

COMPARISON OF AMD TREATMENT PROCESSES AND THEIR IMPACT ON SLUDGE CHARACTERISTICS

BERNARD C. AUBÉ

Noranda Inc. Technology Centre, 240 Hymus Blvd
Pointe-Claire, Québec H9R 1G5, Canada

and

JANICE M. ZINCK

CANMET, Mining and Mineral Sciences Laboratories
555 Booth Street, Ottawa, Ontario K1A 0G1, Canada
jzinck@nrcan.gc.ca

Abstract

Lime neutralisation for the treatment of acid mine drainage is one of the oldest water pollution control techniques practised by the mineral industry. Several advances have been made in the process in the last thirty years, particularly with respect to discharge concentrations and sludge density. However, the impact of different treatment processes on metal leachability and sludge handling properties has not been investigated.

A study of treatment sludges sampled from various water treatment plants has shown that substantial differences can be related to the treatment process and raw water composition. This study suggests that sludge densities, excess alkalinity, long-term compaction properties, metal leachability, crystallinity and cost efficiency can be affected by the neutralisation process and specific process parameters. The study also showed that the sludge density and dewatering ability is not positively correlated with particle size as previously suggested in numerous studies. The treatment process comparisons include sludge samples from basic lime treatment, the conventional High Density Sludge (HDS) Process, and the Geco HDS Process.

Key Words: acid mine drainage, treatment, sludge, density, viscosity, stability, lime efficiency, leachability.

Description of Processes

All processes compared within the scope of this investigation use either quicklime or hydrated lime for the precipitation of heavy metals. In all cases, the acid mine drainage (AMD) was provided from the oxidation of sulphides in mine wastes and/or mine workings from base metal mines. The reviewed sites apply variations of three basic processes as described in Figures 1 to 3 and the following paragraphs.

Basic Lime Treatment

The basic lime treatment is simple addition of lime to an AMD stream followed by solid/liquid separation in a settling pond (Fig. 1). The lime is added to attain a pH suitable for precipitation of the heavy metals to be removed from solution. Falconbridge and Kidd Metallurgical Division apply this treatment process. A higher pH setpoint is often necessary to insure complete precipitation of metals throughout the pond. When the pond overflow pH exceeds a regulated maximum, carbon dioxide can be used to depress pH without adding toxicity to the treated water.

HDS Process

Figure 2 describes the Conventional High Density Sludge (HDS) process. The AMD is generally fed into a Rapid Mix Tank (RMT), where it is contacted with a lime/sludge slurry to bring the pH of the combined slurry to 9 or 9.5. The RMT (shown in dashed lines) is often used to offer better pH control in the process, but is not necessary. The retention time in this vessel varies normally from 2 to 10 minutes.

is used for raw water storage thus the raw water chemistry remains seasonally stable with changes recorded mostly on a yearly basis.

Table 2. Approximate Raw Water Characteristics (mg/L).

Site	Al	Cd	Cu	Fe	Mn	Ni	Pb	Zn	SO ₄	pH	Eh (mV)
Brunswick	20	0.10	3	145	20	0.1	2	120	2300	3.2	540
Falconbridge Sud.			1.1	8		3			500	3.1	
Geco	10	0.05	0.5	600	10	<0.5	<0.5	20	4500	3.9	500
Heath Steele	25	0.15	10	180	38	0.1	2	160	3000	3.0	660
Kidd Metallurgical			3	25	<0.01	0.05	0.1	60	2100	4.5	
Mattabi	34	0.50	14	145	33	0.6	<0.5	245	4000	3.0	680
Waite Amulet	15	<0.02	1	75	4	<0.5	<0.5	5	1000	2.5	725

When all factors are compared, the raw water has the greatest influence on the sludge characteristics. The sludge is essentially composed of the metals treated. For this reason, sites such as Geco and Waite Amulet which contain primarily Fe, will form a more stable sludge. Mattabi and Kidd are among the few sites that generally treat more Zn than Fe, therefore forming a sludge slightly more sensitive to pH. Brunswick and Heath Steele have only slightly more Fe to treat than they do Zn. Cd, Cu, Pb, and Ni are often minor elements treated easily when there is an excess of Fe and Zn. At Falconbridge Sudbury Division, the raw water metal concentrations are low and therefore require excess lime for treatment in order to decant the few solids formed in neutralisation.

Sludge Characterisation

As shown in Table 3, the sludge densities, pH and Eh vary significantly. The data in this table was gathered primarily by CANMET for comparing the sludges from various Noranda and Falconbridge sites. Some particle sizing was also done at Noranda, but since these were from different samples, the solid contents of the sludge differ. As the Brunswick Mining Division was co-depositing the sludge with tailings at the time of sampling, no aged sludge could be collected for the operating treatment process. The Heath Steele WTP had not been in operation long enough to have produced aged sludge from the existing process. The aged sludge from all other sites was at least a year old.

Sludge Densities

The solid content of a fresh sludge depends primarily on the raw water composition and applied process. Brunswick, for example, had the highest density of fresh sludge. The Brunswick WTP is highly automated, has an over-sized clarifier, and sufficiently high concentrations of iron and zinc to produce a high-quality, high-density sludge. Geco's raw water contains very high concentrations of iron and is also automated sufficiently for efficient treatment control. The smaller clarifier at Geco does not offer as much retention time to allow for compaction of the sludge prior to disposal.

On the other end of the scale, the two sites without any recirculation result in fresh sludges with less than 5% solids. Both the Falconbridge Sudbury Division and the Kidd Metallurgical Division produce relatively voluminous sludges. This can be explained by the physical attributes on a micro-scale. Scanning electron microscope (SEM) pictures have shown that sludges particles from Brunswick or Geco produce small spherical particles that are apparently solid. The particles from lower density processes seem typically larger but particularly more porous or "fluffy". A visual analogy could show HDS resembling ball-bearings, while LDS can be compared to cotton balls.

Lime sludges tend to densify with ageing due to evaporation and freeze-thaw effects. In general, sludges deposited without a water cover display a greater degree of densification because of surface evaporation. However, the degree of densification observed with ageing can vary significantly from site to site. For example, Falconbridge sludge aged (in a sludge pond with a water cover) for 17 years contained only 7% solids while Geco sludge aged only one year (on a dry tailings beach) contained 60% solids.

These examples show the importance of the process on the sludge density. A low density sludge will never attain the solid content of a high density sludge even with ageing and freeze-thaw. The most significant control is during formation of the sludge. The change in density may be affected by the disposal scenario, but the effect of leaving the same sludge either under water or disposing it dry has not been investigated. Only clues are available showing the relative difference, and these do not seem consistent.

Table 3. Physico-Chemical Characteristics of Sludges.

Site	Sample	Density (% solids)	Particle size CANMET (μm , D_{50})	Particle size NTC (μm , D_{50}) [% solids]	pH	Eh (mV)
Brunswick	Fresh	32.8	4.09	3.1 [24%]	10.04	166
Falconbridge Sudbury	Fresh	3.7	5.74		9.45	161
	Aged	7.2	7.96		9.51	315
Geco	Fresh	27.8	2.89	2.7 [16%]	9.32	222
	Aged	60.0	3.88		9.32	221
Heath Steele	Fresh	20.8	4.13	4.1 [16%]	9.48	270
Kidd	Fresh	3.4	6.67		10.85	239
	Aged	4.1	21.06		10.56	201
Mattabi	Fresh	16.1	5.27	4.7 [16%]	9.30	301
	Aged	22.5	5.92		9.95	213
Waite Amulet	Fresh	18.0	3.96	4.7 [6%]	8.90	262
	Aged	24.8	5.27		9.62	300

Both the Waite Amulet sludge and the Geco sludge were deposited on surface in draining conditions. Geco sludge more than doubled in solid content while the Waite Amulet sludge increased by less than 1.4 times. Mattabi sludge, maintained under water, increased exactly 1.4 times in solid content. The samples of LDS densified by 1.2 to 1.9 times the original formed density. These data suggest that even the long-term densification is due primarily to the type of solids formed in the process (i.e. particle size, shape and density).

Particle Size

The sludge particles observed in these samples were generally aggregated masses displaying either bimodal or multi-modal distributions. Some of the plant sludges were sampled both as a part of CANMET work and within Noranda, to be analysed at NTC (Noranda Inc. Technology Centre). In both cases, HDS-type treatment sludges display narrower size distributions, indicative of a greater homogeneity. High density sludges also tend to have lower median particles sizes than other treatment sludges. The fresh high-density sludge from Geco possessed the lowest median particle size (2.89 μm). The highest median particle size was measured on a sample from Kidd Metallurgical, from Basic Lime Treatment.

Sludge densities of fresh material from numerous treatment sites are plotted against mean particle size distribution as presented in Figure 4. The particle sizes for the HD sludges

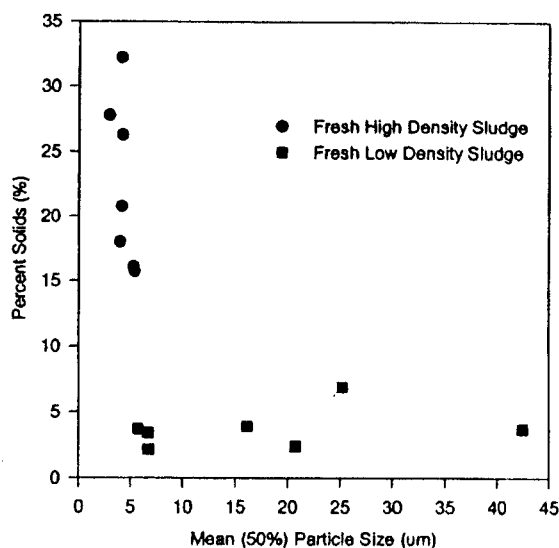


Figure 4: Relationship between Solid Content and Particle Size

cluster around 3-5 μm regardless of the sludge density. Low-density sludges report higher median particle size distributions either because of a higher degree of porous particle aggregation or the presence of larger calcite particles (Zinck et al. 1997a). In HDS systems, there is better control of the precipitation mechanism. This was particularly apparent for the Geco sludge. In this case, the particles were small and uniformly shaped, suggesting chemical growth rather than simple particle aggregation. In all cases, median particle size increases with ageing. This may be due to localised particle dissolution and re-crystallisation, leading to particle growth and aggregation.

Sludge pH and Eh

In general, the sludge pH is essentially the same as the treatment control pH. In cases where the sludge pH is higher, this suggests an incomplete reaction of lime (Zinck and Aubé, 1999). The redox values are mostly inversely proportional to pH, as the system follows the slope of the water stability field. There are exceptions when this balance is affected by some redox reactions. Some reactions that may occur include the on-going oxidation of ferrous or biodegradation of polymers in the sludge, both leading to oxygen consumption.

Thermodynamic equilibrium is not attained in a WTP as the retention times are too short and the accompanying reactions too numerous. No relation was found between aeration during lime treatment and higher redox values of sludges. Aged sludges may be approaching equilibrium as they often remain in the same environment for years. As a result, sludges maintained under water cover may display lower redox values than fresh sludges or surface-disposed sludges. This may be due to a limited amount of available oxygen in the disposal environments. As mentioned, residual ferrous iron can consume oxygen as can the polymers used for clarification. The polymers are long organic carbon chains which may slowly biodegrade.

Sludge Chemistry

Table 4 shows a summary of the sludge chemistry. Most sludges contain primarily iron, particularly for the Geco sludge. The Kidd and Mattabi sludges contain more zinc than iron. Other metals vary from site to site. Copper is generally less than one percent, while calcium ranges from 1% to as much as 25%. Sludges produced from basic treatment systems tend to have higher calcium contents. TIC (total inorganic carbon) is generally associated to calcium, likely as CaCO_3 . A concentration of 1% TIC represents 8.3% CaCO_3 .

Lime Consumption

Neutralisation potential (NP) can be used as a rough indicator of plant efficiency with respect to lime consumption. The NP can generally be related to the carbonate content (TIC), as some of the added lime forms calcite with CO_2 dissolved in the slurry either from air or from the raw water. In cases when the AMD is neutralised to a high pH, the sludge may contain unreacted lime. Without measuring the actual NP of the sludge, it can be estimated by a mass balance around Ca. Much of the sulphate in the sludge may be associated with Ca as gypsum is formed in the process and by subtracting the associated fraction of Ca, we obtain a rough estimate of the excess alkalinity. Without a TIC or carbonate measurement, this Ca can either be unreacted lime or calcium carbonates. Mg and other metals can also precipitate in carbonate form.

Generally, HDS-type processes yield sludges with lower TIC contents. Geco sludge has the lowest TIC content. This follows the theory that carbonates formed in the process are used to partially neutralise the AMD in the first reactor (Aubé and Payant, 1997). The theory of calcite dissolution in the process suggests that step-wise neutralisation using sludge for the primary pH increase is beneficial in terms of lime efficiency.

The TIC and NP of the Brunswick, Mattabi and Heath Steele sludges suggest that these plants have good lime neutralisation efficiency. The differences between each of these plants can be due to a variety of reasons, including the TIC of the raw water, the pH control efficiency, the retention time, and the lime slaking efficiency.

Waite Amulet has considerable free alkalinity (315 kg CaCO_3 /tonne sludge) for an HDS plant (50-200 kg CaCO_3 /tonne sludge). The Waite Amulet WTP was constructed in 1984 and lime is added intermittently for pH control in the plant.

This results in pH fluctuations of more than a full pH unit. Large oscillations are detrimental to lime efficiency, but as even the upper peaks of the oscillations are relatively low (~9.6 pH units), such a high NP is surprising. Another difference with the Waite Amulet plant is the use of hydrated lime. Sizing analysis of the Waite Amulet lime slurry showed that the $\text{Ca}(\text{OH})_2$ particles had a D_{50} of 7.3 μm and a D_{90} of 45.5 μm . Typical lime slurry slaked on-site will have sizes closer to 50% passing 5.5 μm and 90% passing 22 μm (Zinck and Aubé, 1999).

Table 4. Chemical Composition of Sampled Sludges (%).

Site		Al	Cd	Ca	Cu	Fe	Mg	Pb	S	SO ₄	Zn	TIC	NP
Brunswick	fresh	3.9	0.0137	3.8	0.12	15.0	3.13	<0.43	4.14	11.80	14.2	0.48	142
Falconbridge Sudbury	fresh	0.1	0.0002	26.6	0.05	4.8	5.8	<0.43	1.43	3.70	0.007	7.06	-
	aged	0.5	0.0009	22.9	0.02	7.1	6.3	<0.43	1.10	3.60	0.021	6.38	725
Geco	fresh	1.4	<0.02	1.8	0.52	46.5	1.34	<0.01	0.78	2.16	2.1	0.15	76
	aged	1.4	<0.02	1.8	0.05	46.3	1.10	<0.01	0.91	1.73	1.8	0.12	62
Heath Steele	fresh	3.54	0.010	2.74	0.72	16.3	3.39	0.22	3.09	8.09	14.6	0.21	164
Kidd Metallurgical	fresh	1.3	0.0674	17.2	0.15	2.3	5.7	<0.42	2.92	6.90	8.5	4.39	471
	aged	0.6	0.1390	10.0	0.20	3.0	7.8	<0.43	4.58	11.29	14.4	2.36	523
Mattabi	fresh	3.47	0.067	5.76	1.40	14.05	5.59	<0.12	2.88	7.73	22.0	0.92	127
	aged	1.85	0.029	7.14	0.63	9.14	5.52	0.038	2.54	7.51	17.6	1.56	197
Waite Amulet	fresh	2.8	0.0029	8.3	0.33	10.8	5.5	<0.43	1.85	5.20	1.4	1.42	319
	aged	3.3	0.0025	7.4	0.27	26.1	2.6	<0.43	1.34	3.77	1.1	2.23	210

At Kidd and Falconbridge, the pH setpoint is higher and reactions do not occur in a stirred tank. As a result, lime consumption and the amount of excess alkalinity in the sludge is higher. The NP of the Falconbridge sludge (725 kg CaCO_3 /tonne sludge) is more than 10 times the NP for the Geco sludge. Like Waite Amulet, Falconbridge uses hydrated lime for neutralisation. A lime excess may be required at the Falconbridge site to enhance settling as the concentration of precipitates formed is low due to the dilute AMD treated.

For Kidd, the high neutralising potential is explained by the setpoint pH of 11 and the lack of effective mixing, both of which reduce the lime dissolution efficiency and increase lime consumption (Zinck and Aubé, 1999). The high pH is required here to enhance settling of the fresh precipitates when the AMD is dilute and to offer excess alkalinity for subsequent thiosalt oxidation.

Sludge Composition and Crystallinity

Sludge composition is affected by the raw water chemistry, reagents used in neutralisation, and the type of treatment process. Most sludges are composed of a major amorphous oxy-hydroxy-sulphate phase which serves to adsorb and collect metals. In contrast, sludge produced at Geco was found to contain a crystalline iron compound, lepidocrocite ($\gamma\text{-FeOOH}$). Crystalline hydrolysis products are rarely observed in sludges produced from lime treatment plants. Any degree of crystallinity in the normally amorphous iron phase is seen as a significant improvement on past treatment results. Sludge crystallinity is an indication of effective supersaturation/crystallisation control during precipitation. As Geco raw water is high in ferrous and low in other metals, the mechanism to grow and crystallise particles is not affected by other co-precipitating and competing metal species as can be the case for other waters. The ferrous could first form 'green

rusts' which are later oxidised to lepidocrocite (Gehring and Hofmeister 1994). The effectiveness of this plant design in producing crystalline precipitates from other raw water compositions is presently under study at CANMET.

The amorphous iron oxy-hydroxides often serve to adsorb other metals such as Zn and Cd (Zinck and Dutrizac, 1998). If crystallisation occurs, these metals are either stabilised in the complexes or precipitated on their own as hydroxides. Both these scenarios represent stronger stabilisation of the metals involved and should thus result in decreased release in the long term. Adsorption effectively scavenges such metals but there is no evidence that this phase serves to immobilise the species. Metal release from crystalline precipitates is generally lower than from amorphous material. Crystalline sludges are also easier and cheaper to handle as they tend to be less viscous and have a higher solids content.

Sludge Stability

Experimental Procedures

The collected sludge samples were leached using two procedures, a leaching test using acetic acid and another using a synthetic acid rain leachant. Use of acetic acid mimics the organic acids expected to be present in a municipal landfill and assumes co-disposal of mineral processing and municipal wastes. The synthetic acid rain was a mixture of 60/40 wt% sulphuric/nitric acid diluted to pH 4.5. The synthetic acid rain procedure simulates the inorganic acids that are likely to come in contact with the sludge through precipitation and as such is more representative of the typical disposal scenarios.

In both cases, the samples were diluted with water by a factor of 16:1 (water:sludge by dry weight). The pH of the leaching solution was monitored at set intervals during the course of the extraction and manually adjusted to pH 5.0 if the pH was greater than 5.2. A maximum of 200 mL of the acid solution was added. Because of the relatively high alkalinity of the sludge samples, this maximum volume of acid was required in all tests. In this situation, the acetic acid test provides more acid for neutralising excess alkalinity than did the synthetic acid rain procedure. It must be noted however, that the synthetic acid rain procedure represents many years of acidic precipitation under the existing disposal scenarios.

Results

Table 5 presents the results of the acetic acid leach tests and Table 6 those of the synthetic acid rain tests. In general, the test using synthetic acid rain leached less than the acetic acid tests. Note that concentrations of all metals except Zn are in µg/L.

Table 5. Acetic Acid Leachate Results.

Site		Cd (µg/L)	Cr (µg/L)	Cu (µg/L)	Fe (µg/L)	Ni (µg/L)	Pb (µg/L)	Se (µg/L)	Zn (mg/L)
Brunswick	fresh	376	<24	<196	<40	<91	<5	<35	27
Falconbridge	fresh	<1	<7	<13	<40	4.9	<2	<24	<0.05
Sudbury	aged	<1	<7	73.3	<40	769	<2	<24	<0.05
Geco	fresh	25.8	7.5	6.3	<10	81	<0.5	<8	2.4
	aged	26.6	5.8	17.1	<10	127	<0.5	<8	6.0
Heath Steele	fresh	1430	9.1	452	<70	138	4.7	<2	216
Kidd	fresh	47.8	<24	<196	<50	<112	<1	132	0.48
Met.	aged	3.7	<7	<13	<40	8.2	<2	186	<0.05
Mattabi	fresh	1620	39.7	361	<40	508	1.3	<48	147
	aged	484	37.3	312	<40	623	1.8	<48	219
Waite	fresh	<4	<34	<235	<40	32.8	<10	<97	0.3
Amulet	aged	<4	<34	<235	<40	11.8	<10	<97	0.2

Most metals in the sludges did not dissolve to any significant degree. Particularly for the acid rain tests, many of the elements are below detection. In comparing these results to field expectations, we must consider the fact that these sludges were submitted an amount of acid (acid to sludge solids ratio) which would take many years to achieve and would be contacted considerably less vigorously in realistic situations. This means that the concentrations expected in a field leachate would be considerably lower. The difference between these tests and reality will depend on the disposal scenario, but if water-covered sludge is taken as an example, the resulting concentrations may be more than 2 orders of magnitude lower.

Table 6. Synthetic Acid Rain Leachate Results.

Site		Cd ($\mu\text{g/L}$)	Cr ($\mu\text{g/L}$)	Cu ($\mu\text{g/L}$)	Fe ($\mu\text{g/L}$)	Ni ($\mu\text{g/L}$)	Pb ($\mu\text{g/L}$)	Se ($\mu\text{g/L}$)	Zn (mg/L)
Brunswick	fresh	<3	<8	<28	<40	<6	<5	<35	<0.06
Falconbridge	fresh	<1	<7	<13	<40	5.2	<2	<24	<0.05
Sudbury	aged	<1	<7	28	<40	26.5	<2	<24	<0.05
Geco	fresh	<5	<3	<14	<20	<10	<0.1	<9	<0.07
	aged	<5	<3	<14	<20	<10	<0.1	<9	<0.07
Heath Steele	fresh	<2	<9	<32	<200	<46	<35	<1.2	<0.10
Kidd	fresh	9.7	<9	<13	<40	19.9	<9	89.8	<0.05
Met.	aged	3.5	<7	<13	<40	7.2	<2	177.5	<0.05
Mattabi	fresh	<2	<134	<28	<40	<78	<7	<28	<0.02
	aged	<2	<134	<28	<40	<78	<7	<28	<0.02
Waite	fresh	<4	<34	<235	<40	<27	<10	<97	<0.06
Amulet	aged	<4	<34	<235	<40	<27	<10	<97	<0.06

Zn and Cd are the two metals most likely to be mobilised. This is true even though the Cd concentrations are typically very low. With the synthetic acid rain tests, the highest leachate concentration was of $9.7 \mu\text{g/L}$, which meets typical discharge limits. For the more aggressive acetic acid test, the highest measured concentration was 1.6 mg/L . Considering the expected rate of release and the volume of water needed to amount to this much acidity, these concentrations remain very low.

Both the neutralisation potential and the initial metal concentration in the sludge strongly affect the amount of metals released (GML, 1987; Zinck et al., 1997 a,b). Two of these sludge samples with similar zinc concentrations showed very different results: Brunswick, a high density sludge with 14.2% Zn and Kidd, low density sludge with 14.4% Zn, showed significantly different degrees of zinc leaching. The resulting acetic acid leachate zinc concentration for Brunswick (27 mg/L) was two orders of magnitude greater than the zinc concentration in the Kidd leachate (0.48 mg/L). This is due primarily to the fact that the Kidd sample has a much higher neutralisation potential than the Brunswick sample, which resulted in a final leachant pH for Kidd of 8.5 while that for Brunswick was 6.8. This comparison illustrates the impact of NP on final leachant pH and hence on long-term stability of sludges.

Samples with similar neutralisation potentials will also show variations in the degrees of metal leaching in accordance with their metal contents. These factors are attributed to the treatment process. thus the type of treatment can impact on metal mobility. In a basic treatment system such as Kidd, the amount of lime consumed is higher and results in sludge with a higher degree of excess alkalinity, as discussed above. Consequently, this excess alkalinity is sufficient to maintain the leachant pH in an alkaline or neutral range, which will reduce metal mobility. Although both sludges are essentially stable for surface disposal, the neutralising potential of the basic treatment sludge suggests that this material could have a practical use instead of simple disposal. Use of sludges as tailings covers is currently under investigation.

Conclusions

1. The Conventional HDS and Geco HDS processes yield sludges with lower particle size distributions than low density processes. The increased bulk density of the sludge seems due primarily to the physical attributes of the precipitates on a microscopic scale.
2. More efficient high density sludge plants tend to produce sludge with less neutralisation potential and a higher degree of crystallinity than sludge produced from basic treatment. This indicates lower lime consumption, but less buffering capacity in the sludge.
3. The metals released when the sludges were tested with synthetic acid rain were minimal, thus suggesting that all of the tested sludges are essentially stable when disposed of in a natural environment (sludge ponds or under water).
4. The Geco process yielded the lowest NP (TIC) and highest degree of crystallinity of all processes examined.
5. Co-disposal of sludges with tailings, use of sludges as covers (oxygen barriers) over tailings and long term densification in dry disposal versus under water disposal should be investigated.

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