Physicochemical and radioactive study of Colombontatalite ore from the Kisengo quarry in the DRC

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ABSTRACT: This research concerns the assessment of radioactivity and the physicochemical characterization of columbotantalite ore extracted from the Kisengo quarry, located in the Tanganyika province of the Democratic Republic of Congo.

The sample was taken from the Kisengo open-pit mine in Nyunzu territory located about 170 km from the town of Kalemie in Tanganyika province/DRC. It contained about 32% niobium, 21% tantalum, 6% iron, 7% manganese, 7% tin, 4% silicon and about 3% titanium and was analyzed by XRF. The results of the radioactivity tests revealed that the average dose rate, measured in nGy/h, was 329.72, or 0.038 mSv with the Identifinder device.

This sample does not pose a risk of radioactive exposure, as the dose levels are very low, in accordance with the WHO range of 20 mSv and DRC Law 017/2002. Analysis of optical microscopy images of Kisengo coltan reveals the presence of a siliceous gangue (quartz) associated with pyroxenes as well as columbotantalite, which is the mineral of tantalum and niobium. Also present are cassiterite, a tin mineral (SnO_2), and pyrolusite, a manganese mineral (MnO_2).

Granulochemical characterization revealed that tantalum and niobium are concentrated in the 500 micrometer fraction. The loss on ignition test indicated that niobium is the most abundant element in the analyzed sample, representing 31.97%, followed by tantalum at 20.75%. Finally, silicon and titanium are present and removed at 4.18% and 17.716%, respectively.

KEYWORDS: Coltan, Radioactive, Hydrometallurgy, Environmental, Minerals

I. INTRODUCTION

The Democratic Republic of Congo is a country rich in precious mineral resources such as diamonds, gold, copper, cobalt, zinc, cassiterite, and coltan. It is true that our country's soil contains economically valuable metals. Unfortunately, the DRC's numerous mining operations do not lead to refining and processing them into finished products. Such rational exploitation would be economically profitable and would have helped reduce, if not eliminate, poverty among the DRC's population.

Indeed, although the DRC has significant coltan deposits underground, the country does not have any processing plants for these minerals to produce the metals. This situation is mainly due to the lack of development of an adequate extraction and purification process. In the DRC, coltan is concentrated only before being exported to other countries. However, it should be noted that coltan contains several valuable metals such as: tantalum, niobium, titanium, iron, etc.

The processing of ore in hydrometallurgy requires several stages of treatment and before processing it, certain characterizations are carried out such as physicochemical characterization, granulometric, and radioactive measurement, etc. the radioactivity test because, this ore sometimes seems to be radioactive which, during laboratory experiments can create environmental damage. The term "coltan" is born from the contraction of columbite and tantalite. (Shikika, 2023)

Coltan in the DRC is primarily mined in the eastern part of the country, particularly in the provinces of South Kivu, North Kivu, Maniema, and Tanganyika.
According to IPIS mapping (2018), there are approximately 160 artisanal coltan mining sites in eastern DRC. Although coltan reserves are not accurately estimated, the DRC holds approximately 80% of Africa's reserves. (Shikika, 2023)

Tantalum and niobium play a crucial role in modern technology due to their physical properties useful in various fields such as electronics, chemistry and nuclear chemistry. It is important to note that, despite the presence of these elements in coltan as composite minerals and that of some other radioactive metallic elements, they must be separated for refining. (Sofia, 2011)

Other researchers have undertaken several methods for the extraction and purification of niobium and tantalum. However, developing new knowledge for the development of dissolution methods that can lead to the dissolution of tantalum and niobium remains a major challenge. (Kitungwa, 2022)

Indeed, environmental constraints and toxicity have prompted manufacturers to explore new production methods that are more environmentally friendly and require less drastic conditions. Traditional processes using fluorinated compounds have a negative impact on the environment. (Sofia, 2011; Deblonde, 2015; Kitungwa, 2022)

The production of pure Nb and Ta metals or their salts is essential for certain applications but it poses a serious challenge in terms of separation process (Deblonde, 2015). In this research, we were interested in controlling the level of radioactive substances found in columbotantalite ore before moving on to the extraction of the tantalum contained within it.

II. MATERIALS, METHODOLOGY, AND QUALITY ASSURANCE

This section of the research focuses on presenting the sample collection site, describing the materials and apparatus, and the experimental procedure used throughout the research.

II.1. Sample Origin

The sample covered in this study is coltan ore. Investigations into the sample's origin revealed that it was taken from the Kisengo open-pit mine, located at longitude 28°16'43.6"E and latitude 5°35'16.4"S, approximately 170 km from the town of Kalemie in Tanganyika Province, specifically in Nyunzu Territory.

II. 2. Materials (see ISO 17025 standard)

After receiving the sample, we headed to the Polytechnic Faculty where the sample preparation and actual testing were carried out, followed by various chemical analyses.

II. 3. Radioactivity Testing

The purpose of radioactive characterization is to determine the radioactive dose rate generated by the ore. From this determination, it is possible to assess whether the sample presents a risk of exposure or not.
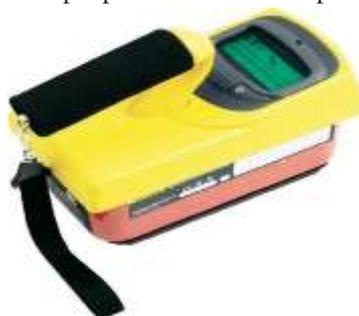

Fig. 1. Identifinder (Google)

The measurement was carried out by spreading the sample to cover the surface of the device's detector. The stability of the value displayed on the screen requires about twenty seconds. Thus, the standards of doses admitted (in sievert: Sv) by the World Health Organization (WHO) are shown in the following table 12:

Table I. Dose standards (Sv) accepted by the World Health Organization (WHO)

Dose interval	Dose designation
0 to 20 mSv (0.02 Sv)	Very low doses
20 to 200 mSv (0.2 Sv)	Low doses
200 to 2000 mSv (2 Sv)	Medium doses
2 to 10 Sv	High doses
Over 10 Sv	Very high doses

II. 4. Particle Size Test

The particle size characterization aims to identify in which particle size range the majority of tantalum and niobium was trapped. To achieve this, eight sieves with different apertures were used in decreasing order of size and subjected to a vibrating screen for 30 minutes. At the end of the sieving process, a certain weight of oversize was retained on each sieve and ground for chemical analysis by particle size range.

To determine the chemical particle size distribution, the following experimental procedure was followed:

• Place 1000.00 grams of coltan on the first sieve stacked in descending order;

• Start the vibro-sieve for 15 minutes to separate the grains according to their size, as determined by the 500, 425, 315, 250, 160, 125, 100, and 75 µm sieves;

• Collect the residue from each sieve separately at different pitches;

• Weigh each residue collected and place it in a 0.5 g bag;

• Grind approximately 5.00 grams of residue collected from each sieve for chemical analysis by X-ray Fluorescence (XRF);

From the results of the chemical analysis, proceed to calculate the weight of the metals as well as the chemical particle size distribution using the formulas below:

$$Metal\ weight\ (g) = \frac{(\%\ element\ (chemical\ analysis)\ *\ weight\ of\ residue\ on\ the\ sieve)}{100}$$

The distribution of the element on each sieve is given by the following formula:

$$Distribution\ of\ the\ element(\%) = \frac{mass\ of\ metal\ per\ sieve}{Total\ mass\ of\ metal} * 100\%$$

II. 5. Mineralogical Testing

Mineralogical analysis identifies and quantifies the minerals present in a sample, which is essential for the evaluation of mineral resources and their efficient exploitation. It helps understand the composition and structure of rock formations, which is crucial for geological studies and research on the evolution of the Earth's crust.

This analysis was carried out in the mineralogical laboratory of the Faculty of Sciences in the Department of Geology using an optical microscope in transmitted and reflected polarized light.

II. 6. Geochemical Test

This test was conducted to determine the oxide content of the various chemical elements present in the sample using a loss on ignition analysis. To do this, 25g of finely ground samples were placed in a furnace at 950°C for 60 minutes to remove any chemical matter likely to volatilize, ensuring accurate inclusion in the oxide calculation (Yuma P., 2019).

Calculations of oxides and loss on ignition are carried out using the following formulas:

+ $Oxide\ content\ (\%) = \frac{\%element\ (chemical\ analysis)*Mm\ of\ the\ oxide}{Atomic\ mass\ of\ the\ element}$ **(II.3)**

+ **PF = weight of the sample before calcination – weight after calcination (II.4)**

II. 7. Chemical Analysis

This analysis was performed to determine the composition of various chemical elements contained in the sample. During our investigations, three spectral instruments were used for chemical analysis:

II. 7. 1. X-ray Fluorescence (XRF)

To reveal the weight composition of chemical elements contained in solid samples, we used XRF from the company Chemaf (Chemical Africa) under the Olympus brand, as shown in the image below.

Fig. 2. XRF device brand OLYMPUS from the company CHEMAF (photo 2021)

II. 7. 2. How XRF Works

The sample is bombarded by photons emitted from an X-ray tube. Electrons belonging to the inner shells of atoms are then ejected. It therefore makes it possible to obtain the chemical composition of a sample quickly, accurately, and non-destructively.

II. 7. 3. Induction Plasma-Optical Emission Spectrometer (ICP-OES)

This spectrometry was used for the analysis of chemical elements in solution. For this analysis, we also used the ICP-OES from Chemaf, branded Spectro-Arcos.

Fig. 3. The ICP – OES of the CHEMAF company (photo 2021)

III. PRESENTATION OF RESULTS AND DISCUSSION

This section presents the results of the radioactive test as well as the physicochemical analysis of the coltan ore sample collected at the Kisengo mining site.

III. 1. Radioactive Test Results

This analysis was conducted to determine the radioactivity levels present in the ore under examination. The results obtained by the FGR detector at the Faculty of Sciences of the University of Lubumbashi, specifically in the Geology Department, at various points along the well-spread sample are shown in Table 1 below.

Table II. Rate of doses of radioactivity detected at different points

Dose rates in nGy/h			Average dose rates in nGy/h		
332.5	316.1	328.0	329.72		
324.6	326.5	321.6			
317.6	321.1	320.6	**Sample dose rate in mSv/h**		

338.5	340.9	332.5	0.038
331.5	346.9	346.9	

This analysis shows that the value obtained is 0.038 mSv/h. This shows that the sample does not present a risk of radioactive exposure. The value is well below the recommended threshold, i.e. it is in the WHO very low dose range (0 to 20 mSv).

III.2. Results of the chemical composition of the KISENGO coltan ore

This analysis was conducted to quantify and characterize the nature of the ore under investigation. The results were obtained by XRF analysis and are shown in Table II below.

Table III. Results of the content of chemical elements contained in coltan ore

Elements	Ta	Nb	Si	Sn	Fe	Mn	Ti	Cr	As	Zr	Bi
Contents (%)	20.75	31.97	4.18	6.85	5.88	6.76	2.97	0.50	0.03	0.09	0.44

Chemical analysis of the sample showed that the ore studied is largely composed of 31.97% Niobium and 20.75% Tantalum followed by 6.86% Tin; 6.76% Manganese; 5.88% Iron; 4.18% Silicon and 2.97% Titanium. It also contains trace metals such as Chromium (0.50%); Arsenic (0.03%); Zirconium (0.09%) and Bismuth (0.06%).

These results confirm that the sample under examination is much more dominated by tantalum and niobium, that is to say, we are in the presence of an ore which is full of both of the elements mentioned above.
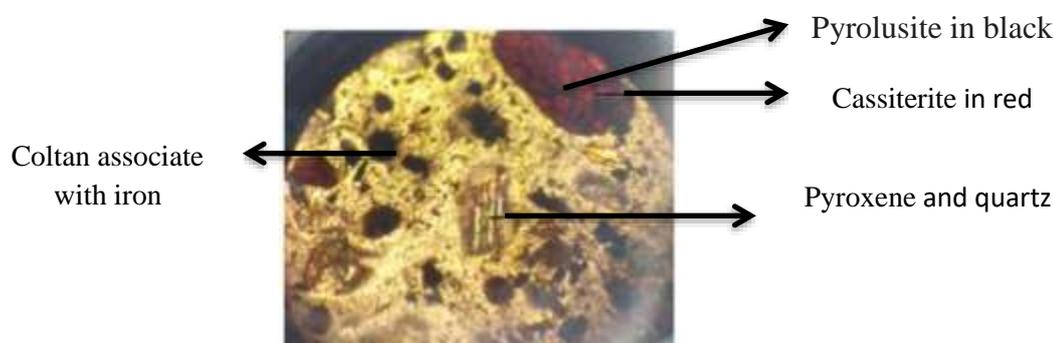
Fig. 4. Optical microscopy image for the determination of minerals in coltan ore

Examination of the image from optical microscopy of Nyunzu coltan reveals that it contains, among other things:
- siliceous matrix (quartz) associated with pyroxenes, which are iron and magnesium minerals associated with silica, with the general formula: $(Mg, Fe, Ti, Al) (Si, Al)_2O_6$, the color of which in the image is between white and light yellow (Bédard P., 2011);
- columbotantalite, a tantalum and niobium mineral also associated with iron, and in the image it appears as more or less large black spots trapped between quartz and pyroxene;
- Cassiterite, a tin mineral, SnO_2, which appears in red in the image under examination;
- Pyrolusite, a manganese mineral, MnO_2, which is represented in the image by the black spot trapped in the red color of cassiterite. This information will be compared with the results of the chemical analysis to confirm the chemical elements that constitute these different elements.

III.4. Granulochemical Characterization

This characterization was carried out to determine how tantalum and niobium are distributed in the different particle size ranges. In the case under examination, we used eight sieves, including 500, 425, 315, 250, 160, 125, 100, and 75 μm. Exactly 1,000.00 grams of coltan ore, previously ground to -2.00 mm, were fed onto the sieve with a large mesh opening.

The results obtained are classified in Table 3 below. And for the sake of facilitating the presentation, interpretation or discussion of the results, we considered tantalum, niobium, tin, manganese; iron, silicon and titanium are present in the sample under examination.

Table IV. Results of the granulo-chemical distribution of elements according to the granulometric ranges

| Sieve mesh (µm)	Weight refusal (g)	refusal %	Cantent of chemical elements in (%)							Weight of chemical elements in (g)							Granulo-chemical distribution (%)	
			Ta	Nb	Si	Sn	Fe	Mn	Ti	Ta	Nb	Si	Sn	Fe	Mn	Ti	Ta	Nb
500	370.50	37.24	20.75	31.97	4.18	6.85	5.88	6.76	2.97	76.89	118.43	15.47	25.39	21.80	25.03	11.00	42.34	53.45
425	17.10	1.72	20.69	20.96	3.75	2.24	7.91	6.45	4.90	3.54	3.58	0.64	0.38	1.35	1.10	0.84	1.95	1.62
315	159.2	16.00	18.57	18.57	3.88	2.22	11.19	5.34	8.69	29.56	29.56	6.18	3.54	17.82	8.49	13.83	16.28	13.34
250	134.3	13.50	16.18	15.98	3.48	1.90	12.28	3.86	9.50	21.73	21.46	4.67	2.55	16.49	5.18	12.76	11.97	9.69
160	138.90	13.96	14.47	14.14	3.79	1.90	15.78	3.28	19.01	20.10	19.64	5.26	2.63	21.92	4.55	26.40	11.07	8.86
125	114.70	11.53	15.96	15.50	3.90	2.10	17.36	3.97	23.52	18.30	17.78	4.47	2.41	19.91	4.55	26.97	10.08	8.02
100	39.50	3.97	18.06	17.63	3.77	2.95	13.71	4.09	21.38	7.13	6.97	1.49	1.17	5.42	1.61	8.44	3.93	3.14
75	6.80	0.68	18.33	18.23	3.88	2.95	13.63	4.35	21.31	1.25	1.24	0.26	0.20	0.93	0.30	1.45	0.69	0.56
-75	14.00	1.41	22.01	20.92	3.90	3.61	9.96	4.68	11.45	3.08	2.93	0.55	0.51	1.39	0.65	1.60	1.70	1.32
Total	995.00	100.00								181.57	221.58	38.99	38.79	107.03	51.47	103.30	100.00	100.00
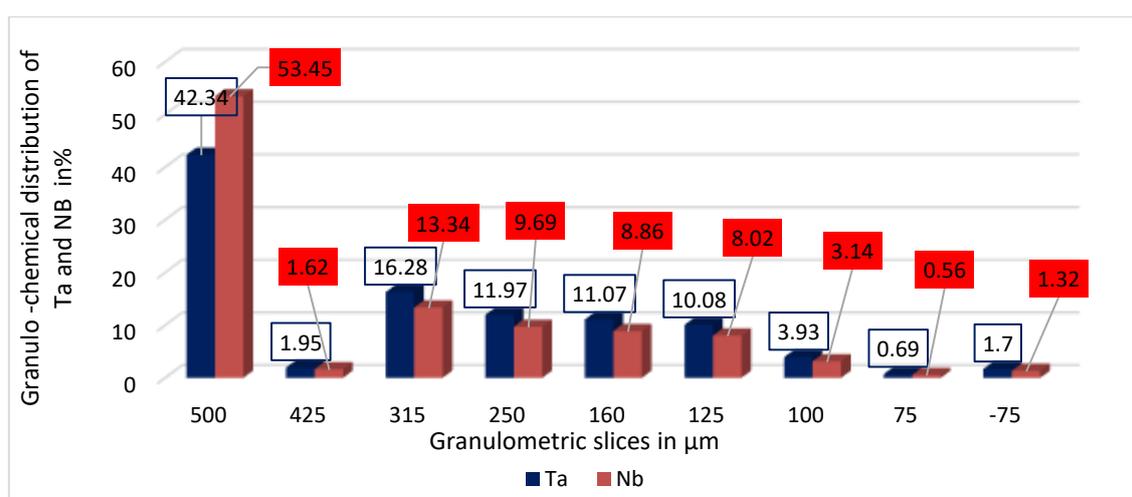
Fig. 5. Shape of the histograms of the granulo-chemical distribution of tantalum and niobium in the different sieve slices

Analysis of Table IV and the histograms presented in Figure 5 above shows that tantalum and niobium are most concentrated in the 500 micrometer range. This observation shows that at this range, the targeted metals are more in the coarse mineral particles; that is, the 500µm range in which tantalum and niobium are respectively concentrated at 42.34% and 53.45%.

Based on the theory of Blazy P., 2003; it is obvious that the ore under examination must be fragmented to release the mineral particles of the desired metals. Thus, the rest of the tests will be carried out with a particle size of -75µm in view of leaching.

III.5. Results of the chemical and geochemical analysis of the ore

The results of the chemical and geochemical analysis of the coltan ore are presented in Table V below.

Table V. Results of chemical and geochemical analysis of the ore

Content of chemical elements in sample in %						
Ta	Nb	Si	Sn	Fe	Mn	Ti
20.75	31.97	4.18	6.85	5.88	6.76	2.97
Chemical element content after test (%)						
Ta	Nb	Si	Sn	Fe	Mn	Ti
23.751	21.406	4.697	1.998	14.416	3.972	17.716
Oxide content after loss on ignition test (%)						
Ta_2O_5	Nb_2O_5	SiO_2	SnO_2	Fe_2O_3	MnO_2	TiO_2
57.99968	61.226	10.065	2.5353	41.189	6.283	29.527
Loss on fire = 0.8 g						

It is noted in Table V above that niobium is the most abundant element in the sample under examination with 31.97%; followed by tantalum with 20.75% and tin, manganese, silicon, iron, silicon and titanium represent respectively 6.85%, 6.76%, 5.88%, 5.88% and 4.18% in the sample.

By comparing the results obtained here with those obtained during the mineralogical analysis, we note that the latter confirm the results of the mineralogical analysis. Indeed, tantalum, niobium and iron are present in columbotantalite while iron, titanium and silicon with implied magnesium are present in pyroxene, silicon and iron are present in quartz and manganese in pyrolusite while tin is present in cassiterite.

The results of the geochemical analysis confirmed that the ore under investigation contains more tantalum and niobium oxides than it is indeed columbotantalite. These assertions also meet the results of the mineralogical analysis which presented pockets of black spots composed of ferrous columbotantalite. As with the chemical analysis, titanium oxide is the least represented in this series.

CONCLUSION

This work aimed to assess radioactivity levels and perform physicochemical characterizations of tantalum and niobium contained in coltan from the Kisengo quarry in the Democratic Republic of Congo, Tanganyika Province.

- ➢ The acquisition of coltan ore of approximately 4 kilograms in the form of a package wrapped in a black bag, from Kalemie to Lubumbashi and containing information on the origin of the sample. This information revealed that it came from the Kisengo open-pit mine in the territory of Nyunzu, in the province of Tanganyika;
- ➢ The granulochemical characterization of the sample was carried out using a series of 8 sieves including 500, 425, 315, 250, 160, 125, 100 and 75 µm (Sieves placed in our possession during this study);
- ➢ Chemical, mineralogical and geochemical characterization was carried out for the quantification and qualification of the ore under examination.

This procedure was carried out in the metallurgical laboratory of the Polytechnic Faculty (for sample preparation and experimental tests, in the Geological Laboratory of the Faculty of Sciences for mineralogical analysis and in the CHEMAF company laboratory for chemical analyses.

Following our investigations, the following points should be noted:

- - For the mineralogical analysis, the ore on which this study was based is a columbotantalite whose gangue is made up of quartz, pyroxene, cassiterite as well as pyrolusite. These results were confirmed by chemical analysis, which in turn

revealed that the ore consisted of 32% niobium, 21% tantalum, 6% iron, 7% manganese, 7% tin, 4% silicon, and about 3% titanium. These are just some of the elements that XRF from CHEMAF was able to reveal to us. These observations were also consistent with geochemical tests revealing the contents of these elements in the form of oxides.

- For the granulometric test, it was found that the Kisengo coltan ore consisted mainly of coarse particles with a granulometry greater than 100 µm. While the granulochemical analysis had shown that tantalum and niobium were found at the extremes of the values of the granulometric sieves used.

This observation showed the need to grind the sampled ore in order to release the desired metals and increase the contact surface during subsequent processes.

From all the above, it is therefore possible to extract tantalum and niobium in their pure state by observing some of the procedural conditions as outlined in this work. However, it must be said that the intention of this research has only given part of the expected results due to the lack of availability of certain specific analysis devices such as DRX, SEM (Scanning Electron Microscope) and IR (Infrared). With these devices, we suggest that this reflection be deepened by other researchers in the near future.

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