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Characterization of mining discharges from the Mining and Industrial Company of Kivu (SOMINKI). Cases of releases from the KALIM Acassiterite treatment plant, Maniema-R D Congo

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Abstract:*This work analyzed the issues of solid mine waste management, taking into account reclamation, the environment and sustainable development. The methodology applied has led us to obtain the following characteristics: the difference of 0.62 to 0.78 between $pH_{water}$and pH_{KCl} attributes to these discharges an average exchangeable acidity; these discharges are dominated by light (density of 1 to 2) and medium-heavy (density of 3 to 4) minerals that are semiconductors (conductivity ranging from 10^{-7} to 10^{5} S/m); some discharges are made up of large porous blocks with cracks because the moisture content varies between 3.8 and 6.6%. The chemical composition of these rejects was dominated by twelve major elements (Sn, Fe, Si, Ti, W, Ca, Al, K, Mn, Mg, Nb and Ta) (>0.1%) of which four oxides (SnO_2, Fe_3O_4, SiO_2 and TiO_2) accounted for more than 85% of the total. Acidity indices greater than 1 classify these discharges in the category of poor hydraulic binders. Three samples (K101, A103 and RA104) were made up of coarse particles of which more than 71% of the upper +75µm contain more than 71% tin. The optical microscope revealed the presence of quartz (SiO_2) and cassiterite (SnO_2) in all samples and other minerals are specific to the samples ; and X-ray diffraction revealed many tin (Sn) minerals such as cassiterite, ixiolite, varlamoffite, megawite, malayaite, wodginite, sorosite, stistaite, nordenskioldine and cernyite where they are associated with the other minerals. The radioactivity of these discharges ranged from 0.0066 to 0.0420 µSv/h per 100 g of material. These values show that there is no danger of occupational exposure. The NAG test revealed that samples A103, RA104 and RA205 in hydrogen peroxide are potentially acid-generating because the solutions have a pH of less than 4.5. This claim was reinforced by the static chemical test because sulphur (Stotal) >0.3%, PNN< 20 kg $CaCO_3$/tonne and NPR<3; simple extraction yields in both solvents, H_2O and $CaCl_2$, are low (2.8-15.3%).*

Keywords*: Cassiterite discharges, characterization of discharges, environmental impacts, recovery of discharges*

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I. Introduction

The problem of managing mining waste is a topical issue because such waste is likely to contain trace metal elements (TMEs), which are indestructible and sometimes toxic at very low levels.

From a sustainable development perspective, the revival of the Congolese mining industry cannot be envisaged without having inventoried and characterized the various historical and currently active mining sites (Kaniki, 2008).

In Maniema, cassiterite, wolfram, coltan and gold mining began in the 1930s, creating several mining companies whose merger in 1974 gave rise to the SociétéMinière et Industrielle du Kivu (SOMINKI) based in Kalima, which was liquidated in 1997 (Mayundo, 2006), leaving huge quantities of mining waste which, until then, had not yet been the subject of any particular study.

In recent years, the issue of mining waste recovery has been of concern to several researchers in the Democratic Republic of Congo, for example, Kaniki (2008), Kitobo (2009), Shengo (2013), Kyona (2014), ..., who have worked on copper-cobalt-rich waste from the former province of Katanga, some for its characterisation and others for its recovery. In this study, the characterisation approach was applied to cassiterite mining waste produced by the SOMINKI-Kalima processing plant; this made it possible to obtain relevant information on this waste in order to be able to assess and monitor its properties, behaviour, characteristics and suitability for recovery.

II. Material And Methods

2.1. Materials

The samples used in this study were collected in a clustered manner relative to the location of the plant. 83 sampling points totalled 62 kg from 5 areas of the site (Figure 1), which were selected based on the storage history of these discharges; RA205 is the oldest (1976-1979) and is located downstream of the discharge channel that flows into the Kamisuku River, followed by RA104 (1980 - 1984) and K202 (1985 - 1990). The A103 area, to the left of the catchment area, is also the oldest (1976 - 1979) but is still disturbed by landslides from Mount Abuki. The K101 area is upstream and recent (1990 - 1997) because the SOMINKI heir company continues to store their waste (Sominki, 1980; Francart, et al., 1982).

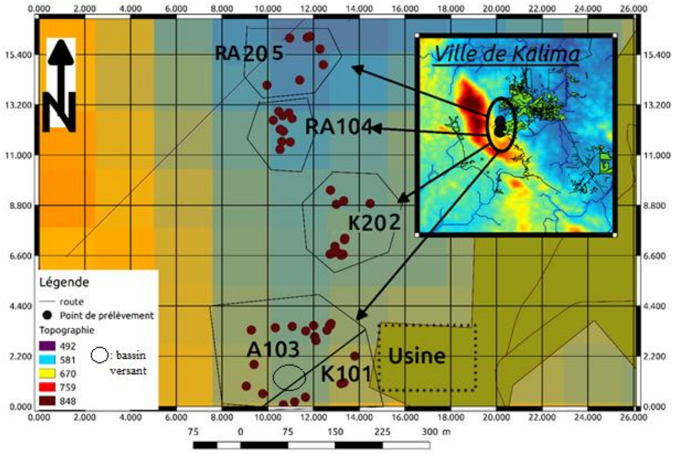


Figure 1: Mapping of sampling points in the city of Kalima

2.2. Methods

The physico-chemical analyses were based on measurements of pH_{water} and pH_{KCl} (CEAEQ, 2005; ISO 10390, 2005); density according to the ISO 17828 standard procedure (Robert, et al., 2018); electrical conductivity (CEAEQ, 2015) and permanent humidity according to the ISO 18134-1 standard (Robert, et al., 2018).

Chemical analyses were performed by X-ray fluorescence spectrometry (XRF) (CEAEQ, 2014), where elemental contents, elemental oxide contents (Kaminski, 2001; Merkel, 2003; Kyona, 2014), acidity indices (Thomas, 2013) and granulochemical distribution (Abdelouafi, et al., 2016; Shengo, 2013; Sutherland, 2003) were determined.

Mineralogical analyses have included both metal-bearing minerals and gangue minerals by light microscopy (Boulmier, et al., 1979; Shengo, 2013) and X-ray diffraction (XRD) (Ghorbel, 2012).

Methods for assessing other characteristics that may have an impact on the environment have been carried out, such as radioactivity by portable INSPECTOR dosimeters using appropriate techniques (Sebastien, 2017), the risk of acid mine drainage (NAG and static chemical tests) (Bouzahzah, et al, 2014a; Bouzahzah, 2013) and simple extraction tests using AFNOR techniques (AFNOR X 31-210, 1988; CEAEQ, 2005; Blanchard, 2000; Hlavackova, 2005; Lassin, et al., 2002; Kitobo, 2009).

III. Results and discussion

3.1. Physico-chemical analysis

The results of the physico-chemical analysis are shown in Table 1. It can be seen from this table that the values of pH_{water} are all higher than those of $pH_{KCl}$with a range between 0.62 and 0.78. This range of deviation gives these samples an average exchangeable acidity. The order of the pH values in relation to the samples also cannot be plausibly explained immediately. All the values found for density are much lower than that of cassiterite (6.80 to 7.30) but samples K101 and A103 are denser than calcite (d=2.70) and quartz (d=2.65) which proves that these rejects are made up of several minerals of different densities. Sample RA205 has an electrical conductivity of 217.00 S/m higher, followed by K202 (119.80S/m) and K101 (117.00S/m), while the others have values below 50.00S/m, so they consist of semiconductor materials (10^{-7} to 10^{5} S/m) which can only be explained using quantum mechanics and the theory of energy bands in solids. Sample A103 has a moisture content value of 6.6%, followed successively by samples RA205 (6.10%), K202 (4.20%), RA104 (4.10%) and K101 (3.80%). The first two samples, visibly, consist of large porous blocks with cracks, which justifies a higher moisture absorption than the others.

Table 1: Physico-chemical analysis of the rejects

Samples	pH			Density	Electrical conductivity (S / m)	Humidity (%)
	Water	KCl	difference			
K101	5.38	4.71	0.67	3.01	117.00	3.80
K202	6.58	5.96	0.62	2.25	119.80	4.20
A103	6.43	5.65	0.78	3.77	29.00	6.60
RA104	6.16	5.40	0.76	1.67	41.70	4.10
RA205	5.51	4.80	0.71	1.48	217.00	6.10

3.2. Chemical analysis of releases

3.2.1. Basic composition of rejects

Chemical analysis of these releases revealed in Table 2 that the five samples consist of twelve major elements (Sn, Fe, Si, Ti, W, Ca, Al, K, Mn, Mg, Nb and Ta) (>0.1%) to which tin (Sn) is considered to be the useful and more concentrated element in the releases; the others are its companions (impurities). It was also noted, in the right vertical direction of the channel (Figure 1), a tendency for tin contents to decrease as one moves away from the discharge point of the discharges (Plant); therefore, the order of density values was much more influenced by the tin contents in the samples.

Table 2. Content of releases

Samples	Percentage content									
	Al	Yes	Fe	S	K	It	Ti	Or	Cr	Mn
K101	0.141	4.008	15,540	0.070	1,175	1,493	3,613	0.010	0.090	0.867
K202	0.260	6.838	24,629	0.090	0.329	0.359	3,229	0.010	0.020	0.899
A103	0.100	2,768	3.919	0.509	1,879	2,699	0.989	0.010	0.020	1.349
RA104	0.133	10,999	14,072	0.386	0.945	0.399	1.309	0.009	0.009	0.263
RA205	0.127	2,731	18,268	0.404	1.056	1,598	5.033	0.008	0.100	1,988

Samples	Percentage content									
	Zr	Zn	Sn	W	Mg	N / A	Th	U	Nb	Your
K101	0.396	0.050	43.184	1.108	0.888	0.858	0.078	0.024	0.244	0.348
K202	0.118	0.013	32,179	1.199	0.618	0.928	0.024	0.009	0.203	0.168
A103	0.006	0.010	57,779	1,329	0.818	0.528	0.003	0.002	0.241	0.378
RA104	0.022	0.010	38.802	1.233	0.627	0.607	0.005	0.003	0.250	0.237
RA205	0.254	0.009	39,488	1,113	0.719	0.132	0.012	0.007	0.591	0.629

3.2.2. Averageoxide content of the waste

Table 3 shows that four oxides accounted for more than 85% of the total, namely SnO_2, Fe_3O_4, SiO_2 and TiO_2. In all samples, cassiterite (SnO_2) dominated in this order 71.18% (A103), 54.65% (K101), 49.95% (RA205), 49.06% (RA104) and 40.68% (K202). We noted that this order was influenced by the tin contents reported in Table 2. Sample A103 was characterized by a higher loss on ignition value of 4.08% compared to 3.79; 3.28; 2.74 and 2.09% for RA205, K101, RA104 and K202 respectively. This decreasing order is comparable to that of CaO in the samples, which justifies the phenomenon of decarbonation by removal of CO_2 from the calcite; therefore, there was no weight gain from the oxidation of $Fe^{2+}$to Fe^{3+} (Shengo, 2013). In view of these SnO_2 contents, these discharges are considered as a deposit focus capable of producing other quantities of cassiterite.

In the field of buildings and public works, these discharges are classified in the category of poor hydraulic binders because the acidity index values (hydraulicity index) are greater than 1 (Table 3) for all samples (Kaniki, 2008); on the contrary, they are considered as mineral binders that release elements with difficulty in acid solution.

Table 3. Oxide contents in the samples

Sam.	Percentage content											PF	Acidity index (I)
	Al_2O_3	SiO_2	Fe_3O_4	Na_2O	K_2O	CaO	TiO_2	MnO_2	SnO_2	WO_4	MgO		
K101	0.13	8.59	17.57	1.16	1.36	2.09	6.05	1.38	54.65	1.48	1.48	3.28	2.40
K202	0.25	14.66	30.07	1.25	0.42	0.53	5.42	1.46	40.68	1.64	1.03	2.09	9.60
A103	0.09	5.94	4.05	0.71	2.29	3.81	1.68	2.17	71.18	1.82	1.37	4.08	1.20
RA104	0.12	23.57	16.02	0.82	1.16	0.56	2.18	0.41	49.06	1.66	1.05	2.74	14.70
RA205	0.11	5.85	20.18	0.18	1.28	2.25	8.43	3.16	50.45	1.50	1.20	3.79	1.70

3.2.3. Granulochemical analysis

It can be seen from Figure 2 that more than 65% of the tin is located in the size ranges 250 to 45 µm for all samples and the remainder is distributed in the >250 and <45 µm size ranges. This distribution is related to the method of treatment used by SOMINKI, which consisted of concentrating dense coarse particles with a particle size greater than 250 µm.

In the perspectives of recovery of these rejects, it will be necessary to pass through the grinding of three samples, K101, A103 and RA205, at less than 20% of the rejects at the 75 µm sieve for a better release of the recoverable metal (Shengo, 2013). Grinding tests on 1kg of each of these samples revealed respective consumptions of 10, 13 and 9 minutes.

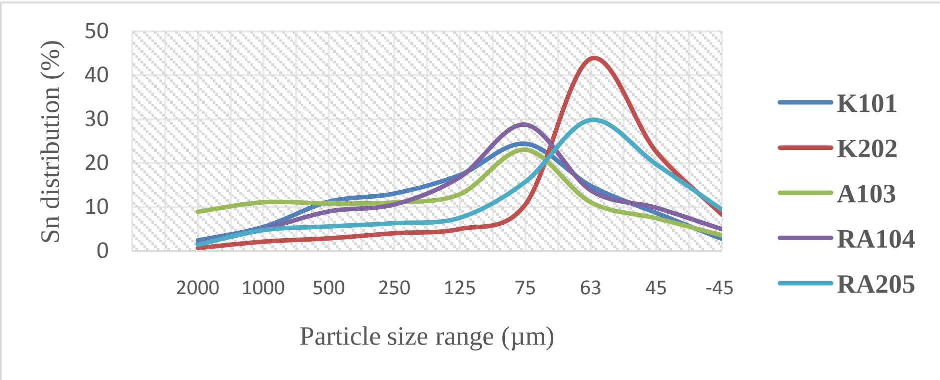


Figure 2. Distribution of the useful element (Sn) in the size ranges

3.3. Mineralogical analysis

The mineralogical analysis by light microscopy in Figure 3 revealed that all samples consist of quartz (SiO2) and cassiterite (SnO_2) as major minerals with rutile (TiO_2) in samples K101, K202 and A103 ;columbite-tantalite [(Fe, Mn)(Ta, Nb)$_2$O$_6$] found in samples K101, K202 and RA205 and augite [Ca(Mg, Fe, Mn, Al, Ti)$_2$(SiAl)$_2$O$_6$] found in samples K101 and K202. The other minerals were considered specific to the samples; they are ilmenite ($FeTiO_3$) in K101, mica [K_2(Mg, Fe)$_6$($Al_2Si_6O_{20}$)(OH)$_{4]}$ and amphibole [NaFe7Si8O$_{22}$(OH)$_2$] in K202, feldspar ($NaAlSi_6O_{20}$), wolframite ($FeWO_4$), phyllite [KCa($Al_3Si_5O_{16}$). 6H_2O], stistaite (SnSb), galena (PbS), qitianlingite ($Fe_2Nb_2WO_{10}$) and pyroxene $(SiO_3)_2$NaFe (Aegyrine) in A103, scheelite ($CaWO_4$) in RA104.

As for the analysis by X-ray diffractometer (XRD), the minerals of tin (Sn) origin were revealed in the diffractograms of Figure 4, these are cassiterite SnO_2 (in K101, K202, A103 and RA205), ixiolite (Ta, Nb, Sn, Mn, Fe)O_2 (in K101), varlamoffite (Nb, Ni)SnO_2 (in K101), megawite $CaSnO_3$ (in K202), malayaite $CaSnSiO_5$ (in K202), wodginiteMn(Sn, Ta)(Nb, Ta)$_2$O$_8$ (in A103), sorositeSnCu (in K202), stistaiteCuSnSb (in K202), nordenskioldine $CaSnB_2O_6$ (in RA205) and cernyite Cu_2CdSnS_4 (in RA205). The gangue has a complex mineralogical composition consisting mainly of oxide minerals, silicates, phosphates, sulphates and other minor classes.

On the basis of these results, it was estimated that these discharges came from deposits associated with albitized or greisenified granitic domes in which pegmatites and quartz veins develop because moderate development of sulphides [PbS and $KAl(SO_4)_2.12H_2O$] has been noted (Varlamoff, 1953).

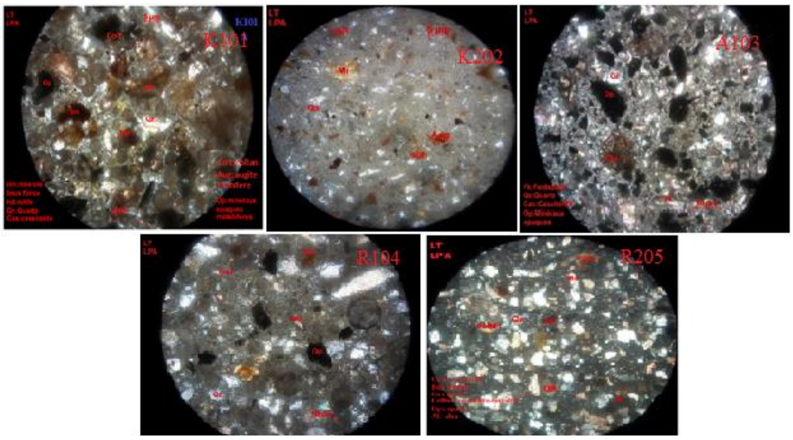
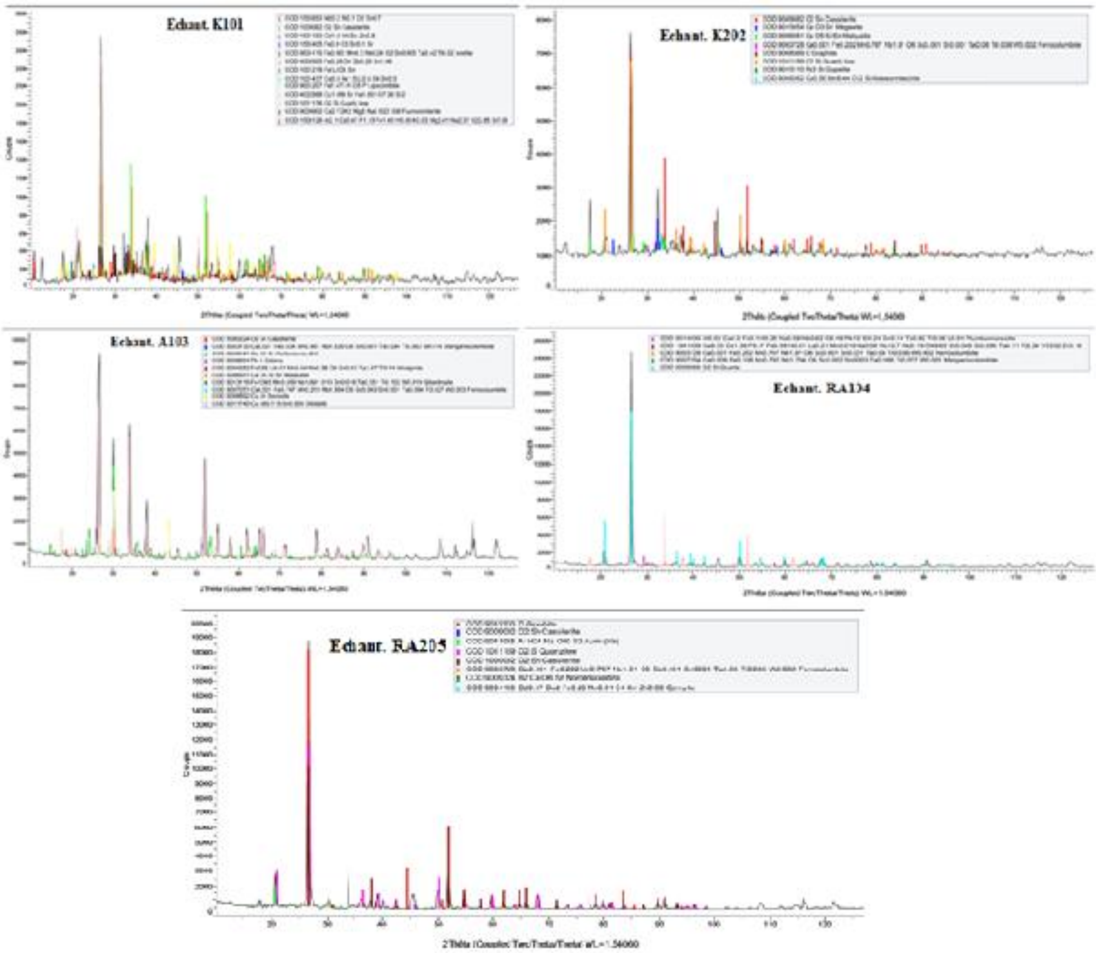


Figure 3. Aspects of minerals in samples under an optical microscope



Figures 4. X-ray diffraction patterns of samples

3.4. Radioactive analysis

It can be seen from Table 4 that sample K101 stood out with a gross activity of 639.54 strokes per minute, which is 11 times the background noise (BF) of the experimental medium against samples K202, A103, RA104 and RA205, which have nearly 2.2, 1.1, 1.7 and 1.5 times the BF respectively. None of these samples is considered radioactive (Journal Officiel de la RDC, 2005). A higher activity of 0.0419 µSv/h per 100g of material in the K101 sample can be justified by high levels of Th (0.078%) and U (0.024%) because the order of their activities in the samples is consistent with that of their Th and U contents.

Table 4. Activities of samples measured in the laboratory

Samples	Weight (g)	Gross activity (A_b)		Corrected activity (A_c) (100gr)		
		In cpm	In µSv / h	In cpm	In µSv / h	A_b / BF
K101	425.23	639.54	0.178	150.40	0.0419	11.0
K202	421.45	128.39	0.036	30.46	0.0085	2.2
A103	405.12	64.40	0.018	15.90	0.0044	1.1
RA104	402.31	96.83	0.027	24.07	0.0067	1.7
RA205	342.30	85.00	0.024	24.83	0.0070	1.5
Background Noise (BF)	-	58.36	0.016	-	-	1.0

3.5. Acid mine drainage analysis (DMA)

The static NAG test revealed that samples A103, RA104 and RA205 mixed with hydrogen peroxide yielded solutions with pH<4.5 (Table 5), which gives these samples potential acid-generating power (Bouzahzah, 2013).

This finding was confirmed by the static chemical test using the method of Sobek (1978), whose values are also recorded in Table 5. Samples A103, RA104 and RA205 are classified as acid-generating by Canadian Mining Industry Directive 019 (Melanson, 2006) as they have values of PNN<17, NPR<3 and % S_{total}>0.3 (Sobek, et al., 1978). There are many reasons for this prediction, as solid mine wastes can contain sometimes non-negligible amounts of unminable metallic minerals, such as the lead sulphides identified in A103. These, when exposed naturally to the action of air (oxygen) and moisture (water), oxidize and generate acidity ($PbS + 8Fe^{3+} + 4H_2O \rightarrow Pb^{2+} + 8Fe^{2+} + SO_4^{2-} + 8H^+$). The presence also of iron in these samples in high quantities is a factor that justifies the acid-generating potential because oxidation of ferrous ion into ferric ion or its precipitation into ferric hydroxide releases a high concentration of acidity (H^+) and causes the formation of a gelatinous precipitate that covers the surface of organic materials and coats the limestone grains (Melanson, 2006).

Table 5. PA, PN, PNN, NPR and pHperoxide values of samples

Sample	% S_{total}	PA	PN	PNN	NPR	H_2O_2 solution pH (NAG)
K101	0.07	2.19	28.75	26.88	15.30	4.9
K202	0.09	2.81	31.25	28.44	11.10	5.8
A103	0.50	15.63	26.25	10.63	1.70	4.1
RA104	0.40	9.38	25.00	15.63	2.70	4.4
RA205	0.40	12.50	28.75	16.25	2.30	4.2

It has been reported that artisanal miners in the area exploit sites RA104 and RA205 where their waste is dumped into the canal leading to the Kamisuku River, the waters of which are used for domestic purposes; a hazard is predicted.

3.6. Testing of simple extractions

The assessment of the ability of trace metal elements (TMEs) to be released to the environment, based on solution testing of these releases in water and in CaCl2 solution, has been interpreted by results reported in Table 6. Overall, the simple extraction efficiency is very low, ranging from 2.8 to 15.3%. This can be explained by the type of gangue (acidic) that conditions the extraction solution to become acidic. In this case, the solubilized heavy metals come mainly from the destruction of the carbonates and oxides/hydroxides that participated in their retention (Sebastien, 1997). Reason which points out that magnesium had higher values followed by iron. By diadochemicalphenomena, the Mg2+ ion, being characterized by the smallest radius, gives a more soluble salt when associated with a voluminous anion such as silicate.

Table 6. ETM contents in simple extraction solutions

Solvent	Ech	Fe		Ti		Sn		Mn		Mg		W	
		% S	Rdt	% S	Rdt	% S	Rdt	% S	Rdt	% S	Rdt	% S	Rdt
H_2O	**K101**	1.21	7.8	0.3	8.3	1.33	3.1	0.03	3.4	0.07	7.9	0.04	3.6
	K202	2.32	9.4	0.31	9.5	1.5	4.7	0.04	4.3	0.06	9.7	0.06	4.9
	A103	0.28	7.1	0.07	6.9	1.81	3.1	0.03	2.2	0.06	7.3	0.05	3.7
	RA104	1.26	9.0	0.11	8.4	1.96	5.1	0.01	3.8	0.06	9.5	0.07	5.7
	RA205	1.81	9.9	0.49	9.7	2.06	5.2	0.11	5.5	0.08	11.1	0.09	8.1
$CaCl_2$	**K101**	1.26	8.1	0.33	9.1	1.32	3.1	0.04	4.6	0.09	10.1	0.04	3.6
	K202	2.4	9.7	0.32	9.8	1.3	4.0	0.04	4.3	0.09	14.5	0.05	4.1
	A103	0.31	7.9	0.09	8.9	1.62	2.8	0.04	2.9	0.07	8.5	0.05	3.7
	RA104	1.32	9.4	0.12	9.2	1.85	4.8	0.02	7.7	0.08	12.7	0.07	5.7
	RA205	1.93	10.6	0.53	10.5	1.98	5.0	0.16	8.0	0.11	15.3	0.08	7.2

IV. Conclusion

A research program has been undertaken to characterize the mining rejects of Company Mining and Industry in Kivu (SOMINKI), case of rejects from the cassiterite processing plant of KALIMA, Maniema/R D Congo in order to evaluate and monitor the properties of these rejects, their behaviour, their characteristics and their aptitude to be valorized. Other characteristics likely to have an impact on the environment, such as the risks of acid mine drainage and radioactivity, were also assessed in order to establish the overall risks that these discharges pose to man and the ecosystem. The implementation of this programme has led to various results, the main results of which can be summarised under various headings:

- the pH_{water} values were all higher than those of pH_{KCl} with a difference that varies between 0.62 and 0.78, giving the samples an average exchangeable acidity;
- with respect to density, these discharges are dominated by light (1-2) and medium-heavy (2-4) minerals but less dense than cassiterite;
- the materials that make up these samples are semiconductors (10^{-7} to 10^{5} S/m);
- the moisture content, which varies between 3.8 and 6.6%, shows that these rejects are made up of large porous blocks with cracks;
- the chemical composition of these rejects was dominated by twelve major elements (Sn, Fe, Si, Ti, W, Ca, Al, K, Mn, Mg, Nb and Ta) (>0.1%) of which four oxides (SnO_2, Fe_3O_4, SiO_2 and TiO_2) totalled more than 85% of the total. The higher acidity indices classify these discharges in the category of poor hydraulic binders;
- three samples (K101, A103 and RA104) were made up of coarse particles of which more than 71% of the slices larger than 75µm contained more than 71% tin; their grindability test set the grinding time at 9 to 13 minutes for 1 kg of these discharges;
- the optical microscope revealed the presence of quartz (SiO_2) and cassiterite (SnO_2) as major minerals in all samples and other minerals are specific to the samples. This analysis was completed by X-ray diffraction which revealed many minerals of tin (Sn) origin such as cassiterite, ixiolite, varlamoffite, megawite, malayaite, wodginite, sorosite, stistaite, nordenskioldine and cernyite where they are associated with oxides, silicates, phosphates and sulphates of other elements;
- the radioactivity of these discharges gave values between 0.0066 and 0.0420 µSv/h per 100 g of material. These values do not pose any danger of occupational exposure because Congolese legislation sets the limit at an average effective dose of 20 mSv/year over 5 consecutive years;
- the NAG test revealed that samples A103, RA104 and RA205 in hydrogen peroxide are potentially acid-generating because the solutions gave a pH<4.5. This assertion was reinforced, for the same samples, by the static chemical test, the results of which also proved to be potentially acid-generating because sulphur (Stotal) >0.3%, PNN< 20 kg $CaCO_3$/tonne and NPR<3 (Bouzahzah, 2013) ;
- the simple extraction yield in both solvents (H_2O and $CaCl_2$) is very low (2.8 - 15.3%) due in particular to the low specific surface due to the coarse grain size and the low porosity of the constituent grains. Solubilized heavy metals mainly come from the destruction of the carbonates and oxides/hydroxides that participated in their retention (Sebastien, 1997);

Since part of these releases has been considered potentially acid-generating, their management requires in-depth studies on their containment, or even their stabilization, using appropriate means depending on the techniques adopted. The remainder of static chemical and mineralogical tests must be carried out before the tin processing and extraction techniques are determined.

Rosuvastatin 20 mg on every other regimen had equal effect when compared to daily dose regimen of atorvastatin 40 mg &rosuvastatin 20mg.

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