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Turbidity Mitigation in an Oil Sands Pit Lake through pH Reduction and Fresh Water Addition

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Abstract:

Currently, tailings ponds are used to contain oil sands process-affected water (OSPW) and fluid fine tailings (FFT) generated from the Alberta oil sands mining operations. In 2016, the Government of Alberta introduced a tailings management directive providing guidance for reclamation efforts in Alberta's oil sands mining projects. Pit lakes, one of the proposed reclamation strategies, are designed to support a healthy aquatic ecosystem, and to provide a storage location for FFT and OSPW. In 2013, Syncrude Canada Ltd. began testing a full scale demonstration pit lake project, named Base Mine Lake (BML). However, high turbidity (~71-231 Nephelometric Turbidity Units [NTU]) has been observed in the BML cap water due to the resuspension of FFT; this turbidity inhibits the development of a healthy aquatic ecosystem. The goal of this water column study, which tested two different water dilutions (60% OSPW and 20% OSPW), was to improve BML cap water clarity by reducing pH through CO_2 addition. Of these mixtures, only the one containing 60% OSPW with added CO_2 showed significant water clarity improvement ($P = <0.001$), and a ~60 day lag period preceded a rapid increase in water clarity (96.8% turbidity reduction). By contrast, only minor improvement in water clarity was observed for other mixtures: 20% OSPW with CO_2 added (77.9% turbidity reduction) and 60%

OSPW with no CO_2 added (81.6% turbidity reduction). Taken together, these results suggest that the water clarity can be improved by CO_2-induced pH reduction, and that the ionic strength of the solution is an important factor in turbidity removal through CO_2 addition.

Introduction

Alberta's oil sands deposits are among the largest in the world (Government of Alberta 2014). Surface mining and bitumen extraction processes generate by-products, such as fluid fine tailings (FFT) and oil sands process-affected water (OSPW; Humphries 2008). As oil sands mining companies currently do not have regulatory approval to discharge OSPW, these by-products are contained in tailings ponds (Government of Alberta 2015). Fluid fine tailings in the tailings ponds settles and releases process water, which is reused in the bitumen extraction process, thereby reducing freshwater consumption. However, it has been estimated that FFT will take 125 to 150 years to completely consolidate though gravity settling (Eckert et al. 1996). The recent rapid expansion of oil sands operations has generated a pressing need for permanent reclamation strategies for FFT and OSPW. A new directive (085), issued by the Government of Alberta in 2016, specifies that new fluid tailings must be ready to reclaim ten years after the end of mine life, and all legacy tailings be ready to reclaim and by the end of mine life (Alberta Energy Regulator 2016). The main objective of tailings reclamation is to support the transformation of oil sands mining projects (including mined or disturbed lands) into "a self-sustaining boreal forest ecosystem" upon completion of mining (Alberta Energy Regulator 2016).

Pit lakes are among the reclamation and storage strategies currently being investigated. In 2013, Syncrude Canada Ltd. commissioned a full scale demonstration pit lake named Base Mine

Lake (BML), in which a mixture of OSPW and fresh water was placed on top of a layer of FFT (Dompierre et al. 2016). Additional fresh water is added during the summer months from the Beaver Creek Reservoir (BCR; an adjacent fresh water body) to simulate runoff from the reclaimed landforms that will eventually make up the watershed of BML (Dompierre et al. 2016). Previous studies have found that typical FFT samples contain 30–40% w/w solids, and the particle size distributions are 10% >44 µm, 57% 1–44 µm and 33% <1 µm (Dompierre et al. 2016; Voordouw 2013). About 34% and 26% of clay in oil sands FFT are kaolinite and illite, respectively, with a size range of 0.2–2 µm (Kaminsky et al. 2009). The small size of the fine clay particles (between 0.2 and 2 µm) means they will likely remain suspended upon disturbance (from wave actions and/or seasonal turnover events), reducing water clarity (Charette 2010; BGC Engineering Inc. 2010). Water clarity is an important parameter for the development of healthy aquatic ecosystem, since it impacts nutrient recycling processes and dissolved oxygen replenishment (Tundisi and Tundisi 2012). The impact of suspended clay particles on water clarity in the BML demonstration needs to be better understood prior to the full-scale implementation of pit lakes as an oil sands reclamation strategy.

The stability of suspended clay particles can be influenced by several parameters, such as pH and ionic strength (Siddique et al. 2014; Zbik et al. 2008). Several studies have shown that decreasing the pH of FFT with dissolved carbon dioxide (CO_2) can destabilize suspended clay particles and improve fine tailings densification (Siddique et al. 2014; Ibanez et al. 2014; Zhu et al. 2011). However, limited research has been done to study the effect of CO_2 addition on water clarity.

The current laboratory study explores the effect of CO_2 addition on turbidity reduction in the BML water cap. It is hypothesized that CO_2-induced pH reduction will flocculate suspended clay particles, thus reducing the turbidity in the BML cap water.

Materials and Methods

Materials

Fluid fine tailings (FFT), Oil Sands Process-Affected Water (BML-OSPW), and fresh water from the Beaver Creek Reservoir (BCR) were collected and provided by Syncrude Canada Ltd. in 2013. FFT was collected from BML at a depth of 12 m below the FFT-water interface (from the SW platform) in July. The BML-OSPW was collected from the surface layer of BML (from the SW platform) in October, and represents a 90% OSPW and 10% BCR fresh water mix. All samples were stored at 4°C until use.

FFT characterization

FFT was characterized prior to the experiment. Key analysis included zeta potential, solids content, methylene blue index (MBI), total carbonate minerals, and X-ray powder diffraction (XRD). The zeta potential was measured using a Zetasizer Nano ZSP (Malvern Instruments Ltd.). MBI was determined using the CONRAD method previously described (Kaminsky 2014), on four replicate FFT samples and the results were averaged. Solids content was determined for five samples by drying overnight at 105°C as described in ASTM D5907 (ASTM 2013) with their results averaged. The total carbonate minerals in the FFT solid phase were analysed using the acid digestion method previously described (Siddique et al. 2014). XRD analysis of the powdered FFT and sediments from BML-OSPW samples was performed using a Geigerflex powder x-ray diffractometer equipped with a D/Tex detector. XRD data was analyzed using JADE 9.1 software, with peak locations being referenced using the International Centre for Diffraction Data (ICDD 2016) and Inorganic Crystal Structure Databases (ICSD 2016).

Experimental setup

To evaluate the effect of pH change on turbidity reduction, two Townsend acrylic columns (2.5 m in height, 0.305 m in diameter, 6.35 mm wall thickness) were used. Each acrylic column was equipped with 10 Swagelok SS Quarter-Turn Instrument controlled sampling ports (at the heights of 0.4, 0.8, 1.2, 1.6, and 2 m from the bottom; Fig. 1). FFT was placed at the bottom of the column (0.2 m deep) and capped with various ratios of water mixtures (columns filled with 2 m of water; Fig. 1). To test the effect of OSPW dilution on the CO_2-induced turbidity reduction, various BML-OSPW to BCR fresh water ratios were tested (Fig. 1). Either 60/40 or 20/80 OSPW:BCR %v/v water (Table 3) were tested to represent the expected BML water cap composition in the short-term (~5 years) and mid-term (~10 years), respectively. A no-CO_2 control column containing a 60/40 water mixture was also constructed (Table 3).

CO_2 (99.9% purity PRAXAIR) was added to two of the columns using a SEAIR microbubbling diffusion system (SEAIR Inc.) at a rate of 4 L/min. The water in the column was withdrawn from the sampling ports at 1.40 m and 1.80 m above the water/FFT interface (Fig. 1) and passed through the diffusion chamber to supersaturate the fluid with CO_2. The CO_2-saturated water was then returned to the column via sampling ports at 0.60 m and 1.00 m above the water/FFT interface (Fig. 1). Approximately 4000 L (~17 hours) and 1000 L (~4 hours) of CO2 were used for treating 60/40+CO_2 column and 20/80+CO_2 column respectively.

Secchi depth and PAR light intensity measurements

Prior to Secchi depth and light intensity measurement, the side of the column was covered with an opaque curtain to minimize ambient laboratory light interference, and a globe workshop halogen work light (250 Watts, model # 60568) was placed (28.5 cm) above the column.

The visual water clarity was evaluated using a 20 cm diameter Secchi disk (Science First 78-010 Fieldmaster; Tundisi and Tundisi 2012).

The photosynthetically active radiation (PAR) within the column was measured using an LI-192SA underwater quantum sensor and LI-1400 data logger (LI-COR). The measured PAR data was used to determine the euphotic zone depth, the depth at which PAR intensity is 1% of its surface value (Kirk 1994).

Water characterization

Water samples were collected from each of the five ports, and various parameters were measured, including pH, conductivity, alkalinity, turbidity, concentration of naphthenic acids (NAs), concentration of cations and anions, total suspended solids (TSS), total dissolved solids (TDS), total organic carbon (TOC), and dissolved organic carbon (DOC).

pH was measured using an Accumet AR50 dual channel meter (Fisher Scientific). Conductivity was measured using an ExStik®II Meter (Extech). Alkalinity was measured using a Mettler Toledo DL53 with 0.02 N H_2SO_4 as a titrant. The TOC and DOC were measured using a Shimadzu DOC analyser (TOC-LCPH). Prior to DOC analysis, the liquid samples were filtered through a 0.45 µm membrane filter, and the filtrates were diluted by 1:10. The non-purgeable organic carbon (NPOC) method was used for the organic carbon analysis (Brown et al. 2013).

Turbidity was measured using an Orbecco-Hellige Digital Direct-Reading Turbimeter. TDS and TSS were measured following the ASTM D5907 standard method (2013).

Anion and cation concentrations were determined as previously described by (Abolfazlzadehdoshanbehbazari et al. 2013) and NA concentrations were determined as previously described (Mahdavi et al. 2015).

Geochemical modelling

The water chemistry profile within the column was estimated with PHREEQCi (Version 3.3.11.12535; Parkhurst and Appelo 2013), and mineral saturation indices (SIs) were calculated using the WATEQ4F (Ball et al. 1991) thermodynamic database. Piper diagrams were constructed using AqQA software (Version 1.5.0, RockWare Inc.).

Results and Discussion

Prior to the experiment, dissolved ion concentrations (Na^+, Mg^{2+}, Ca^{2+}, SO_4^{2-}, and Cl^-) from the FFT pore water were determined and found to be consistent with those previously measured in the field (Table 1; Dompierre et al. 2016). The physical parameters of the FFT were examined, and the solids content and MBI results were within the range previously reported (Table 2; Dompierre et al. 2016; Osacky et al. 2014). The XRD results showed that clay minerals (kaolinite and illite) and carbonate minerals (calcite) were present in both BML cap water and FFT (Fig. S1). The concentration of carbonate minerals within the FFT was also determined (Table 2).

Initial cap water chemistry was measured prior to CO_2 treatment (Table 3), and most of the parameters were found to be comparable to the values previously reported for tailings pond OSPW (Table S1; Charette 2010; Jiang et al. 2016). However, the concentration of certain ions (Cl^-, Na^+, K^+, and Ca^{2+}) and total dissolved solids were lower than previously reported OSPW values; this can be attributed to dilution of the BML water cap with fresh (BCR) water before the samples was collected (Table 2 and S1). The initial water chemistry for both 60/40 (no-CO_2 and +CO_2) columns were similar (Table 3).

The pH of the 60/40-noCO_2 column was stable throughout the experiment (Fig. 2). After initial CO_2 addition to the 60/40+CO_2 column, the pH decreased; however, further CO_2 addition was required (between day 43 and 51) to maintain a stable pH for the duration of the experiment (Fig. 2).

Water Clarity

Water clarity in the columns was monitored by Secchi depth, turbidity, and photosynthetically active radiation (PAR) light intensity measurements. Of the three columns tested in this study, only the 60/40+CO_2 column had a notable improvement in water clarity, according to each of these variables.

The 60/40+CO_2 column initially had a gradual increase in Secchi depth, and a sharp increase occurred (from 0.22 m to 1.92 m) between days 78 and 113 (Fig. 2). The 60/40-noCO_2 column had a slow and insignificant increase in Secchi depth (from 0.08 to 0.55 m, $P = 0.25$) over 133 days (Fig. 2). Likewise, while the pH decreased in the 20/80 column after CO_2 treatment compared to the 60/40+CO_2 column, only a slight increase in Secchi depth (0.055 to 0.3 m, $P = 0.25$) was observed over 133 days (Fig. 2).

The PAR trends were similar to those of Secchi depth. The 60/40+CO_2 column showed the same dramatic increase in PAR as the Secchi depth between days 78 and 98, and the final euphotic zone depth was >1.8 m on day 113 (Fig. 3). The 60/40-noCO_2 column showed very little improvement in light penetration, and the final euphotic zone depth at day 151 was >0.8 m, which was higher than the observed Secchi depth (0.55 m). Light penetration in the 20/80+CO_2 column gradually increased over time, with the first noticeable change occurring on day 30. The highest Secchi depth (0.52 m) and the highest euphotic zone depth (>1 m) was observed on day 58, even though Secchi depths and light intensity curves decreased after this (e.g. on day 161, Secchi depth was 0.38 m and euphotic depth >0.8 m). The discrepancy between the Secchi depth and PAR measurements was likely caused by the water transparency (Luhtala and Tolvanen 2013).

The trends in turbidity were similar to those of the Secchi depth and PAR light intensity. A slow initial reduction in turbidity (32.2%) was observed in the 60/40+CO_2 column within the first 57 days, and then a sharp decline in turbidity was observed after day 57 resulting in a total 96.8% reduction in turbidity. In contrast, the 60/40-noCO_2 column had an initial sharp decline in turbidity (65.8% reduction) in the first 27 days, and a final 81.6% reduction in turbidity was observed at day 133. However, only 77.9% turbidity reduction was observed in the 20/80+CO_2 column within 161 days.

Zhu et al. (2011) reported a 76.9% turbidity reduction in OSPW after treating with CO_2, compared to a 96.8% turbidity reduction observed in the 60/40+CO_2 column in this study. Several factors could contribute to the observed differences. First, fine FFT particles were resuspended from the sediment layer in the study by Zhu et al. (2011). These authors used an autoclave to supersaturate the solution with CO_2, and escaping CO_2 bubbles disrupted the sediment (Zhu et al. 2011). The current study utilized the SEAIR system to introduce CO_2, and the inflow/outflow was oriented to minimize disruption of the sediment. Second, Zhu et al. (2011) examined the turbidity only five hours after treatment, whereas this study tracked the turbidity for over 100 days, allowing other slow chemical processes such as mineral dissolution to occur, which could further contribute to improved water clarity.

Ions

Major cations and anions were analysed, and all of the ion concentrations presented here were normalized with chloride ion concentration to account for water evaporation during the experiment. An increase of divalent cations was observed in the 60/40+CO_2 column, and computer modelling suggested that carbonate mineral dissolution likely occurred in both the 60/40+CO_2 and 20/80+CO_2 columns.

An increase in sodium ion (Na^+) concentration was observed in both the 60/40+CO_2 and 20/80+CO_2 columns (Fig. 4). The diffusion of Na^+ from the underlying FFT layer into the water cap may have contributed to the increase in the Na^+ concentration. The initial Na^+ concentration in the porewater was much higher than in the cap water in both the 60/40+CO_2 and 20/80+CO_2 columns (Table 1 and 2). A slight increase in bicarbonate (HCO_3^-) concentration was observed in the 60/40+CO_2 and 20/80+CO_2 columns. Siddique et al. (2014) also reported an increase in HCO_3^- concentration in an anaerobic column study; they hypothesized that this increase was due to dissolution of CO_2 in the cap water and/or the dissolution of carbonate minerals.

In addition to sodium ion, a slight increase in the concentrations of divalent cations (magnesium and calcium) in the cap water of the 60/40+CO_2 column was observed, along with a slight decrease in potassium and sulphate ion concentrations (Fig. 4). A similar increase in magnesium and calcium ions was also reported in previous studies, as a result of a decrease in pH caused by dissolved CO_2, produced through biotic and abiotic processes (Siddique et al. 2014; Zhu et al. 2011). This decreased pH caused the dissolution of carbonate minerals, increasing the magnesium and calcium ion concentration (Dompierre et al. 2016; Siddique et al. 2014). In this study, the total concentration of carbonate minerals was also determined (Table 2), and XRD results suggest that calcite was the major carbonate mineral (Fig. S1). Calculations using PHREEQCi showed a -2.6 fold change in the saturation index for calcite (Table S2), suggesting that the CO_2 addition resulted in decreased saturation of calcite and that dissolution was likely to occur. Additionally, studies performed by Siddique et al. (2014) and Dompierre et al. (2016) suggested that the dissolution of carbonate-containing minerals may contribute to FFT dewatering.

The change in ionic concentrations in the 60/40+CO_2 column were different from those in the 20/80+CO_2 column. The 20/80+CO_2 column had an increase in magnesium and sulphate concentrations, a decrease in potassium, and no observable change in calcium concentrations (Fig. 4). Interestingly, the saturation indices calculations for the 20/80+CO_2 column showed a -3.2 fold change in the saturation index for calcite. The 20/80+CO_2 column had a lower buffering capacity than the 60/40+CO_2 column, likely resulting in a greater magnitude of the observed chemical changes (Table 3). Although the modelling suggested that carbonate-containing mineral dissolution should have been similar in the two columns, the differences in water clarity was likely due to the lower ionic strength in the 20/80+CO_2 column (approximately half of the 60/40+CO_2 column; Table 3). Ionic strength is one of the most important parameters in the coagulation/flocculation of suspended clay particles, as the thickness of the diffusion double layer is inversely related to ionic strength of the solution (Siddique et al. 2014; Hunter 2001).

The concentrations of most ions were stable during the experiment in the 60/40-noCO_2 column, except for sulphate which decreased in concentration (Fig. 4).

Piper diagrams constructed using data on ionic concentrations suggested that the water was mixed in all three columns (Fig. S2). In addition, the anion zone for the 60/40+CO_2 and 20/80+CO_2 shifted from the "no dominant type" to the “bicarbonate type”, and the overall water type changed from the “sodium chloride type” to a “mixed type” (Fig. S2a & c), indicating that carbonate minerals might have dissolved. By contrast, the 60/40-noCO_2 column retained "sodium chloride type" water over time (Fig. S2b).

Conclusion

This study suggests that it is possible to increase water clarity by adjusting the pH of OSPW. However, an additional question is raised concerning the lag time between pH adjustment and when the water began to clear. The 60% OSPW and 20% OSPW treatments were used to determine if CO_2 addition can improve water clarity, and if there was an associated lag time. The pH was reduced to pH 6 through the addition of CO_2, but neither column had an initial increase in water clarity. The marked improvement of Secchi disk depth was observed in the 60% OSPW treatment between 78 and 113 days. However, the 20% OSPW treatment did not increase in clarity after 133 days. This significant time lag is very important for any future studies on coagulation, flocculation and settling in pit lakes, and for industry efforts to manage turbidity in pit lakes. Sufficient time must be allowed to fully observe the effects of any method of coagulation, flocculation and settling. In addition, the improvement in water clarity in the 60% OSPW treatment but not the 20% OSPW treatment indicates that ionic strength may play an important role in coagulation at pH 6, likely due to the reduction of the diffuse double layer thickness (Siddique et al. 2014; Hunter 2001). OSPW is characterized by higher ionic strength than freshwater; hence, a higher ratio of OSPW in the pit lake in conjunction with CO_2 addition may be necessary for maximum water clarification. Additional studies should confirm the optimal ratio of OSPW to fresh water to optimize water clarification in pit lakes. Increasing the extent and rate of coagulation, flocculation and settling is essential to increase light penetration and improve the health of the littoral zone and pit lakes in general.

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Supplemental Data

Tables S1-S2 and Figs. S1-S2 are available online in the ASCE Library (https://ascelibrary.org)

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Table 1. Cation and anion concentrations in the porewater.

Porewater ion	**Concentration (mg/L)**
Cl^-	1296 ± 10
SO_4^{2-}	354 ± 4
Na^+	1118 ± 135
Mg^{2+}	24 ± 2
K^+	10 ± 2
Ca^{2+}	15 ± 1

Table 2. Physical parameters of FFT.

Parameters	**Value**
MBI (meq/100g)	3 ± 2
Solids Content (% by mass)	37 ± 7
Zeta potential (mV)	-28 ± 2
Carbonate (wt %)*	4.5

*=Calculated on oven dry weight basis.

Table 3. Initial parameters of 60/40+CO_2, 60/40-noCO_2, and 20/80+CO_2 columns

Parameter	**60/40+CO2**	**60/40-noCO2**	**20/80+CO2**
pH	**8.26 ± 0.07**	**7.85 ± 0.06**	**8.03 ± 0.04**
Conductivity (mS/cm)	**2.12 ± 0.03**	**2.29 ± 0.06**	**1.05 ± 0.04**
Turbidity (NTU)	**299 ± 9**	**301 ± 17**	**208 ± 12**
Alkalinity (mg $CaCO_3$/L)	**561 ± 7**	**468 ± 10**	**267 ± 4**
TSS (mg/L)	**384 ± 145**	**-**	**422 ± 43**
TDS (mg/L)	**1818 ± 101**	**1651***	**985 ± 106**
Cl^- (mg/L)	**396 ± 16**	**337 ± 3**	**143 ± 1**
SO_4^{2-} (mg/L)	**203 ± 1**	**185 ± 2**	**51 ± 1**
Na^+ (mg/L)	**574 ± 10**	**523 ± 38**	**210 ± 38**
Mg^{2+} (mg/L)	**14 ± 0.3**	**11 ± 1**	**10 ± 2**
K^+ (mg/L)	**14 ± 3**	**10 ± 1**	**15 ± 3**
Ca^{2+} (mg/L)	**29 ± 1**	**24 ± 3**	**34 ± 7**
Ionic Strength (mmol/L)	**30.6**	**26.8**	**13**
Total Organic Carbon (mg/L)	**-**	**37.6 ± 2.3**	**35 ± 1.3**
Dissolved Organic Carbon (mg/L)	**42 ± 3.0**	**38.5 ± 1.6**	**29 ± 1.6**
Naphthenic Acids (mg/L)	**33 ± 3.5**	**32 ± 2.0**	**12 ± 1.5**

- = not tested, * = estimation based on conductivity and ions concentrations calculated from http://www.lenntech.com/calculators/tds/tds-ec_engels.htm