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Effect of cement kiln dust, lime and fly ash on metal leaching characteristics of oil sands tailings from Alberta, Canada

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Abstract - *The oil sands industry of Alberta generates huge amounts of tailings in a slurry form that typically require up to 40 years to consolidate in very large tailings ponds which are up to 150 m in height. Cement kiln dust (CKD), a byproduct of the ordinary Portland cement manufacturing process, as well as lime and fly ash, collectively referred to as geopolymers, may have the potential to reduce the tailings slurry consolidation period from 40 years, thus affecting the sustainability of such tailings facilities. However, first, it must be demonstrated that these geopolymers will also decrease the metal leaching from thickened tailings (TT) and mature fine tailings (MFT) from the oil sands industry. This study was focused on the use of geopolymers to reduce the environmental impact of TT- and MFT-tailings in the Alberta oil sands industry. Toxicity characteristics leaching procedure (TCLP) and static leaching test (SLT) was used to examine the leaching of metals from tailings, with the SLT test effectively mimicking the leaching process in the tailings ponds environment. Under non-acidic conditions corresponding to the SLT test results, iron concentrations with values of about 530-705 ppm were found to be lower than previous studies on oil sand tailing ponds (2400 ppm). Results showed that geopolymer amendment of TT and MFT significantly reduced the leaching of heavy metals. SLT tests showed that amendment of MFT with 7% CKD decreased Pb, Mn, and Fe leaching, whereas TT-amended with 4% CKD decreased Cu, Pb, Zn, Mn, and Fe. Overall, the CKD amendment of TT showed more than 95% efficiency in the reduction of leaching of all heavy metals. In TCLP tests, TT-amended with 2% FA decreased the leaching of Pb and Ni to acceptable levels with substantial efficiency in reducing the leaching of Fe, Cu, and Zn. TCLP tests also showed that among different amendments, TT-amended with 4% CKD or 2% FA were the most effective proportions for controlling metal leaching from TT, while MFT-amended with 7% lime/FA or 3% CKD were the effective proportions for reducing metal leaching from MFT. Thus, it is deduced that CKD at 3%-4% w/w amendment would work best for reducing leachate levels of both TT and MFT. While amendment of tailings by means of a combination of fly ash and lime also were effective in reducing the leaching of metals, these two geopolymers were not as effective as the CKD amendment.*

***Keywords** – Oil sands industry, thickened tailings, mature fine tailings, geopolymers, CKD, heavy metals, leaching tests.*

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1. Introduction

Significant amounts of tailings are generated by the oil sands industry in Alberta. By the end of 2013, the Alberta oil sands tailings ponds and associated structures covered an area of about 220 km^2, with 88 km^2 being the liquid surface area of the ponds[1]. By September 2017, all of the Alberta tailings ponds held approximately 1.2 trillion L of contaminated water.

The Alberta oil sands tailings contain a highly variable composition of different compounds including water, fine silts, residual bitumen, salts and soluble organic compounds, heavy metals, and solvents [2]. This huge amount of oil sands tailings has led to concerns about negative impacts on soil, surface and groundwater, air quality, as well as adverse effects on ecology and wildlife. Heavy metals are one of the toxic contaminant groups in the oil sands tailing ponds water that originate from tailings [2].

Various technologies have been studied and employed to manage the Alberta oil sands tailings [1]. Among them only a few technologies such as sedimentation/self-weight consolidation and composite/consolidated tailings (CT) have demonstrated commercial applications [1]. Stabilization and coagulation by alkaline materials have also been used by researchers [3].

Ordinary Portland cement (OPC) has been used to stabilize oil sands mine tailings. However, there are some limitations to the OPC stabilization of tailings, such as poor metal immobilization, high energy consumption, as well as carbon footprint and greenhouse gas (GHG) emissions associated with the production process of OPC [4].

Geopolymers, which are stable materials produced by the reaction by aluminosilicates in a highly concentrated alkali hydroxide or silicate solution, can also be used to stabilize oil sands tailings [4]. They are amorphous polymeric-structured materials having amorphous polymeric structures with interconnected Si–O–Al bonds [5], [6]. Compared to OPC, geopolymers show some advantages such as rapid development of mechanical strength, acid resistance, immobilization of toxic materials, and reduced GHG emissions. These characteristics have made geopolymers a great potential sustainable alternative to OPC [5]. However, to date, there are not many research results published on the geopolymerization of oil sands mine tailings [4], [7], [8].

Cement kiln dust (CKD) is a powdery by-product collected from kiln exhaust flow in the OPC manufacturing process. Ground and mixed calcium carbonate and clay are calcined by heating to very high temperatures to produce calcium carbonate. During calcination in the kiln, in addition to the main product, a dust named CKD is also generated. It is separated and collected from the exhaust gas as part of air quality control measures of OPC manufacturing plants. CKD can be collected and landfilled, fed back to the process for further calcination, or used outside of the production plant for a variety of purposes such as mine tailings stabilization, chemical admixes, subgrade improvement [9], as a stabilizing binder in oil and gas industry [10], and in agricultural applications. For example, CKD can be used to reduce the total heavy metal concentrations of sludge in the stabilization and solidification of the waste from water treatment plants. It can also be used for soil stabilization as an alternative to lime [9].

The chemical and physical characteristics of CKD are dependent in great part on the manufacturing process adopted, method of dust collection employed at the facility, and the raw materials [11]. However, CKD basically contains very fine particles of cement clinker, unreacted or partially reacted particles of clay or calcium carbonate and raw materials and fly ash of fuel that has been used in rotary cement kiln [4], [12], [13]. Oxides such as Al2O3, CaO, SiO2, and Fe2O3 are present in CKD in such proportions that can produce intrinsic cementitious properties [14], [15]. pH of CKD is naturally alkaline due to the presence of Ca, Na and K.

Nehdi and Tariq (2007) reported strength gain, decrease of hydraulic conductivity and immobilization of heavy metals due to hydration of CKD used to stabilize sulfidic mine tailings [10]. Ahmari and Zhang (2013) studied the enhancement of the physical and mechanical properties and the durability of mine tailings-based geopolymer bricks by adding CKD [4]. They investigated the effects of CKD content on the unconfined compressive strength (UCS), water absorption, and durability of mine tailings-based geopolymer bricks at different conditions, reporting significant improvement of the physical and mechanical properties as a result of CKD addition. Latifi et al. (2014) reported that CKD can be employed in soil stabilization as stabilizer with the same mechanisms of traditional chemical stabilizers such as lime and OPC.

With rising concerns about GHG and carbon emission from industries such as cement manufacturing, partial or total replacement of OPC in applications such as tailings stabilization can not only reduce the carbon footprint of both tailings management and cement industry but also reduce the cost and carbon footprint of CKD from OPC plants.

While several studies have been conducted on the physical and geotechnical properties of oil sands tailings [16]–[19] and treatment of oil sands tailings [18]–[23], published research on the effect of CKD amendment of oil sands tailings and their leaching properties is rare.

The present study investigates the effect of CKD amendment of thickened tailings (TT) and matured fine tailings (MFT) with respect to leaching of metals. Other geopolymer amendment mixtures, containing fly ash, lime and CKD, were also investigated. Toxicity characteristic leaching procedure (TCLP) and static leaching tests (SLT) were utilized for examining the leaching of metals from the various geopolymer-amended mixtures.

2. Materials and methods

2.1 Materials and geopolymer-amended experiments

Tailings materials

Thickened tailings (TT) are tailings that are composed of silt and clay sized particles and have been significantly dewatered in a thickener with chemical additives to a point where they form a homogeneous non-segregated mass when discharged from the end of a pipe. A dense mixture of clay, silt and water on the bottom of the disposal sites of tailings from which the upper clarified layer of water is extracted to re-use in extraction process is referred to as matured fine tailings (MFT). Both types of the tailings (TT and MFT) samples contained water. However, no additional water was used in the preparation of the TT and MFT samples. The TT and MFT samples were provided by Canmet Canada, acting as agent and intermediary with an oil sands leaseholder.

Geopolymers

Cement kiln dust (CKD) was supplied by Lafarge's Exshaw plant, west of Calgary, Alberta, Canada. Fly ash and lime were also used as geopolymer amendment in this study. The mineralogical properties of the CKD are included in Table 1, together with those of lime and fly ash. The mineralogical properties of these geopolymers were determined by X-ray fluorescence (XRF).

The major component that makes up CKD is calcium oxide. Silicon oxide is another major component of CKD. Major components of fly ash are oxides of silicon, aluminum, and iron. Oxides of magnesium, potassium, calcium, sodium, and sulphur are also present to a lesser degree as minor phases. Higher pH values of CKD, lime, and fly ash make them good amendment materials to neutralize acidic waste water as well as precipitate/stabilize heavy metals in slurries and acidic wastewater.

Table 1. Mineralogical Properties, pH, moisture content, and loss on ignition of CKD, Lime, and Fly Ash

Constituent	CKD	Lime	Fly Ash
Silicon Dioxide (SiO_2) %	15.2	1.09	43.5
Aluminum Oxide (Al_2O_3) %	3.96	0.33	25.2
Iron Oxide (Fe_2O_3) %	2.21	0.09	11.5
Calcium Oxide (CaO) %	47.0	97.7	1.85
Magnesium Oxide (MgO) %	2.80	1.12	1.04
Sodium Oxide (Na_2O) %	0.35	<0.01	0.43
Potassium Oxide (K_2O) %	2.08	0.08	2.26
Sulfur Trioxide (SO_3) %	6.42	0.06	0.21
pH	13.0	12.1	8.4
Moisture Content %	0.15	0.13	24.8
Loss on Ignition (LOI) %	19.2	2.14	10.8

Geopolymer-amendment of tailings materials

Geopolymer-amended experiments were conducted with combinations of raw tailings materials and various geopolymers. Table 2 presents the proportions of materials tested in this study. Various samples were prepared by mixing the tailings in batches for approximately 2-3 minutes to homogenize. 300 g of pre-mixed tailings was then weighed out into a 500 mL plastic container and mixed with specific amounts of CKD, lime, or fly ash to achieve targeted proportions. The mixing rate was increased by 100 rpm increments every 30 seconds for a total of two minutes. In order to ensure proper mixing, the mixing container was moved up and down to ensure that the total volume of slurry was thoroughly mixed. Neither alkali hydroxide nor silicate solution was used in the geopolymer-amendment experiments. Leaching tests were performed on raw and geopolymer-amended tailings samples after one hour and one day. 18-day and 28-day leaching tests were also performed on CKD-amended TT samples.

2.2 Tests and analyses

The Toxicity Characteristic Leaching Procedure (TCLP) was conducted according to US EPA Method 1311 [24] using extraction fluid at pH 4.93. The extraction fluid was prepared by adding 5.7 mL of glacial acetic acid to 500 mL of reagent water. The leachate was then analyzed for dissolved metals right away.

The Static Leaching Test (SLT) was conducted using similar procedures to the TCLP tests except that water was added instead of the pH 4.93 extraction fluid. Analytical samples were prepared by adding 100 g of sample to 2.00 L (20:1 ratio) of reagent water, followed by mixing for a period of 72 hours. The leachate was then analyzed for dissolved metals. Sample preparation and the leaching tests were carried out at SGS's laboratory in Vancouver, BC, and later at the Alberta Innovative Future Technology Laboratory in Vegreville, Alberta.

Leachate samples were analyzed for dissolved metals by Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) method. Mercury was analyzed using the Cold Vapour Atomic Absorption (CVAA) method. Results from the leaching tests were evaluated against a) the Alberta Surface Water Quality (ASWQ) standards, and b) the Canadian Water Quality Guideline for Aquatic Life [25] standards.

Table 2. Materials and mixtures tested for metal leaching

Tailings Materials	Geopolymer-Amended Mixtures
TT	TT + 1% CKD
	TT + 4% CKD
	TT + 1% FA
	TT + 2% FA
	TT + 3% Lime/FA
MFT	MFT + 3% CKD
	MFT + 7% CKD
	MFT + 2% FA
	MFT + 3% Lime/FA
	MFT + 7% Lime/FA

2.3 Quality control

All batch-mixing experiments were conducted in duplicate for quality control purposes. Average results are reported here. PACS-2, TILL-1, and TILL-3 certified reference material from the National Research Council (NRC) of Canada were used as the standard samples for quality control of chemical composition and metals analyses. Measured concentrations were found to be within the concentration ranges provided by NRC. For all TCLP and SLT leaching tests, blank samples were run and analyzed for quality control purposes.

3. Results and discussion

3.1 Heavy metals leaching from raw tailings

As the basis for comparison and evaluation of the (beneficial) effects of the amendments, untreated tailings (TT and MFT) were tested for metals leaching. Metal leaching results obtained from TCLP and SLT tests along with the regulatory and reference standards are presented in Table 3.

Table 3. Leaching of metals from untreated TT and MFT (ug/L)

Dissolved Metal	TT[a]		MFT[b]		ASWQ[c]	CWQG[d]
	TCLP	**SLT**	**TCLP**	**SLT**		
Antimony (Sb)	0.48	0.516	0.448	0.525	NS[f]	NS
Arsenic (As)	0.892	7.44	1.17	0.946	5	50
Barium (Ba)	1225	96.9	2190	191	NS	NS
Beryllium (Be)	0.854	0.16	1.56	0.19	NS	NS
Boron (B)	589	879	1020	1320	NS	NS
Cadmium (Cd)	0.46	0.022	0.657	0.034	NS	0.8
Chromium (Cr)	2.72	4.66	4.11	0.735	1-8.9	2-20
Cobalt (Co)	79.2	1.44	34.6	3.53	NS	NS
Copper (Cu)	**20.3**	**11.2**	**12.8**	**2.26**	4	2
Iron (Fe)	**2443**	**704**	**2670**	**531**	300	300
Lead (Pb)	**7.9**	**2.59**	**7.75**	1.91	7	2
Manganese (Mn)	845	10.3	1770	87	NS	NS
Mercury (Hg)	0.0045	0.0047	<0.01	<0.01	0.1	0.1
Molybdenum (Mo)	2.65	3.46	2.43	6.42	73	NS
Nickel (Ni)	**105**	2.58	**63.6**	4.87	25-150	65
Selenium (Se)	0.375	0.643	0.324	0.475	NS	1
Silver (Ag)	0.0145	0.0396	0.0121	0.0332	0.1	0.1
Uranium (U)	0.985	0.528	0.776	0.502	NS	NS
Vanadium (V)	0.843	10.8	2.08	2.06	NS	NS
Zinc (Zn)	**156**	14.3	**293**	6.56	30	30

[a] Thickened tailings
[b] Matured fine tailings
[c] Alberta surface water quality standards
[d] Canadian water quality guideline for aquatic life, 2008
[f] NS Not specified

TCLP test results

Results revealed that the concentrations of barium, chromium, copper, iron, lead, manganese, nickel and zinc of both raw TT and raw MFT exceed either the ASWQ or CWQG standards. TT's leachate contains more chromium than ASWQ. Copper concentration of both TT and MFT is higher than ASWQ and CWQG. Pb concentration of TT and MFT also exceeds ASWQ and CWQG as well. Ni and Zn concentrations are also both higher than ASWQ and CWQG. However, it should be noted that the conditions which prevail under TCLP test are the worst possible acidic conditions, which are very unlikely to occur in the Alberta oil sands tailings ponds.

SLT test results

Table 3 shows the concentrations of chromium, copper, iron, and lead in the leachates from SLT tests on TT and MFT exceed standard levels of ASWQ and CWQG. However, it is noted that it is only the iron concentrations that show significantly higher values that the "acceptable" values (of 300 ppm) given in the standards.

Summary

Thus, the risk of contamination of TT and MFT tailings ponds and adjacent surface through metal leachates under "normal" conditions (ie those mimicked by the SLT tests) might be higher, given that the pH of tailings pond water varies in the range of 8.2-8.4 [2] while the pH at which SLT tests were conducted was about 9. The lower pH, as demonstrated in the section above through highly acidic conditions mimicked by the TCLP tests, can consequently lead to more metal leaching from tailings ponds into the surface and ground water surrounding the tailings ponds.

On the other hand, there is a potential greater risk of heavy metals release in case of any sudden or gradual significant decrease in the pH of tailings ponds water, as indicated by the TCLP test results where higher concentrations of metal leachates were measured than the standards.

The (TCLP test) results indicate that there is a risk of metals leaching from TT and MFT into the tailings pond water, leading to probable contamination of surrounding surface and ground water due to heavy metals such as Cu, Zn, Ni, Fe and Cr in the event that highly acidic conditions might develop in tailings ponds (ie. pH values of 4.93 which corresponds to the TCLP test protocols). In support of these findings, some previous studies on the water quality of tailings pond water confirmed Cu and Fe concentrations of 70 and 2400 ug/L, respectively, which exceed the ASWQ and CWQG values [2], [26]. Furthermore, Allen (2008) has also reported the abundance of Pb, Ni and Zn in oil sands tailings pond water at concentration levels higher than the Canadian environmental quality guideline [27] and US EPA water quality criteria for protection of aquatic life [28]. These previous studies corroborate the findings of the current study, namely that heavy metals would leach under highly acidic conditions, thus presenting a potential contamination problem in tailings management. However, under static conditions, corresponding to non-acidic conditions, which are derived from the SLT test results, only iron concentrations were found to be lower than the previously reported concentrations in oil sand tailing ponds, (2400 ppm in the MacKinnon 1993 study), but exceeded the 3 ppm concentration reported by the Allen 2008 for oil sands pond water, as compared with about 530-705 ppm in the current study.

3.2. Heavy metal leaching from geopolymer-amended tailings

TCLP tests results

To reduce the metal leachability of tailings, CKD, lime, and fly ash were used in different mixing proportions as presented in Table 2. Figure 1 depicts the results of metal leaching from TT and MFT amendments at different proportions.

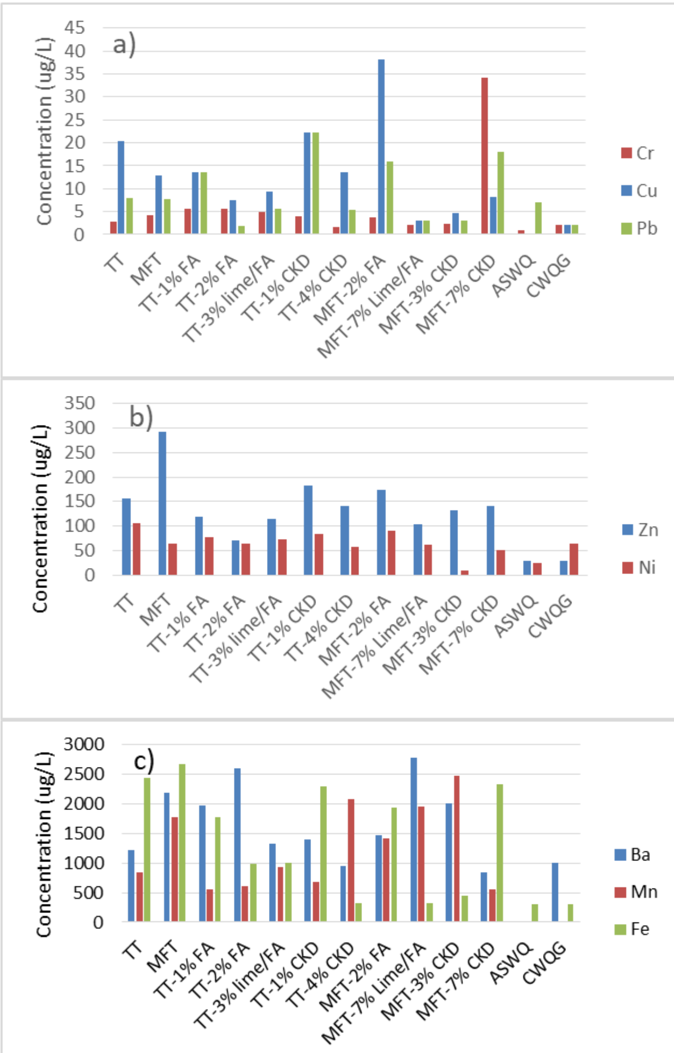

Figure 1. TCLP test results for metal concentrations in leachate from TT, MFT, and different geopolymer-amended tailings

The results from the worst-case scenario of leaching conditions (namely the TCLP tests) show that almost none of the amendments used decreased the leaching of all target metals to levels lower than the selected water quality standards (ASWQ and CWQG). However, some amendments were effective in reducing some metals to acceptable levels. For example, TT-amendment with 4% CKD decreased the level of Cr, Ni and Ba to concentrations lower than the standards, as illustrated in Fig. 1 a, b, and c respectively. However, while leaching of some other metals such as Cu and Fe decreased to 33.5% and 87%, respectively, their final leaching concentrations are still greater than acceptable levels given in the standards.

TT-amended with 2% FA decreased the leaching of Pb and Ni to acceptable levels, while achieving substantial efficiency in reducing the leaching of Fe, Cu and Zn to about 60%, 63% and 55%, respectively. However, the leaching of Ba and Cr did not decrease for TT with 2% FA amendment.

TT-amended with 1% CKD, 1% FA and 3% lime/FA did not decrease the leaching concentrations of any of the targeted metals to levels lower than the standards.

Figs. 1 a and b show that among different MFT amendments, the one with 7% lime/FA showed the best results in decreasing the leaching concentrations of Cr, Cu, Pb, Zn, and Fe under the worst-case scenario (TCLP tests). This amendment reduced the leaching of Pb to an acceptable level relative to the ASWQ standard.

MFT-amended with 7% lime/FA also decreased Ni and Cr leaching concentrations to acceptable levels with regard to the CWQG standard, while the concentration of Fe was close to the CWQG standard. However, the concentrations of Mn and Ba increased from the original concentrations in the raw MFT (Fig. 1 c).

MFT-amended with 3% CKD also showed great leaching decrease of Cr, Cu, Pb, Ni, Zn, and Fe. Leaching concentrations of Ni and Pb decreased to acceptable levels compared to the standards. Increasing CKD proportions from 3% to 7% in the MFT-amended mixtures decreased the Ba and Mn leaching concentrations, while increasing the leaching concentrations of other metals (Fig. 1 a, b, and c).

Overall, results of TCLP tests showed that among different amendments, TT-amended with 4% CKD or 2% FA were the most effective proportions for controlling metal leaching from TT, while MFT-amended with 7% lime/FA or 3% CKD were the effective proportions for reducing metal leaching from MFT. Thus, it is deduced that CKD at 3%-4% w/w amendment would work best for reducing leachate levels of both TT and MFT.

It is hypothesized that mechanisms that lead to CKD-amendment of TT and MFT reducing leaching of heavy metals are attributed to chemical precipitation and adsorption. Dissolved heavy metals in suspension could be precipitated as hydroxides and carbonates. Some previous studies have proven the effectiveness of CKD in heavy metals removal from solutions [29]–[31]. Zaki et al. (2007) reported that basic metal carbonates and hydroxides such as $Pb_3(CO3)_2(OH)_2$, $Cu_4(CaSO4)_2(OH)_6 \cdot 3H_2O$, Cd(OH)Cl, and hydrated $Ni(OH)_2$ and $Co(OH)_2$ are formed in the amended mixture as a consequence of CKD addition. Regarding the chemical composition of lime, which comprises of mostly CaO (Table 2), the major mechanism of metal stabilization by lime in the amendments is attributed to chemical precipitation of metal hydroxides.

In addition to chemical precipitation due to geopolymer amendment, increasing the pH in suspensions containing tailings and CKD or FA can also result in enhancing the adsorption of metals and partially hydrolyzed metals [32], [33].

SLT test results

Fig. 2 depicts the results of targeted metals concentrations in the leachates obtained from SLT tests of different geopolymer-amended tailings mixtures.

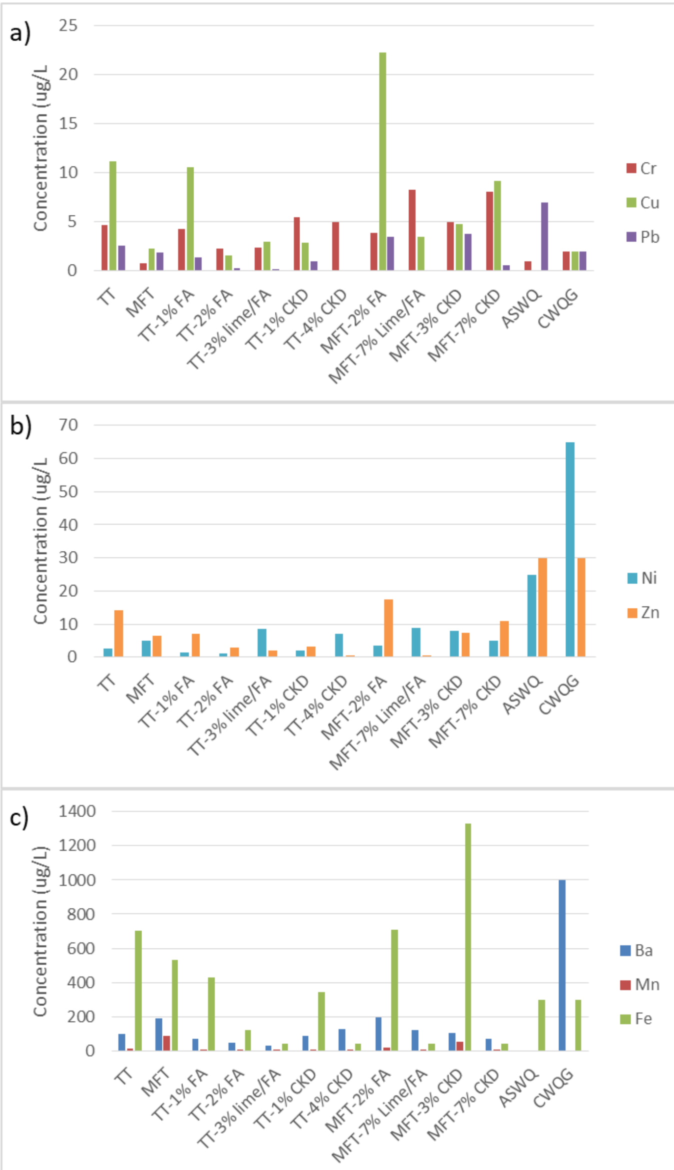

Figure 2. SLT test results for metal concentrations in leachates from TT, MFT and different geopolymer-amended tailings

Results show that for raw TT, Cr, Cu, Pb, and Fe leach at levels exceeding the CWQG standards. For MFT, Cu and Fe are the only metals that leach at concentrations that are greater than the acceptable levels of the ASWQ and CWQG standards.

The most effective amendment for TT for reducing Cr and Ni concentrations is with 2% FA (Figs. 2 a and b). TT amendment with 4% CKD decreased concentrations of all metals, except for Cr, to levels lower than the standards. The 4% CKD amendment of TT also reduced concentrations of Fe, Mn, Zn, Pb and Cu to the lowest values, compared to other TT amendments (Figs. 2 a, b and c). Leachate concentration reduction efficiencies of TT-amended with 4% CKD for Cu, Pb, Zn, Mn and Fe were about 100%, 99%, 96%, 99% and 94%, respectively. However, this amendment led to slight increases in Ba and Cr concentrations.

For MFT, amendment with 7% lime/FA showed the most promising results in reducing the concentrations of Cu, Pb, Ni, Zn, Mn and Fe. Final concentrations of all metals studied, except for Cr and Cu, were lower than the standards (Figs. 2 a, b and c). MFT amended with 7% CKD also showed good efficiencies for Pb, Mn and Fe at about 77%, 98% and 94%, respectively. MFT-amended with 3% CKD showed increase in some of the heavy metals concentrations (i.e. Ba, Cr, Pb, Ni, Mn and Fe).

Overall, the results showed that amendment with 4% CKD and, to a lower extent, 2% FA, were effective in decreasing the leaching of metals from TT to levels lower than or near the acceptable standards. Similar results were achieved for MFT through amendment with 7% lime/FA or 7% CKD.

4. Conclusion

This study investigated the effect of different proportions of CKD, lime and FA in controlling leaching of heavy metals from thickened tailings (TT) and mature fine tailings (MFT) a) into water and b) under highly acidic conditions. Results revealed that geopolymer amendments significantly decreased metals leaching into both tailings pond water and under highly acidic conditions. Although, none of the amendments applied decreased the concentrations of all metals to surface water quality standards, the leaching of some harmful metals (Cu, Pb, Zn, Mn and Fe) decreased to more than 90% efficiency. Thus, geopolymer-amendment of TT and MFT is shown to be effective in improving water quality of tailings pond water to levels suitable for recycling into process water system treatment feed in oil sands tailings industry. Given that CKD and lime/FA amendments showed promising results in decreasing a number of metals including Ba, Cr, Ni, Cu and Zn in some cases, further investigations on a wider range of amendment proportions would be prudent. As well, X-ray diffraction, X-ray fluorescence, scanning electron microscope/energy dispersion using X-ray (SEM/EDX) can also reveal the (physical and chemical) mechanisms that are affecting the stabilization of heavy metals in TT and MFT amended by the geopolymers studied.

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