geochemical literature (e.g. Stumm and Morgan, 1996).

However, as is often the case in real world scenarios, this textbook understanding of the thermodynamic redox ladder fails to illustrate some natural systems (Bethke et al., 2011). Bethke et al., 2011 observed no fixed thermodynamic hierarchy between iron reducers, sulfate reducers (SRBs) and methanogens. Natural systems, for example, may allow for sulfate reducers to be active alongside iron reducing processes (Bethke et al., 2008), indicating microbial activity is not governed by usable energy levels alone. In addition, the prescribed competition for electron donors was not observed in that study. Instead a co-operative existence was observed, where, for example, Fe^{2+} and HS^- was produced in a one-to-one relation forming precipitated FeS minerals (Bethke et al., 2011). This mechanism was also observed by Ramos-Padron et al. (2011), where large concentrations of dissolved HS^- reacts with iron in oil sands tailings material, forming extensive FeS mineralization, trapping a large proportion of sulfides within the tailings. Previous microcosm tailings research has also shown this characteristic FeS discoloration in the upper-most FFT layers (Stasik et al., 2014; Chen et al., 2013).

In addition to the cycling of Fe and S within these systems, methanogenesis also plays a significant role in the carbon degradation processes, and is perhaps more complex in nature. From a metabolic energy perspective it is the least satisfying for microbial uptake of organic matter (assuming a thermodynamic hierarchy of redox processes) and is a concern due to possible greenhouse gas (GHG) emissions. Several studies provide insight into the competition between sulfate reducing bacteria and methanogens in oil sands environments (Holowenko et al., 2000; Fedorak et al., 2002). Though, as described earlier, perhaps this competition for electron donors is more of a delicate balance between the sulfate reducers and methanogens (Bethke et al., 2011), thus increasing our need for a holistic understanding of these systems from a broad biogeochemical framework.

Sediment oxygen demand (SOD) is an indicator of the water column quality, indicating DO concentrations directly influenced by processes at and below the tailings-water interface (Chen et al., 2013). The term applies to both biological (BOD) and chemical (COD) processes dominating sediments, and represents the consumption of DO by the various chemically driven processes, as well as the decay of organic matter through aerobic respiration (APHA, 1992). This consumption of oxygen at the sediment-water

interface can exceed the downward flux of atmospheric O_2 within the water column leading to hypoxia. It is generally accepted that DO levels below $\sim 2 \text{ mg L}^{-1}$ ($62.5 \mu\text{mol L}^{-1}$) are too low to sustain most fish species, and at levels below $\sim 6 \text{ mg L}^{-1}$ ($187.5 \mu\text{mol L}^{-1}$) some cold water fish species (i.e. Salmon, trout etc.) are unable to reproduce (Carter, 2005).

Chen et al. (2013) investigated this DO behavior and the sulfur cycling associated with FFT collected from West-In Pit Lake (WIP) storage basin operated by Syncrude Canada Ltd. The study identified the aforementioned sulfidic zone within biotic systems, providing insight into the microbial role in the cycling of S within that FFT material. Further, in oxic systems, O_2 consumption was enhanced in the biotic systems compared to the abiotic (sterile) controls, due in part to the biota forming reduced reactants over time. However, both abiotic and biotic treatments reported negligible O_2 within the FFT at the onset of the experiment, suggesting that both biologically and chemically driven processes contribute to overall SOD, though the DO gradient of the biotic system indicated increased consumption, thus $\text{BOD} > \text{COD}$ (Chen et al., 2013). Hydrogen sulfide production showed a positive correlation with the biotic system, with HS^- concentrations in the FFT reaching $86 \mu\text{mol L}^{-1}$ after 5 weeks, while the abiotic systems remained at a background of $< 10 \mu\text{mol L}^{-1}$ throughout their study (Chen et al., 2013).

To gain a comprehensive understanding of the geochemical nature of various sediments and tailings, it is often beneficial to determine diffusive flux measurements. The diffusive chemical flux is a term used to describe the mobility of molecules within a certain substance, transporting from an area of high concentration to low concentration, directly correlating to the steepness of a measured concentration gradient. Diffusive flux measurements across water-sediment boundaries have been studied extensively over several decades (Boudreau and Guinasso, 1982; Jørgensen and Revsbech, 1985; Jørgensen and Marais, 1990; Lorke et al., 2003). The so-called diffusive boundary layer (DBL), or benthic boundary layer (BBL) creates distinct concentration gradients, and has direct impact on the overlying water column (Brand et al., 2007). In this thin layer above the sediment, the dominant transport mechanism is molecular diffusion (Gundersen and Jørgensen, 1990), therefore, it is highly advantageous to determine diffusive flux values across this interface in accordance with both O_2 and HS^- concentrations. Diffusive flux across the DBL directly relates to the physical properties of the FFT, and can correlate directly to the diffusive properties within its mineralogical matrix. Furthermore, biofilm structure, be it thin and porous or tightknit and thick affect the diffusive characteristics of various molecules across the DBL (Tijhuis et al., 1994).

In our study, comparisons are made of the geochemical development of FFT materials collected from two different tailing pond sources from Suncor Energy Inc., representing both fresh and aged tailings. Previous research has focused on single pond characterization, as far as these researchers are aware (ex: Chen et al., 2013; Stasik et al., 2014). The aged tailings are from an inactive, water recycling pond containing FFT approximately 38 years old while the fresh tailings are acquired from an active tailings pond, which receives fresh mixed tailings on an ongoing basis. This study will investigate the behavior of DO and S across the DBL associated with two different FFT materials using a series of microcosms after Chen et al., 2013. Tracking the DO concentrations in the water column over the course of this study will give insight into the long-term SOD, and whether or not DO increases or decreases over time. Further, a brief summary of the distinct redox zonation will be presented, explaining influences on the cycling of key elements such as Fe and S species within each respective pond. Specific diffusive flux measurements will be presented in order to gain an

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