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Hydrometallurgical assessment of oxides of Nb, Ta, Th and U from Ethiopian tantalite ore

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ABSTRACT

Tantalite ore is the main source of the metals niobium (Nb) and tantalum (Ta). Today, the needs for these economic and irreplaceable Nb, and Ta metals are becoming imperative for the technological world, to meet the demand, a lot of research is needed to ore reserves, extraction methodology and simplified refining processes should be in plethora. This study focuses on the Ethiopian tantalite ore located south of the Neoproterozoic Adola belt of the rare element Kenticha pegmatite deposit (global source of tantalite). Successive beneficiation of pegmatite ore results in a highest dissolution of 60.83 wt% of Ta₂O₅ with 4.58 wt% of Nb₂O₅ including the removal of U, Th, Ti, Fe and Si, resulting in a percentage low weight, which allows transporting ore concentrates. The concentrated ore is leached with a mixture of binary acids HF and H₂SO₄ with 6:1 ratio at temperatures from 100 to 400 °C. The filtered solution was extracted with MIBK then, precipitated with ammonium solution. The hydrometallurgy of Nb and Ta oxides is affected by leaching temperature and the presence of radioactive oxides of U and Th. Elemental analysis and surface topography were studied using SEM/EDS, revealing the highest percentage composition of Nb and Ta oxides at 200 °C of leaching temperature. Comparatively, the compositions of U and Th showed higher amounts at the leaching temperatures of 150 and 200 °C, and when the temperature reached about 350 and 400 °C, the percentage composition of U and Th became very low, the economic metals Nb and Ta were completely leached. The stability of the precipitated samples was analyzed using TGA-DTA and found to be thermally stable, and not contain significant moisture at each temperature studied. Studies for recovery of Ta, Nb, U and Th from Kenticha tantalite are required.

1. Introduction

Columbo tantalite, one of the most economical mineral and listed as an important mineral in the United States, is derived primarily from pegmatite constituents of the form $((Fe,Mn)(Ta,Nb)_2O_6)$ [1–4]. Tantalum is being utilized more frequently in new technologies and has grown its importance as a technological component in the modern economy [3]. Ethiopian Kenticha pegmatite is classified into four groups based on the mineral composition: spodumene, albite-spodumene, muscovite feldspar, and beryl. In order to ensure that the enriched product satisfies export standards, pegmatite-spodumene, which has the highest composition of Mn-tantalite, also contains radioactive elements like Th and U, which need to be removed with caution [4,5]. Fe, U, Th, Ti, and Si are examples of associated

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impurities that are frequently present and coexist with valuable minerals and must be removed in order to treat and beneficiate Nb and Ta oxides. Since these elements have similar chemical characteristics, they are hard to separate [6], and in order to remove Ta and Nb from traditional multi-element analyses, solid samples need to undergo intricate mineralization processes [3].

When naturally occurring radioactive materials are extracted from raw materials using hydrometallurgical processes, mining and processing-related activities can lead to high levels of these materials in products, by-products, and wastes [6–8]. Due to the extreme harsh conditions required for the process, the leaching of Ta and Nb is still very difficult [8–12]. Concentrated HF is currently used to treat the majority of tantalite ore. This reaction can be used to illustrate how Mn-tantalite dissolves in HF (Eq. (1)) [11].

$$MnNb_2O_6/(MnTa_2O_6) + 16HF \rightarrow 2H_2NbF_7/(2H_2TaF_7) + MnF_2 + 6H_2O$$
(1)

On the other hand, these decomposition processes are linked to HF volatilization losses, which is harmful to both human (operator's health) and equipment (deleterious and corrosive). When removing HF pollution at the source during the production of Ta and Nb, these reactions need to be carefully considered [12]. Furthermore, all commercial hydrometallurgical processes that produce Nb and Ta requires the presence of F^- [4,12]. Thus, in order to lower the process's level of toxicity, a specific quantity of HF should be swapped out for H₂SO₄ [13–15]. Using organic extractants like tributyl phosphate, cyclohexanone, 2-octanol, methyl isobutyl ketone, and high molecular weight amines, the Nb and Ta ions are removed from the dissolved solution. On the other hand, because of its great solubility, high chemical and operational stability, and low density and cost, methyl isobutyl ketone is the most widely used extractant [12,13].

For this reason, radioactive oxides of U and Th have been found in Kenticha pegmatite ore (an exportable concentrated tantalite ore) [14,15]. The nature of the ore, or the concentration of value metals and impurities in this case U, Th, Fe, Ti, and Si, determines the hydrometallurgical properties of tantalite ore. Tantalum ore leaching parameters like temperature, time, and particle size have all been investigated before [14,15]. The quantity of U and Th metal ions leached from tantalite ore revealed a significant amount. Furthermore, uranium, which is normally present in uranium minerals found in U^{4+} and U^{6+} oxidation states. Pegmatite deposits are the primary source of uranium and thorium ores [1,2]. Eqs. (2) and (3) describe the dissolution chemistry of U in acidic solutions, where U^{4+} must first be oxidized to U^{6+} . Strong oxidants, like Fe³⁺, are necessary to enhance its dissolution during acid leaching, which results in the formation of uranyl ions (Eq. (4)) [16–22]. Ores frequently contain Fe as a constituent. The leaching efficiency of U^{6+} is about 95 % [3]. Eq 2 through 5 illustrate the process by which U and Th dissolve in acid [16–22].

$$\begin{array}{ll} UO_{3} + 2H^{+} \rightarrow UO_{2}^{2+} + H_{2}O \ (dissolution)) & (2) \\ 2UO_{2} + O_{2} + 4H^{+} \rightarrow 2UO_{2}^{2+} + 2H_{2}O \ (oxidation) & (3) \\ UO_{2} + 2Fe^{3+} \rightarrow UO_{2}^{2+} + 2Fe^{2+} & (4) \\ ThO_{2} + 4H^{+} \rightarrow Th^{4+} + 2H_{2}O & (5) \end{array}$$

Thorium complexes are created when thorium oxide combines with acids. At high temperatures, these reactions are completed more quickly [1]. During leaching, Ta–Nb ore experiences a sequence of high-temperature reactions in addition to strong acid reactions, where U–Th remains in solution at high concentrations [3]. The oxidation state, matrix composition, contamination, solubility, economic viability, tailing management, and other environmental factors all play a role in the lixiviant selection process for U and Th leaching [15–22].

The assessment of the hydrometallurgical oxides of Nb, Ta, U, and Th from the Ethiopian Kenticha pegmatite ore is the ultimate goal of this work. Taking temperature into account, radioactive oxides of U and Th were eliminated from the binary acid-leaching oxide of Nb and Ta, and the oxides of Ta and Nb were then extracted, precipitated, and provided with additional ideas for pretreatment and extraction. XRD, TGA-DTA, EDX-SEM, and AAS spectroscopies were used to examine both liquid and solid samples.

2. Materials and methods

2.1. Equipment and reagents

Employing an Anton Paar PerkinElmer Multiwave 3000 microwave reaction system fitted with an 8SXF100 rotor and eight PTFE reaction vessels, a concentrated tantalite sample was leached using a solution of 6 M HF and 1 M H_2SO_4 . TGA was used to analyze the weight fraction, weight loss, and thermal stability of tantalite ore samples as their temperature increases. Model TA 50, Shimadzu America, and platinum cell in nitrogen with a flow rate of 30 mL/min were the instruments used. SEM with EDS (JEOL JSM-848 model) was used to examine the compositional and topographic changes that occurred both before and after the samples were leached. The dissolution procedure was conducted in a stainless batch reactor (SUS316) within a high-temperature furnace set at different temperatures of 100 -400 °C. No additional purification was required when using research-grade HF, H_2SO_4 , ammonia solution, and methyl isobutyl ketone (MIBK) from Sigma-Aldrich.

2.2. Methods

The ore was mined from the upper zone of the Kenticha pegmatite deposit in Oromia Regional State, Ethiopia (Fig. 1, Sample 1) [4, 8,13,14]. 10 kg of Kenticha pegmatite ore (Sample 1) was crushed and grinded by hand with a mortar and pestle to <200 mesh size.

These steps are performed by washing and decanting to remove residues. The sample was then shaken to recover the dark brown to black portion of the ore. Further processing was performed by gravity separation (25 °C, 250 rpm, 1 h) and magnetic separation to recover 1 kg of enriched Mn-tantalite, commonly known as tantalite (sample 2 in Fig. 1).

Based on the author's studies, the decomposition of a tantalite ore sample was conducted in a high-temperature furnace using a stainless steel (SUS316) batch reactor [4,12–14]. In a separate sample holders, the 5 g of sample 2 was introduced in the furnace with 100 mL of 6 M HF + 1 M H₂SO₄ and reacted for an hour at a 100, 150, 200, 250, 300, 350, and 400 0 C reaction temperatures and at a constant string speed at atmospheric pressure. Each sample was then allowed to fuse for 1 h (labeled samples 3 to 9, respectively). Each of the products was cooled at room temperature, and then leached with 100 cm³ de-ionized water (resistivity of 18.2 MΩ cm). The filtrates and residues were obtained by filtration. Quantitatively, each filtrate was moved to a 100 mL volumetric flask, and the leached residue was collected for further study (Fig. 1, sample R) [12–14].

Leaching, filtration, separation, and extraction were among the sequential processes that comprised the solvent extraction technique [13–17]. By altering the study on solvent extraction in fluoride media, the solvent extraction procedures were carried out [4]. For 30 min, seven samples (labeled as samples 3 through 9, respectively) were agitated in an atmospheric dry box with 50 mL of each filtrate solution containing sample with 50 mL of MIBK (1:1) [15,22,23]. The mixture was divided into organic/upper phase (anticipated to contain Ta, Nb, U, and Th) and aqueous/lower phase (expected to contain active metals like Fe, Al, and Si and considered the raffinate). This was done using a separatory funnel. In trials involving triplets, the aqueous phase was extracted once more to optimize the extraction efficiency using 10, and 5 mL of MIBK extractant, respectively, and all upper phases were collected together to each labeled sample container. All lower phases are collected and stored together for further analysis, removal, and F⁻ processing.

For each of them, an excess of 30% ammonia solution was added to the collected organic/upper phase sample to precipitate it, forming a cake, which was then dried for three days at 100 °C in an oven. Once more, the dried solid samples were calcined for an hour at 1200 °C in a furnace. For every sample of three to nine, a comparable producer was completed (see Scheme 1). XRD, EDXRF, TGA, and SEM-EDS were used to examine the calcined solid samples.

3. Results and discussions

3.1. Ore characterization

Based on previous works; and quantitative analysis of an tantalite ore is blessed in high elemental compositions of Ta, and Nb, the Kenticha-Ethiopia ore revealed that the composition of Kenticha deposit varied significantly, and the activity of some concentrations of radionuclides is the main source of the radiation dose [4,8,12,14,15]. The amount of U and Th oxides in enriched ores is always related to beneficiated tantalite ores, as there are the main drawbacks when performing Nb and Ta extraction, transporting and exporting [4]. Sequential processing of pegmatite ore is the best alternative approach to obtain higher grades of tantalite ore than primary processing of pegmatite ore (Table 1), thus, the concentrated Tantalite ore (sample 2) was scored 60.83 wt% of Ta₂O₅ starting from 2.32 wt% of Ta₂O₅ of the deposit pegmatite ore (sample 1). Similarly, 4.58 wt% Nb₂O₅ was obtained from 0.45 wt% Nb₂O₅, respectively (Table 1). These sequential beneficiations of pegmatite ore attributes to the increase in wt% of Ta₂O₅ and Nb₂O₅, refining and preparing them for further processing by removing untargeted minerals as considered as tailings. In addition, these results attributes the increments of Nb and Ta oxides directly related with the increments of U and Th oxide and indirectly related composition of Ti, Fe and Si oxides. In addition, the results are supported by SEM-EDS spectroscopic pattern (Fig. 2). These results are similar with a previous studies by the authors, XRD spectra of Kenticha Tantalite ore by using acidic leaching (Fig. S1) [4,12].



Fig. 1. XRD diffraction of Kenticha pegmatite ore (sample 1), concentrated tantalite ore (Sample 2) and leached residue (Sample R) [4].



Scheme 1. Schematic diagram of a connected approach hydrometallurgy of tantalite ore.

Table 1

Typical chemical composition of Kenticha pegmatite ore, beneficiated tantalite and precipitated oxides fraction using EDXRF^a, as metallic oxide.

Ore (sample code)	Percentage, wt%								
	Ta ₂ O ₅	Nb ₂ O ₅	TiO ₂	Fe ₂ O ₃	ThO ₂	U_3O_8	SiO ₂		
Pegmatite ore (Sample 1)	2.32	0.45	3.56	25.54	0.09	0.80	12.14		
Mn-Tantalite (Sample 2)	60.83	4.58	0.96	5.71	0.33	1.23	2.01		
Fe-Tantalite (Sample R)	2.11	6.11	14.48	56.11	0.22	1.01	10.23		
(Sample 3) at 100 °C	46.32	4.87	0.11	3.11	0.26	0.21	0.23		
(Sample 4) at 150 °C	51.23	4.46	0.08	0.11	0.27	0.12	0.21		
(Sample 5) at 200 °C	34.11	0.11	0.03	4.97	0.33	0.07	0.14		
(Sample 6) at 250 °C	32.12	0.11	0.04	0.11	0.41	0.19	0.11		
(Sample 7) at 300 °C	25.65	0.11	0.12	0.11	0.30	0.14	0.13		
(Sample 8) at 350 °C	6.84	0.11	0.11	82.38	0.21	0.05	0.23		
(Sample 9) at 400 °C	0.11	0.11	5.34	86.45	0.01	0.01	6.54		

^a (Tantalite ore leached by 6 M HF with 1MH₂SO₄, extracted by MIBK and precipitated by NH₄OH, scanned the precipitated samples).

3.2. Leached and extracted tantalite ore

According to a prior study, 6 M HF and 1 M H_2SO_4 was chosen as the ideal acid concentration ratio to leach Nb and Ta from tantalite ore [12]. In a batch reactor, the acid leaching solution was run at various leaching temperatures (100, 150, 200, 250, 300, 350, and 400 °C) at constant atmospheric pressure. The filtered aqueous solution was then carefully mixed with a 1:1 (O/A) amount of MIBK to dissolve and extract the impurities from the aqueous/lower phase and the Nb and Ta metal ions from the organic/upper phase. The extracted upper phase (containing Nb, Ta, U, and Th) was separated, stripped and extracted from the aqueous phase, while adding MIBK and then, the collected upper phase solution was precipitated with excess ammonium solution [13–15]. Following the precipitated samples' drying and calcination, EDXRF, EDS-SEM, XRD and TGA are used to characterize the solid samples (labeled samples 3 to 9).

The primary factor influencing the target metals and radioactive oxides dissolution performance in the tantalite ore leaching process is temperature. Ta and Nb are predicted to remain intact at higher leaching temperatures, but generally speaking, raising the leaching temperature can result in an increase in the leaching rate of U and Th (Table 1). When the leaching temperature was raised from 200 to 400 °C, the amount of Ta_2O_5 dropped to 0.11 wt%, according to the EDXRF elemental composition. Initially, Ta_2O_5 increased from 46.32 to 51.23 wt% at the leaching temperatures of 100 and 150 °C (samples 3,4). Furthermore, at a leaching temperature of 100 °C, 4.87 wt% of Nb_2O_5 was obtained (sample 3). This indicates that 100% of Nb_2O_5 precipitated at 100 °C and 84% (2.11 wt%) of Ta_2O_5 precipitated from the tantalite ore at 150 °C. EDS spectroscopy of specific regions of the precipitated ore samples



Fig. 2. SEM-EDS of tantalite ore (sample 2).

(Fig. 3 and Fig. S2) confirms the generalizability of these results, which show that 150 °C is the ideal leaching temperature to separate and extract Nb and Ta from tantalite ore (sample 4). As a consequence, raising the leaching temperature from 100 to 400 °C causes the wt% of Nb, Ta, U, and Th oxides to decrease to a minimum value (0.11 wt%). Nevertheless, even at leaching temperatures of 400 °C, the radioactive oxides of Th and U continued to exist, producing 0.01 wt% ThO₂ and U_3O_8 (Table 1).

The degree of leaching reduces as hydrolysis takes place at leaching temperatures above 300 °C [13,16]. Similar studies under these circumstances showed that Nb leached at a maximum rate of 73.5% at 200 °C (sample 5), while tantalum hydroxide, or 30.3 % of the total Ta, departed the concentrate as leachable species in the liquid [13–16]. However, comparable studies revealed that overheating reduces and dehydrates some rare earth elements, resulting in the formation of an insoluble sulfate that reacts with the leaching agent (H₂SO₄). These investigations also verified that the leaching efficiency of U and Th at various temperatures was less than 45%, which may have been caused by the presence of Si and Ti [18–22]. In comparison to tantalite ore (parent ore, sample 1), this study revealed that Th and U oxides decreased to 3.03% and 0.81% (0.01 wt%), respectively, at extraction temperatures ranging from 100 to 400 0 C. Ti and Fe oxides are responsible for these characteristics.

Based on the sample's limited areas selected by EDS (Fig. 3 and Fig. S2), the surface of the precipitated oxides' selected areas verified a high composition of Ta, Nb, and Mn at leaching temperatures ranging from 100 to 200 °C, corroborating the findings of related studies [11–22]. Comparatively, at leaching temperatures of 150 and 200 °C, the compositions of U and Th were higher, and the value even decreased as the leaching temperature rose above 200 °C (Fig. 3, Fig. S2, and Table 2). Similarly, at the higher leaching temperatures of 350 °C and 400 °C (samples 8,9), respectively), the limited screening area of the sample by EDS diffraction spectroscopy revealed the absence of U and Th. According to these findings, a series of procedures including leaching tantalite ore with 6 M HF and 1 M H₂SO₄ at a high temperature, extracting with MIBK, and precipitating the samples with ammonium solution were used to determine whether radioactive oxides of U and Th were present. However, comparable research revealed that the leaching of tantalite

Table 2

Elemental Composition of tantalite ore using Scanning Electron Microscope Equipped with Dispersion Spectrum Analysis (SEM-EDS) (eZAF Smart Quant Results of selected area^a).

Leaching temperature, ⁰ C	Weight % of element										
	СК	OK	NbL	ThM	UM	MnK	FeK	TaL	SK	SiK	TiK
a) 100, Sample 3	5.76	33.64	4.37	0.25	0.19	9.37	2.72	43.70	-	-	-
b) 150, sample 4	5.85	33.04	4.03	0.25	0.11	8.25	-	48.47	-	-	-
c) 200, sample 5	25.00	32.85	-	0.31	0.05	4.85	4.85	32.08	-	_	-
d) 250, sample 6	-	46.70	-	0.38	0.17	8.66	-	30.97	13.13	-	-
e) 300, sample 7	5.25	50.69	-	0.28	0.11	7.11	-	22.90	13.66	-	-
f) 350, sample 8	2.69	35.42	-	0.18	0.03	-	48.77	6.72	6.19	-	-
g) 400, sample 9	-	38.17		-	-	-	49.25	-	6.88	3.05	2.65

^a (Tantalite ore leached by 6 M HF with 1MH₂SO₄, extracted using MIBK and precipitated using NH₄OH, scanned selected limited area from the precipitated surface of the samples) K, L, M represents atomic shells from where EDS spectroscopy propagates.

ore in the presence of HF in the leaching solution also contributes significantly to the extraction of Nb and Ta using different extractants, resulting in the extraction of Nb and Ta oxide in studies where U exists [12,13,22,23].

As the leaching temperature rose to 150 °C, the extraction and dissolution efficiency of Nb declined; however, Nb did not appear in any limited areas of precipitate samples 5 to 9 at any temperature above 150 °C (Table 2). However, depending on the percentage composition of the extracted Nb and Ta oxides, the extraction and dissolution efficiencies of Ta, Mn, Th, and U increased and then decreased up to 200 °C. The distinctive patterns of tantalite ore were seen at 30° 2Theta, while leaching at temperatures ranging from 100 to 200 °C (Fig. S1). These results were corroborated by XRD (Fig. 4, samples 3,5). Thus, Ta, Nb, U, and Th were the remaining oxides after leaching, separation, and precipitation of the tantalite ore, as indicated in Fig. 3 and Table 2.

Furthermore, these compositions demonstrated the presence of U and Th with leaching temperatures up to 350 °C, as shown in the EDS diffraction spectra in Fig. 3 and Table 2. The dissolution efficiency of Nb and Ta oxides can also decrease with increasing temperature. Since practically all of the polymetallic elements were very low during the extraction process, 100 and 150 °C were thus the preferred optimal leaching temperatures of Nb and Ta from Kenticha tantalite ore with the recommended weight % of Th and U (Table 1). The dissolution and extraction efficiency of Nb, Ta, U, and Th on the precipitated samples dropped to almost zero wt% at temperatures above 300 °C, while non-targeted elements like Ti, Fe, and Si saw an increase in these processes. Consequently, at temperatures higher than 300 °C, the presence of impurities, chiefly, roughly 3.5 times the amount of iron oxide was revealed during leaching with an HF and H_2SO_4 mixture. Studies revealed that during the leaching and precipitation of tantalite ore in the presence of U and Th, these processes occurred more quickly before they were finished at high temperatures [18–24].

As a result, the most distinctive tantalite ore diffraction pattern at 30° 2Theta was identified by the XRD diffraction pattern results (Fig. 4 and Fig. S1 sample 6–9), which were lost. Instead, it is evident that the pattern changes at higher leaching temperatures than at



Fig. 3. EDS of a selected area of precipitated oxides from tantalite ore.

(Tantalite ore leached by 6 M HF with $1MH_2SO_{4.}$, extracted by MIBK and precipitated by NH_4OH , scanned selected limited area from the precipitated surface of the samples).



Fig. 4. XRD diffractions of selected area of acidic leached and extracted tantalite ore.

lower ones. This is because high temperatures cause impurities like sulfate, Fe, Ti, and Si to leach. Studies have verified that the more intense features in the XRD pattern of tantalite ore are located at 30° 2Theta [2,4,12,14,15]. Diffractions therefore support the EDS weight percent findings for tantalite ore that has been leached, extracted, and precipitated. (Table 2, Fig. 4 and Fig. S2).

Selected samples tantalite ore stability was examined using TGA-DTA (Fig. S3), which revealed that the sample loses very little weight (0.2%–2%) when heated to 1000 °C, was indicating that the reaction is exothermic. The low weight loss suggests that the tantalite samples, which are separated and extracted at a processing temperature of 1000 °C, are free of impurities, thermally stable, and contain little water or gaseous decomposition products.

4. Conclusions

This work focuses on the rare element Kenticha pegmatite deposit in Ethiopia, which is situated south of the Neoproterozoic Adola belt and contains tantalite ore. Pegmatite ore undergoes successive beneficiation, resulting in a maximum dissolution of 60.83 wt% of Ta_2O_5 with 4.58 wt% of Nb₂O₅, including the removal of U, Th, Ti, Fe, and Si. This low wt% makes it possible to transport and export ore concentrates. The performance of a mixture of 6 M HF and 1 M H₂SO₄ as a decomposed of concentrated tantalite ore was investigated in relation to temperature. For each sample, MIBK was used to extract the filtrates following stages of decomposition at 100 to 400 °C. A mixture of solutions, including impurities like Ti, Fe, and Si, is collected in the upper phases and precipitated by adding an ammonia solution oxide of Nb, Ta, U, and Th. Utilizing SEM/EDS, elemental and compositional analyses of the acid-leached and MIBK extract revealed the highest compositional percentages of Nb and Ta oxides at 100 and 150 °C, respectively, reaching 100% and 84% leaching temperatures. At leaching temperatures between 100 and 200 °C, surface compositions have high wt% of Ta, Nb, and Mn. By contrast, the wt% of U and Th compositions was high at leaching temperatures of 150 and 200 °C, and it was almost zero at the higher 400 °C leaching temperature. Using SEM/EDS and EDXRF, 0.11 wt% of Nb and Ta and nearly zero (0.01) wt% of U and Th were found in the selected area at higher leaching temperatures of 350 and 400 °C. Generally speaking, 150 ⁰C is the ideal leaching temperature to separate and extract Nb and Ta from tantalite ore. Thermal stability of tantalite ore's was examined using TGA-DTA

analysis, and it was discovered that neither the ore's separation nor extraction at processing temperature contained any appreciable amounts of neither moisture nor gaseous breakdown products. Studies are needed to recover Ta, Nb, U, and Th from Kenticha tantalite.

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Availability of data

The datasets used and/or analyzed during the current study are available from the corresponding author upon reasonable request and attached supported file of XRD (Fig. S1), SEM/EDAX TEAM (Fig. S2) and TGA-DTA (Fig. S3).

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

CRediT authorship contribution statement

Goitom Gebreyohannes Berhe: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization, Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Mulugeta Sisay Cheru:** Writing – review & editing, Resources, Methodology, Investigation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Dr. Goitom Gebreyohannes Berhe reports financial support was provided by Mekelle University, College of Natural and Computational Sciences. Dr. Goitom Gebreyohannes Berhe reports a relationship with Mekelle University College of Natural and Computational Sciences that includes: employment.

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Appendix A. Supplementary data

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References

- N.T. Nassar, S.M. Fortier, Methodology and technical input for the 2021 review and revision of the U.S. Critical Minerals List: U.S, Geological Survey Open-File Report 2021–1045 (2021) 31, https://doi.org/10.3133/ofr20211045.
- [2] Z. Zhu, C.Y. Cheng, Solvent extraction technology for the separation and purification of niobium and tantalum: a review, Hydrometallurgy 107 (1–2) (2011) 1–2, https://doi.org/10.1016/j.hydromet.2010.12.015.
- [3] M. Filella, Tantalum in the environment, Earth Sci. Rev. 173 (2017) 122–140, https://doi.org/10.1016/j.earscirev.2017.07.002.
- [4] G.B. Gebreyohannes, V. del Rosario Alberto, A. Yimam, G. Woldetinsae, B. Tadesse, Alternative beneficiation of tantalite and removal of radioactive oxides from Ethiopian Kenticha pegmatite-spodumene ores, Int. J. Miner. Metall. Mater. 24 (2017) 727–735, https://doi.org/10.1007/s12613-017-1456-8.
- [5] E.M. Martinis, P. Berton, R.P. Monasterio, R.G. Wuilloud, Emerging ionic liquid-based techniques for total-metal and metal-speciation analysis, TrAC, Trends Anal. Chem. 29 (10) (2010) 1184–1201, https://doi.org/10.1016/j.trac.2010.07.013.
- [6] I. Paiva, R. Marques, M. Santos, M. Reis, M.I. Prudêncio, J.C. Waerenborgh, M.I. Dias, D. Russo, G. Cardoso, B.J. Vieira, E. Carvalho, Naturally occurring radioactive material and risk assessment of tailings of polymetallic and Ra/U mines from legacy sites, Chemosphere 223 (2019) 171–179, https://doi.org/ 10.1016/j.chemosphere.2019.02.057.
- [7] D. Klingelhöfer, M. Braun, J. Dröge, A. Fischer, D. Brüggmann, D.A. Groneberg, Environmental and health-related research on application and production of rare earth elements under scrutiny, Global Health 18 (2022) 86, https://doi.org/10.1186/s12992-022-00879-5.
- [8] F.P. Carvalho, M.B. Tufa, J.M. Oliveira, M. Malta, Radionuclides and radiation exposure in tantalite mining, Ethiopia, Arch. Environ. Contam. Toxicol. 81 (2021) 648–659, https://doi.org/10.1007/s00244-021-00858-8.

- [9] K. Kiegiel, A. Abramowska, P. Biełuszka, G. Zakrzewska-Koltuniewicz, S. Wołkowicz, Solvent extraction of uranium from leach solutions obtained in processing of Polish low-grade ores, Journal of radioanalytical and nuclear chemistry 311 (2017) 589–598, https://doi.org/10.1007/s10967-016-5029-5.
- [10] A. Shikika, F. Zabene, F. Muvundja, M.C. Mugumaoderha, J.L. Colaux, M. Aatach, S. Gaydardzhiev, Extraction of Ta and Nb from a coltan bearing ore by means of ammonium bifluoride fluorination and sulfuric acid leaching, Minerals 11 (12) (2021) 1392, https://doi.org/10.3390/min11121392.
- [11] O.M. El-Husaini, M.N. El-Hazek, Removal of radioactive elements from niobium and tantalum ores, Eur. J. Miner. Process. Environ. Protect. 5 (1) (2005), 1303-0868.
- [12] B. Tadesse, G. Berhe, V. Alberto, A. Yimam, G. Woldetinsae, Decomposition of the Kenticha mangano-tantalite ore by HF/H₂SO₄ and KOH fusion, Physicochem. Probl. Miner. Process. 54 (2) (2018) 406–414, https://doi.org/10.5277/ppmp1840.
- [13] O.M. El-Hussaini, M.A. El-Hakam Mahdy, Extraction of niobium and tantalum from nitrate and sulfate media by using MIBK, Min. Pro. E.rt. Met. Rev. 22 (4–6) (2002) 633–650, https://doi.org/10.1080/08827500208547433.
- [14] M.S. Cheru, A.V. Del Rosario, A.Y. Ali, G.G. Berhe, B. Tadesse, Separation of radioactive elements from Ethiopian Kenticha pegmatite ore by hydrometallurgical methods, JOM 70 (2018) 2807–2812, https://doi.org/10.1007/s11837-018-3187-3.
- [15] M. Cheru, A. del Rosario, A. Yimam, B. Tadesse, G. Berhe, Hydrometallurgical removal of uranium and thorium from Ethiopian tantalite ore, Physicochem. Probl. Miner. Process. 10 (2018) 448–547, https://doi.org/10.5277/ppmp18153.
- [16] O.M. El-Hussaini, M.A. Mahdy, Sulfuric acid leaching of Kab Amiri niobium-tantalum bearing minerals, Central Eastern Desert, Egypt, Hydrometallurgy 64 (3) (2002) 219–229, https://doi.org/10.1016/S0304-386X(02)00045-2.
- [17] R. Venter, M. Boylett, The evaluation of various oxidants used in acid leaching of uranium, in: Hydrometallurgy Conference, The Southern African Institute of Mining and Metallurgy, 2009, pp. 1–12.
- [18] Z. Zhaowu, P. Yoko, Y.C. Chu, Separation of uranium and thorium from rare earths for rare earth production a review, Miner. Eng. 77 (2015) 185–196, https://doi.org/10.1016/j.mineng.2015.03.012.
- [19] M. Huang, K. Hu, X. Li, Y. Wang, J. Ouyang, L. Zhou, Z. Liu, Mineralogical properties of a refractory tantalum-niobium slag and the effect of roasting on the leaching of uranium-thorium, Toxics 10 (2022) 469, https://doi.org/10.3390/toxics10080469.
- [20] S.A. Neveen, S. Mahmoud, M. Khalafalla, M. Abdel, R. Mahmoud, T. Khattab, Studying the leaching of uranium, thorium and REE from microgranite at the south eastern part of wadi baroud, north Eastern Desert, Egypt, J. Rad. Nucl. 3 (2021) 211–218, https://doi.org/10.18576/jrna/060304.
- [21] M. Huang, K. Hu, X. Li, Y. Wang, J. Ouyang, L. Zhou, Z. Liu, Mineralogical properties of a refractory tantalum-niobium Slag and the effect of roasting on the leaching of uranium-thorium, Toxics 10 (2022) 469, https://doi.org/10.3390/toxics10080469.
- [22] M. Nete, W. Purcell, E. Snyders, J.T. Nel, G. Beukes, Characterization and alternative dissolution of tantalite mineral samples from Mozambique, Journal of the Southern African Institute of Mining and Metallurgy 112 (12) (2012 Jan) 1079–1086. ISSN 2225-6253.
- [23] Z. Zhu, C.Y. Cheng, Solvent extraction technology for the separation and purification of niobium and tantalum: a review, Hydrometallurgy 107 (1–2) (2011) 1–2, https://doi.org/10.1016/j.hydromet.2010.12.015.
- [24] M.S. El Sayed, Uranium extraction from gattar sulfate leach liquor using aliquat-336 in a liquid emulsion membrane process, Hydrometallurgy 68 (1–3) (2003) 51–56, https://doi.org/10.1016/S0304-386X(02)00159-7.