Eksplorium
Volume 47 No. 1, May 2026: 15–28

p-ISSN 0854-1418
e-ISSN 2503-426X

Preliminary Investigation of Catechin as a Natural Complexing Agent for Selective Recovery of Tantalum and Niobium from Hydrometallurgical Leachate of Bangka Tin Slag 2

Sulaksana Permana[1], Agustino Zulys[2], Widiantari Nofriandani[2], Kurnia Trinopiawan[3,5], Riesna Prassanti[3,5*], Iwan Susanto[4], Asep Mohamad Noor[1], Johny Wahyuadi M. Soedarsono[5]
[1]Department of Mechanical Engineering, Gunadarma University,
Margonda Raya St., No. 100, Depok, 16424, Indonesia
[2]Department of Chemistry, Faculty of Mathematics and Natural Sciences (FMIPA),
Universitas Indonesia, Depok, 16424, Indonesia
[3]Research Center for Nuclear Materials and Radioactive Waste Technology,
Research Organization for Nuclear Energy, National Research and Innovation Agency (BRIN)
B. J. Habibie Science and Technology Park, Tangerang Selatan, 15314, Indonesia
[4]Department of Mechanical Engineering, Politeknik Negeri Jakarta,
Prof. Dr. G.A. Siwabessy St., Depok, 16424, Indonesia
[5]Laboratory of Prof. Johny Wahyuadi Soedarsono, Department of Metallurgy and Materials,
Faculty of Engineering, Universitas Indonesia, Depok, 16424, Indonesia
*E-mail: ries002@brin.go.id

Article received: 10 January 2026, revised: 4 May 2026, accepted: 31 May 2026
DOI: 10.55981/eksplorium.2026.14940

ABSTRACT

Bangka Tin Slag 2 (BTS 2), a byproduct of tin processing, contains strategic metals such as tantalum (Ta) and niobium (Nb) in oxide forms. This study explores the potential of catechin extract from green tea leaves (Camellia sinensis) as a complexing agent for the recovery of Ta_2O5 and Nb_2O_5 from BTS 2 following a staged leaching process. The material underwent a staged leaching process, defined as a sequential alkali-acid treatment (NaOH followed by HCl) to remove major impurities, followed by hydrofluoric acid (HF) digestion. Catechin was isolated with a yield of 4.73%. Leaching optimization revealed that 4 M HF was optimal, achieving extraction efficiencies of 3.879% for Ta and 23.109% for Nb. Interaction studies using Job's method showed a 1:1 stoichiometric ratio (metal:ligand) for the complexation. However, catechin exhibited a dominant complexing efficiency with iron (Fe^{3+}) at 12.23%, compared to only 1.55% with Ta^{5+} and 0.21% with Nb^{5+}. Based on the Hard-Soft Acid-Base (HSAB) theory, this preferential binding occurs because Fe^{3+} acts as a hard acid that matches perfectly with the hard oxygen-donor phenolic groups of catechin, whereas the highly charged Ta^{5+} and Nb^{5+} ions form overly stable fluoro-complexes in the leachate that restrict ligand exchange. These findings indicate that while catechin is a potent complexing agent, its selectivity toward Ta and Nb requires further enhancement or an intermediate iron-removal step.

Keywords: Bangka Tin Slag, tantalum, niobium, catechin, selective extraction

INTRODUCTION

Indonesia is a crucial player as one of the world's largest tin producers, ranking second globally [1]. The tin smelting industry inherently produces tin slag as a byproduct, traditionally considered waste. However, this slag is a significant repository of valuable metals, including strategic refractory metals such as tantalum (Ta) and niobium (Nb) [2]. Tin slag is classified into two main types: Bangka Tin Slag 1 (BTS 1) and Bangka Tin Slag 2 (BTS 2). BTS 2 is produced by remelting BTS 1, which contains 20-30% tin, in a reverberatory furnace [3]. Within the BTS

2 matrix, Ta and Nb are generally found as stable oxides, namely tantalum pentoxide (Ta_2O_5) and niobium pentoxide (Nb_2O_5) [5].

Tantalum and niobium are high-value strategic elements essential to modern industrial applications. Tantalum, known for its excellent corrosion resistance, high melting point, and ability to form stable oxides, is vital in the electronics sector (especially capacitors), superalloys, aircraft components, nuclear reactors, optical lenses, jewelry, and industrial equipment [6]. Similarly, niobium plays a crucial role in producing high-strength steel alloys, superconductors, and rocket components [7]. Given the projected increase in global market demand for both metals, exploring alternative sources beyond primary ores is imperative to ensure a sustainable supply and reduce dependence on limited resources. BTS 2, with its substantial Ta and Nb content, offers a promising prospect as a sustainable secondary source [8], [9].

Previous studies have confirmed the feasibility of extracting Ta and Nb from tin slag. Conventional methods such as chlorination and carbochlorination have been explored [10]–[12], while hydrometallurgical approaches, including staged leaching with alkali-acid solutions followed by HF-H_2SO_4, have also been successfully applied [13]–[15]. Leaching and solvent extraction are common hydrometallurgical techniques for recovering Ta and Nb from various sources, including slag [16]–[18]. However, the main challenges are separating Ta and Nb, which have very similar chemical properties, and effectively removing common impurities such as iron (Fe_2O_3), silicon (SiO_2), and titanium (TiO_2) [18]–[20]. The use of corrosive and toxic inorganic reagents, such as hydrofluoric acid (HF), on an industrial scale also raises environmental and safety concerns, prompting the search for more environmentally friendly and sustainable alternatives.

Given its mineral wealth, Indonesia is also a significant producer of green tea (*Camellia sinensis*). Green tea leaves are rich in secondary metabolites, including catechins. Catechins are water-soluble, colorless, bitter-tasting flavonoids [21]. Catechins are widely known for their antioxidant properties and are used in the pharmaceutical and health industries [22], [23]. However, their molecular structure, characterized by active phenolic hydroxyl groups, shows strong potential as metal-complexing agents [20], [24]. The interactions of catechins with various metal ions have been extensively studied, particularly regarding catechin stability and bioavailability, as well as their ability to bind heavy metals [25], [26]. The potential of catechins as biodegradable natural ligands makes them attractive candidates for developing greener extractive metallurgical processes.

Given the potential of catechins as natural complexing agents, this preliminary study was conducted to investigate the interactions between catechin extract and tantalum and niobium obtained via staged leaching of BTS 2. The main objective of this research is to evaluate the complexing efficiency of catechin toward Ta and Nb, as well as its selectivity for Fe impurities, in the BTS 2 leachate solution. This study also aims to identify challenges and opportunities in the use of natural complexing agents in extractive metallurgy, particularly for the recovery of strategic metals from industrial waste. The results of this research are expected to provide a basis for developing more environmentally friendly and sustainable extraction methods and to pave the way for the utilization of local biomass in the mining industry.

p-ISSN 0854-1418
e-ISSN 2503-426X

METHODOLOGY

This study was designed to investigate the interaction between catechin extract and tantalum and niobium obtained from the staged leaching of BTS 2. The research methodology includes catechin isolation and characterization, initial BTS 2 characterization, staged leaching, catechin interaction with leached metals, and comprehensive data analysis.

Materials and Equipments

The primary material in this study was green tea leaves (*Camellia sinensis*) Assamica variety, obtained from a tea plantation in West Java, Indonesia, as a source of catechin. BTS 2 samples were obtained from a tin smelting facility in Bangka Belitung, Indonesia. Chemical reagents included analytical-grade sodium hydroxide (NaOH) (Merck, 99%), analytical-grade hydrochloric acid (HCl) (Merck, 37%), analytical-grade hydrofluoric acid (HF) (Merck, 48%), analytical-grade sulfuric acid (H_2SO_4) (Merck, 98%), and organic solvents such as ethanol (Merck, 96%) and ethyl acetate (Merck, 99.5%) for catechin isolation. Deionized water was used throughout the experiments.

The equipment used included standard laboratory glassware (Pyrex), a drying oven (Memmert ULM 500), a furnace (Nabertherm L 9/11/P330), a leacher with magnetic stirrer and temperature controller (IKA RCT Basic), a centrifuge (Hettich Universal 320), a UV-Vis spectrophotometer (Shimadzu UV-1800) with quartz cuvettes, Fourier Transform Infrared (FTIR) spectrometer (PerkinElmer Spectrum Two) with ATR accessory, X-Ray Fluorescence (XRF) spectrometer (Bruker S8 TIGER), and Microwave Plasma-Atom Emission Spectroscopy (Agilent 4200 MP-AES) for analyzing metal concentrations.

Catechin Isolation and Characterization

Green Tea Leaf Determination and Preparation

Fresh green tea leaves (Camellia sinensis) were rinsed thoroughly under running water, air-dried at room temperature for 24 hours, and then oven-dried at 50°C for 48 hours until their weight remained constant. The dried leaves were subsequently ground into a fine powder using a blender (Philips, HR2116), resulting in a particle size of less than 60 mesh, suitable for extraction.

Catechin Isolation

Catechin was isolated from green tea leaf powder by maceration followed by liquid-liquid extraction. One hundred grams of green tea leaf powder were macerated in 1000 mL of 70% ethanol at room temperature for 24 hours with constant stirring (150 rpm). The ethanol extract was filtered through Whatman No. 1 filter paper, and the residue was re-macerated twice with the same volume of solvent. The combined filtrates were concentrated on a rotary evaporator (Buchi, R-210) at 40°C to obtain a viscous extract. The viscous extract was dissolved in deionized water and extracted three times with ethyl acetate (1:1, extract:ethyl acetate). The ethyl acetate phase was collected, dried over anhydrous sodium sulfate, and re-concentrated on a rotary evaporator to obtain crude catechin solid. The solid was further purified by recrystallization from an ethanol-water (1:1 v/v) mixture to obtain a purer catechin. Final purity was evaluated by High-Performance Liquid Chromatography (HPLC) (Agilent, 1260 Infinity II) with a UV detector at 278 nm.

Phytochemical Screening Test

The catechin extract was analyzed by phytochemical screening to identify secondary metabolite groups, including flavonoids (Shinoda test), tannins ($FeCl_3$ test), and saponins (foam test). This analysis confirms the presence of catechin as the primary compound and detects any other possible interfering substances.

Thin Layer Chromatography (TLC) Test

TLC was performed to separate the components of the catechin extract and compare them with a catechin standard (Sigma-Aldrich, >98% purity). The stationary phase was silica gel GF254, and the mobile phase was a mixture of ethyl acetate:methanol:water (8:1:1 v/v/v). The TLC plate was visualized under 254 nm UV light and sprayed with 1% $FeCl_3$ solution. The Rf value of the catechin extract was compared with the catechin standard to confirm the presence of catechin and evaluate its purity.

Characterization with FTIR and UV-Vis

The isolated catechin solid was characterized using an FTIR spectrophotometer to identify the functional groups present. Spectra were collected from 4000 to 400 cm^{-1} at a resolution of 4 cm^{-1} with 16 scans. UV-Vis analysis was performed to determine the maximum absorption wavelength of the catechin extract in a 70% ethanol solution [27]. Absorbance spectra were recorded from 200 to 400 nm. FTIR and UV-Vis results were compared with standard catechin data to confirm structure and purity. Figure 1 illustrates the catechin isolation and characterization process.

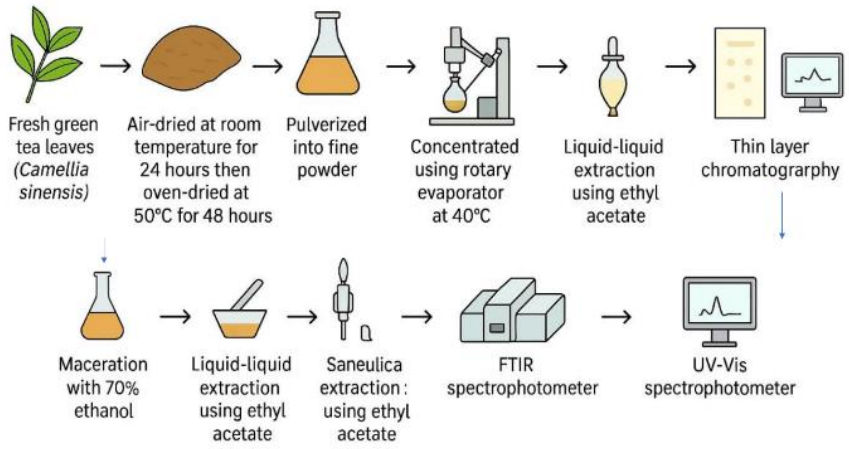


Figure 1. Catechin Isolation and Characterization Process

BTS 2 Leaching Process

Initial BTS 2 Characterization

BTS 2 samples were characterized using XRF to determine the composition of major and minor elements, particularly the levels of Ta, Nb, and other impurity metals, including Fe, Si, and Al. Analysis was performed on homogenized BTS 2 powder samples. Figure 2 shows the leaching process for Bangka Tin Slag 2.

Stage 1: Alkali-Acid Leaching

Ten grams of BTS 2 powder were placed into a leaching flask. Initial leaching was performed using 100 mL of 6 M sodium hydroxide (NaOH) solution at a solid-to-liquid ratio of 1:10 (g/mL) at 90°C for 2 hours with stirring at 300 rpm. After NaOH leaching, the suspension was filtered, and the solid residue was washed with deionized water until the pH reached neutral. The solid residue was then

p-ISSN 0854-1418
e-ISSN 2503-426X

reacted with 100 mL of 3.25 M hydrochloric acid (HCl) at 50°C and 300 rpm for 1 hour. After HCl leaching, the suspension was filtered again, and the solid residue was washed until neutral pH. The filtrates from both stages (NaOH and HCl) were collected and analyzed for their metal content.

Stage 2: Further Leaching with Hydrofluoric Acid

The solid residue from the alkali-acid leaching stage was then reacted with hydrofluoric acid (HF) solutions at concentrations of 2 M, 3 M, 4 M, 5 M, and 6 M. Five grams of solid residue were placed in an HF-resistant leaching flask (e.g., PTFE). Fifty mL of HF solution was added, yielding a solid-to-liquid ratio of 1:10 (g/mL). A small amount (5 mL) of 0.5 M sulfuric acid (H_2SO_4) was added to aid leaching and stabilize metal ions. The leaching process was carried out at 70°C for 3 hours with stirring at 300 rpm. After leaching, the suspension was cooled, filtered, and the filtrate (Pregnant Leach Solution, PLS) was collected. The solid residue was washed with deionized water. The concentrations of dissolved Ta, Nb, Fe, and other metals in the PLS were measured using MP-AES. Extraction efficiency was calculated as the ratio of metal concentration in the PLS to the initial content in BTS 2.

Catechin Interaction Study with Tantalum and Niobium

Determination of Complex Stoichiometric Ratio by Job's Method

Job's method (continuous variation method) was used to determine the stoichiometric ratio for the formation of complexes between catechin and Ta^{5+}, Nb^{5+}, and Fe^{3+} ions. Stock solutions of catechin (1 mM), Ta^{5+} (1 mM), Nb^{5+} (1 mM), and Fe^{3+} (1 mM) were prepared. In this method, the total concentration of catechin and metal ions was kept constant (e.g., 0.1 mM), while the molar ratio of catechin to metal ions was varied from 0:10 to 10:0. The absorbance of the solutions was measured using a UV-Vis spectrophotometer at the wavelength of maximum complex formation (e.g., 278 nm for catechin and specific wavelengths for metal-catechin complexes, if identified). Job's method curves were plotted as absorbance versus the catechin mole fraction to identify the molar ratio at which complex formation reached its maximum.

Catechin-Metal Complex Formation in Leached PLS

Interaction studies were conducted by adding catechin extract to the PLS solution from BTS 2 leaching (optimized at 4 M HF). The PLS solution was filtered, and its pH was adjusted to 2.0 using dilute NaOH or HCl. The initial concentrations of Ta, Nb, and other impurity metals in the PLS solution were measured using MP-AES. Then, a specified amount of catechin extract (e.g., 100 mg/L) was added to 50 mL of PLS solution, and the mixture was stirred at 200 rpm at room temperature for 2 hours. After the contact time, the solution was filtered, and the concentrations of Ta, Nb, and Fe in the filtrate were re-measured using MP-AES. Changes in these concentrations indicated the extent to which catechin interacted with and bound these metals. Characterization of the formed complexes was also performed using FTIR to confirm bonding between catechin and metals. Samples for FTIR were prepared by drying the complex precipitate formed after interaction.

Data Analysis

Data from MP-AES, UV-Vis, and FTIR were analyzed both quantitatively and qualitatively. Leaching efficiency was

calculated as the percentage of metal dissolved relative to the total metal in BTS 2. Catechin complexing efficiency was calculated as the percentage decrease in metal concentration in the solution after adding catechin. Comparing results before and after adding catechin was key to assessing catechin's potential as a complexing agent for extracting Ta and Nb from BTS 2. Descriptive statistics (mean, standard deviation) and inferential statistics (e.g., t-test for significant comparison) were performed using OriginPro 2023 software. All experiments were conducted in triplicate to ensure reproducibility, and results are presented as mean ± standard deviation.

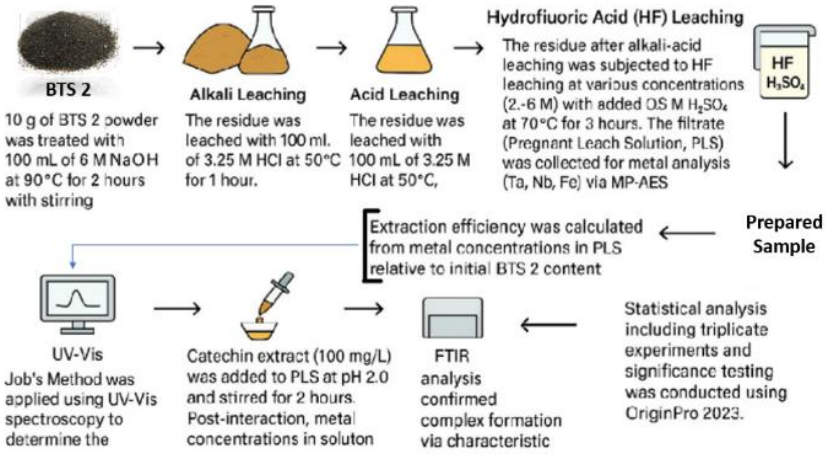


Figure 2. Process of BTS 2 leaching

RESULTS AND DISCUSSION

This section presents the results obtained from the series of experiments conducted, including catechin isolation and characterization, initial BTS 2 characterization, staged leaching process, and catechin interaction study with leached metals.

Isolation and Characterization of Catechin from Camellia sinensis Green Tea

Catechin Preparation and Isolation

Green tea leaves (*Camellia sinensis*) were dried and pulverized to obtain a powder. Catechin isolation from the green tea leaf powder yielded a brownish-yellow solid. The catechin yield from this isolation was 4.73% (w/w), consistent with previous reports [28]. Figure 3 shows the leaves of *Camellia sinensis*.

Figure 3. *Camellia Sinensis*

Phytochemical Screening Test and Thin Layer Chromatography (TLC)

The results of the phytochemical screening test showed that the catechin extract positively contained flavonoids, tannins, and saponins, indicating the presence of polyphenol compounds, including catechin. The TLC test showed a single main spot with an Rf value consistent with the catechin standard, confirming the presence of catechin in the isolated extract and indicating high purity. (Figure 4).

p-ISSN 0854-1418
e-ISSN 2503-426X

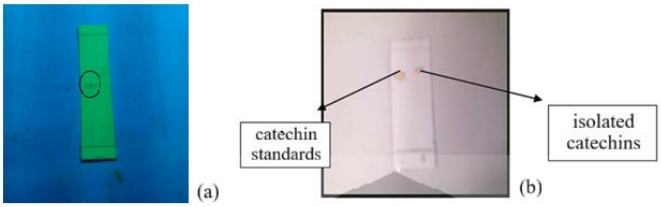
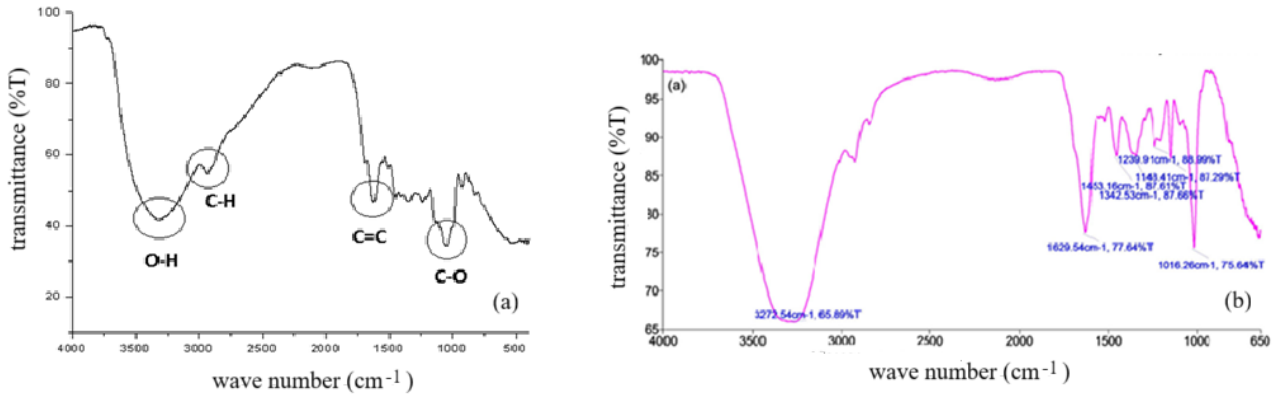


Figure 4. (a) TLC results (b) For Spot Visualization

Characterization of Catechin Extract with FTIR and UV-Vis

The FTIR spectrum of the isolated catechin extract showed characteristic absorption peaks for catechin functional groups. A broad peak at 3350 cm^{-1} indicated the stretching vibration of phenolic hydroxyl (-OH) groups, which is characteristic of catechin. Aromatic C=C stretching vibrations were observed at 1610 cm^{-1} and 1520 cm^{-1}, while C-O stretching vibrations from phenolic and secondary alcohol groups appeared at 1280 cm^{-1} and 1060 cm^{-1}. These peaks are consistent with the FTIR spectrum of standard catechin (Figure 5). UV-VIS analysis showed a maximum absorption wavelength of the catechin extract at 278 nm (Figure 6), which is characteristic of the aromatic ring chromophore absorption in the catechin structure, consistent with literature data.



(a) (b)

Figure 5. (a) FTIR Spectrum of Extracted Catechin (b) FTIR Spectrum of Standard Catechin

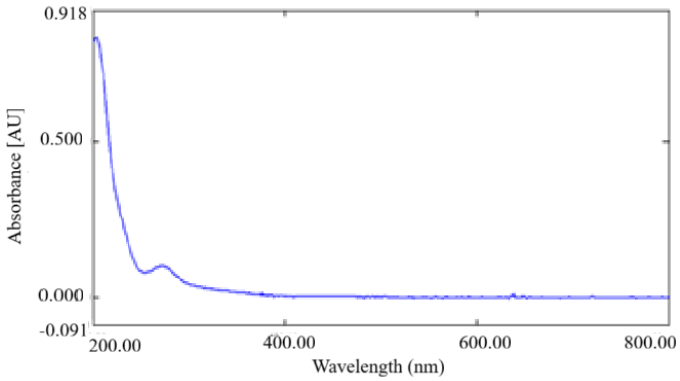


Figure 6. UV-Vis Spectrum of Extracted Catechin

Initial Characterization of BTS 2 Sample

XRF characterization results of the BTS 2 sample showed the composition of major elements in oxide forms. BTS 2 was dominated by quartz (SiO_2), rutile (TiO_2), hematite (Fe_2O_3), zirconium oxide (ZrO_2), aluminum oxide (Al_2O_3), and calcium oxide (CaO). The tantalum oxide (Ta_2O_5) content was 0.33%, and niobium oxide (Nb_2O_5) was 0.64%. In addition, the tin slag also contained significant amounts of major impurities such as iron (Fe). The chemical composition of BTS 2 is presented in Table 1.

Table 1. Chemical composition of BTS 2 based on XRF analysis (in %wt)

Compounds	Content (%)
Ta_2O_5	0.33
Nb_2O_5	0.64
SiO_2	34.26
CaO	15.44
TiO_2	11.92
Al_2O_3	11.7
Fe_2O_3	8.84
ZrO_2	4.78
Others	12.06

Leaching of BTS 2

The staged leaching process was carried out to dissolve Ta and Nb from BTS 2. Initial leaching with 6 M NaOH, followed by 3.25 M HCl at 50°C, was intended to remove most impurities. Further leaching with HF showed that HF concentration was a crucial factor in the dissolution efficiency of Ta and Nb. MP-AES measurements showed that the optimal HF concentration for dissolving Ta and Nb was 4 M (Table 2). At this concentration, the extraction efficiency of Ta and Nb reached its peak, although further increases in HF concentration did not significantly improve efficiency and could even dissolve more impurities. Table 2 presents the concentration of Ta_2O_5, Nb_2O_5, Fe_2O_3, CaO, and Al_2O_3 at various HF concentrations. The composition of the leachate solution (PLS) under optimum conditions (4 M HF) showed dissolved Ta and Nb concentrations, as well as significant Fe concentrations. This highlights the challenge of selectively separating Ta and Nb from iron impurities.

Table 2. Concentration of Ta_2O_5, Nb_2O_5, Fe_2O_3, CaO, Al_2O_3 at various HF concentrations

HF	Ta_2O_5	Nb_2O_5	Fe_2O_3	CaO	Al_2O_3
(M)			(ppm)		
1	0.122	0.881	43.376	6.194	6.192
3	1.032	10.040	73.25	6.863	7.969
4	3.879	23.109	99.879	6.976	9.889

Determination of Complex Stoichiometric Ratio by Job's Method

Job's method was used to determine the stoichiometric ratio of complex formation between catechin and metal ions (Ta^{5+}, Nb^{5+}, and Fe^{3+}). The results of Job's method showed that catechin tended to form a 1:1 stoichiometric complex with Fe^{3+}, indicated by a clear absorbance peak at a catechin mole fraction of 0.5 (Figure 7a). However, for Ta^{5+} and Nb^{5+}, the Job's method curves did not show clear peaks or significant absorbance changes (Figures 7b and 7c), indicating that catechin did not form stable or selective complexes with Ta and Nb under the tested conditions. The absorbance spectra also showed more pronounced changes upon the addition of Fe^{3+} than those of Ta^{5+} and Nb^{5+}, supporting this finding (Figures 8a, 8b, and 8c).

It can be analyzed based on Figure 6 that fraction 0.5 has an optimum absorbance value at a maximum wavelength of 236 nm, with a volume ratio of Ta^{5+}: Ligand, namely 1:1. So the formula of the complex compound obtained is $[TaL]^{5+}$.

p-ISSN 0854-1418
e-ISSN 2503-426X

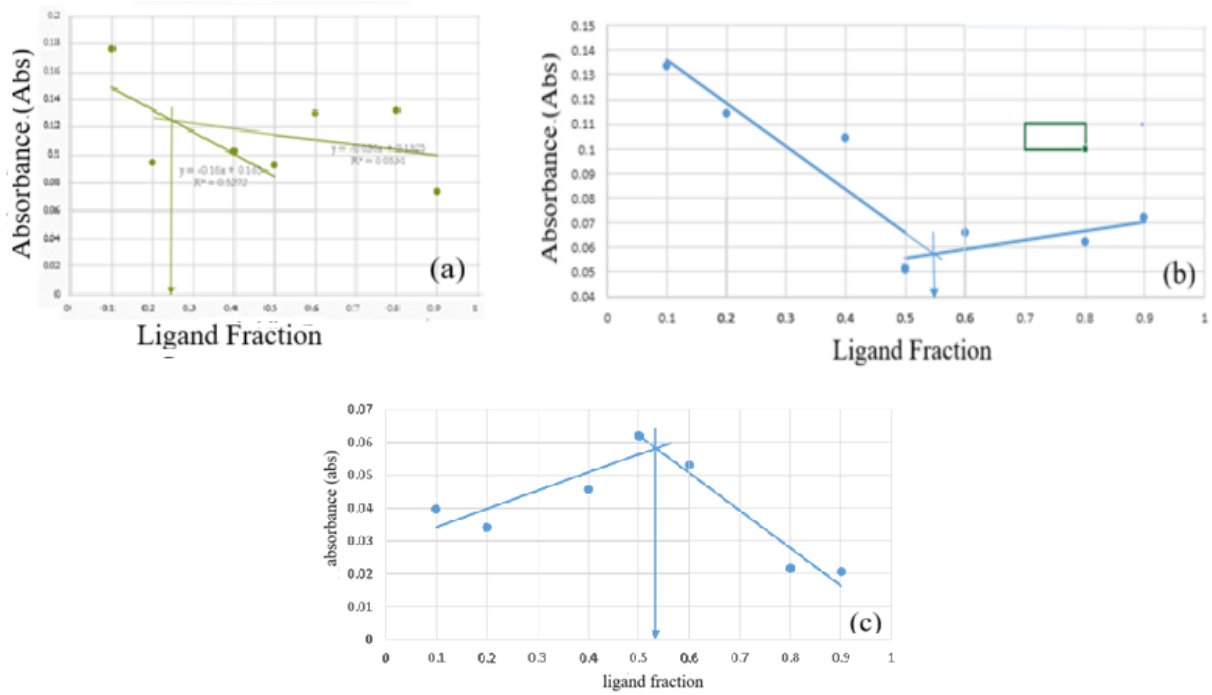


Figure 7. (a) Job's method curve of ligand fraction vs. Fe^{3+}; (b) Job's method curve of ligand fraction vs. Ta^{5+}; (c) Job's method curve of ligand fraction vs. Nb^{5+}

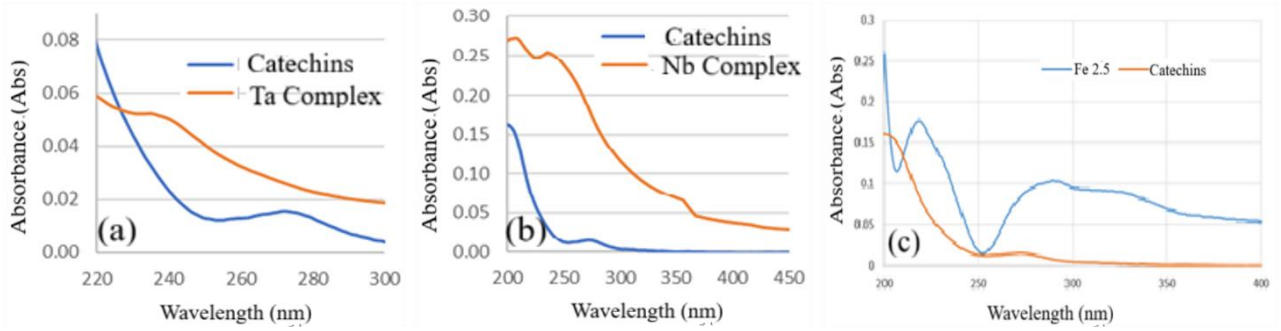


Figure 8. Absorbance spectrum of catechin ligands and the addition of 1 equivalent; (a) Ta^{5+}, (b) Nb^{5+}, and (c) Fe^{3+}

Catechin Interaction Study with Tantalum and Niobium in Leached PLS

The catechin interaction study with the leached PLS solution (from 4 M HF leaching) showed that catechin addition was more effective in binding iron than tantalum and niobium. The iron content in the PLS solution decreased significantly after the addition of catechin, indicating high Fe_2O_3 complexing efficiency. Conversely, the concentrations of tantalum and niobium in the solution did not change significantly after catechin addition, indicating low or non-selective complexing affinity for these two metals. Table 3 presents the metal concentrations in PLS before and after catechin addition, as well as complexing efficiency.

Table 3. Metal concentration in PLS before and after catechin addition

Compounds	Content Before Catechin Addition (ppm)	Content After Catechin Addition (ppm)
Ta_2O_5	3.879	3.819
Nb_2O_5	23.109	23.060
Fe_2O_3	99.879	87.665
CaO	6.976	6.398
Al_2O_3	9.889	9.079

FTIR analysis of the catechin-metal complexes formed after interaction with PLS also supported these findings. The FTIR spectra showed more significant shifts in characteristic catechin peaks (especially -OH groups) when interacting with Fe compared to Ta or Nb (Figures 9a and 9b). This collectively indicates that catechin has a higher complexing affinity for Fe than for Ta and Nb in the complex BTS 2 matrix.

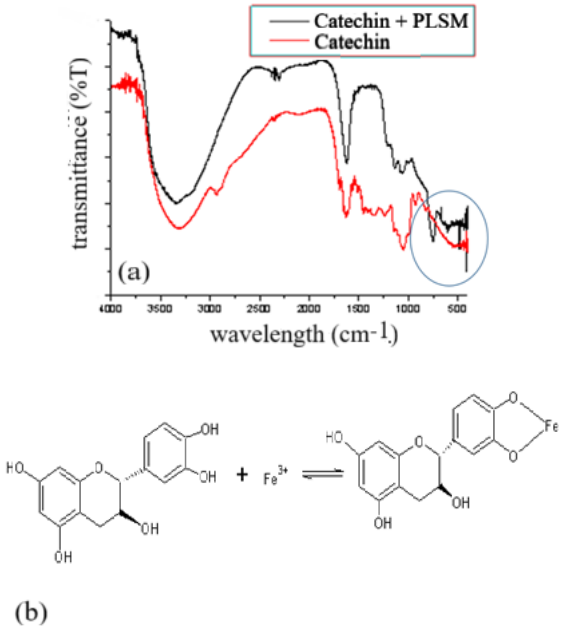


Figure 9. (a) FTIR of Catechin with Catechin – PLSM; (b) The suspected catechin-iron complex formed

This study comprehensively explores the potential of catechin extract as a complexing agent for the recovery of tantalum and niobium from BTS 2. The results provide important insights into catechin-metal interactions within the complex tin slag matrix and highlight the challenges and opportunities in bio-agent-based extractive metallurgy applications [29], [30].

Initial characterization of BTS 2 confirmed the presence of Ta and Nb at significant concentrations (0.33% Ta_2O_5 and 0.64% Nb2O5), consistent with previous reports identifying tin slag as a promising secondary source for these strategic metals [8], [31]. The presence of substantial impurities, especially iron (Fe), was also confirmed. Iron is a common feature of tin slag and poses a major challenge in separating Ta and Nb due to their similar chemical properties and tendency to co-dissolve during leaching [2], [19], [32].

The staged leaching process, starting with alkali-acid treatment (NaOH-HCl) and continuing with hydrofluoric acid (HF), is a standard hydrometallurgical approach effective for dissolving Ta and Nb from oxide matrices [16], [17]. The use of HF is crucial because Ta and Nb form stable, water-soluble fluoride complexes, allowing their separation from the solid matrix. Optimizing the HF concentration to 4 M yielded good dissolution efficiency for Ta and Nb. However, as indicated by the leaching results, impurity metals such as Fe also dissolved significantly. This underscores the need for an efficient, selective separation stage after leaching to obtain high-purity Ta and Nb products.

A key aspect of this study is the evaluation of catechin as a complexing agent. Catechin, a flavonoid with abundant phenolic hydroxyl groups, can theoretically interact with metal ions via chelation [24]. Studies have shown that catechin forms complexes with various metal ions, including Cu^{2+} and Zn^{2+}, which affect catechin's stability and biological activity [20], [29]. However, catechin's interactions with Ta^{5+} and Nb^{5+}, particularly in the context of selective extraction from complex solutions, have been rarely studied and represent a knowledge gap that this research aims to address. This gap is important given catechin's potential as a 'green' complexing agent that can reduce dependence on less environmentally friendly synthetic reagents.

p-ISSN 0854-1418
e-ISSN 2503-426X

The results of Job's method in this study clearly indicate that catechin does not form stable or selective complexes with Ta^{5+} and Nb^{5+} under the tested conditions. The Job's method curves for Ta and Nb did not show clear peaks, in stark contrast to the interaction of catechin with Fe^{3+}, which resulted in a strong, clear 1:1 stoichiometric ratio. This finding is reinforced by FTIR and MP-AES analyses, which showed that catechin tended to bind more with iron in the leachate solution, while the concentrations of Ta and Nb did not change much after catechin addition. The higher affinity of catechin for Fe can be explained by several factors. Iron, especially in the form of Fe^{3+}, is a transition metal ion known to have a strong tendency to form complexes with ligands containing oxygen donor atoms, such as the phenolic hydroxyl groups in catechin [30]. In addition, differences in the ion size, charge, and electronic configuration of Ta^{5+} and Nb^{5+} relative to Fe^{3+} may affect catechin's ability to coordinate effectively and form stable complexes with these ions. Ta and Nb, as period 5 transition metals, tend to form complexes with ligands that have 'harder' donor atoms or with more specific structures that may not be entirely compatible with the catechin structure.

These findings have important implications for the development of bio-agent-based extractive metallurgical processes. First, although catechin is a promising natural complexing agent, its selectivity for target metals (Ta and Nb) in complex matrices such as tin slag must be carefully evaluated. High levels of impurities, such as iron, can significantly interfere with selective extraction. This suggests that for Ta and Nb extraction, catechin may require structural modification (e.g., functionalization to increase selectivity) or be used in combination with other separation methods.

CONCLUSION

This study investigated the potential of catechin extract from Camellia sinensis as a complexing agent for the recovery of tantalum (Ta) and niobium (Nb) from Bangka Tin Slag 2 (BTS 2) after a staged leaching process. The findings indicate that while catechin exhibits a strong complexing affinity for iron (Fe), a major impurity in BTS 2, its selectivity for Ta and Nb is limited under the conditions tested. Job's method and FTIR analysis consistently showed that catechin preferentially forms complexes with Fe^{3+} rather than Ta^{5+} or Nb^{5+}. This suggests that, for effective and selective extraction of Ta and Nb from complex matrices such as tin slag, catechin may need to be structurally modified to enhance its selectivity or integrated into a multi-stage separation process alongside other methods.

Based on the insights gained from this preliminary investigation, several specific recommendations are proposed for future work to overcome the current limitations of catechin selectivity:

1. Structural Modification of Catechin: Future studies should focus on the chemical functionalization or structural modification of the catechin core (e.g., introducing specific electron-withdrawing or bulky hydrophobic groups) to deliberately enhance its steric and electronic selectivity toward highly charged $Ta^{5+}$and Nb^{5+} ions over Fe^{3+}.
2. Investigation of Alternative pH Conditions: The extraction system should be evaluated across a broader pH range, as optimizing the hydrogen ion concentration can heavily

influence the protonation state of catechin's phenolic hydroxyl groups and the speciation of the target metals, potentially opening a window for selective binding.

3. Removal of Fluoride Prior to Complexation: It is highly recommended to integrate an intermediate fluoride-removal or masking step before introducing the bio-ligand, thereby breaking down the hyper-stable fluoro-complexes ($(TaF_7)^{2-}$ and $(NbF_6)^-$) that currently restrict ligand exchange with catechin.
4. Comparison with Other Natural Ligands: A comparative screening with alternative natural polyphenolic ligands, such as quercetin, tannic acid, or gallic acid, should be conducted to evaluate whether different spatial configurations of oxygen-donor groups offer superior coordination efficiency and selectivity for refractory metal recovery.

ACKNOWLEDGMENT

The authors would like to express their gratitude to Universitas Indonesia and the National Research and Innovation Agency (BRIN) for providing the necessary research facilities, laboratory infrastructure, and technical assistance throughout this study. We also thank all colleagues who provided constructive insights during the preparation of this manuscript.

REFERENCES

[1] OECD, *OECD Economic Surveys: Indonesia 2012*, OECD Publishing, Paris, 2012, doi: https://doi.org/10.1787/eco_surveys-idn-2012-en.

[2] I. Gaballah, E. Allain, M.-CH. Meyer-Joly, and K. Malau, "A Possible Method for the Characterization of Amorphous Slags Recovery of Refractory Metal Oxides from Tin Slags", *Metall. Trans. B.*, vol. 23, pp. 249–259, 1992, doi: https://doi.org/10.1007/BF02656280.

[3] E. Zunianto and A. Adhiutama, "Analysis of Material Handling Flow Process on Tin Smelting: a Case Study in Indonesia," *Jurnal Aplikasi Manajemen*, vol. 11, no. 3, pp. 407–416, 2013.

[4] E. Allain, N. Kanari, F. Diot, and J. Yvon, "Development of a process for the concentration of the strategic tantalum and niobium oxides from tin slags," *Minerals Engineering*, vol. 134, pp. 185-192, 2019, doi: https://doi.org/10.1016/j.mineng.2019.01.029.

[5] M. G. Haile, A. Oladunni Oyelola, F. Olufemi Aramide, S. Ojo Seidu, and O. Akinlabi, "Hydrometallurgical, pyrometallurgical and electrometallurgical extraction of niobium and tantalum: an overview," *Mineral Processing and Extractive Metallurgy: Transactions of the Institute of Mining and Metallurgy*, vol. 134, no. 1, pp. 3–12, Mar. 2025, doi: https://doi.org/10.1177/25726641241301982.

[6] U.S. Geological Survey, *Mineral commodity summaries 2024: U.S. Geological Survey*, p. 212, 2024, doi: https://doi.org/10.3133/mcs2024.

[7] M. G. Haile, A. O. Oyelola, and A. Akcil, "Pyrometallurgical and electrometallurgical extraction of niobium and tantalum: an overview. Mineral Processing and Extractive Metallurgy," *Hydrometallurgical*, vol. 134, no. 1, pp. 3–12, Mar. 2025, doi: https://doi.org/10.1177/25726641241301982.

[8] J. M. de Oliveira, I. A. Anes, J. L. Coleti, D. C. R. Espinosa, M. S. de Carvalho, and J. A. S. Tenorio, "Niobium and tantalum recovery from the primary source and from tin slag, an industrial challenge: A review," *Can. J. Chem. Eng.*, vol. 101, no. 4, pp. 1743–1761, 2023, doi: https://doi.org/10.1002/cjce.24621.

p-ISSN 0854-1418
e-ISSN 2503-426X

[9] TIC, "Processing: extraction and refining," 2025. [Online]. Available: https://tanb.org/about-tantalum/processing-extraction-and-refining/.

[10] M. Iguchi, T. Nakatani, and H. Tokunaga, "The Shape of Bubbles Rising near the Nozzle Exit in Molten Metal Baths," *Metallurgical and Materials Transactions B*, vol. 28, pp. 417–423, 1997, doi: https://doi.org/10.1007/s11663-997-0107-2.

[11] F. A. López, I. García-Díaz, O. R. Largo, F. G. Polonio, and T. Llorens, "Recovery and purification of tin from tailings from the Penouta Sn-Ta-Nb deposit," *Minerals*, vol. 8, no. 1, Jan. 2018, doi: https://doi.org/10.3390/min8010020.

[12] J. Spooren, W. Wouters, B. Michielsen, E. M. Seftel, and S.-F. Koelewijn, "Microwave-assisted alkaline fusion followed by water-leaching for the selective extraction of the refractory metals tungsten, niobium and tantalum from low-grade ores and tailings," *Miner. Eng.*, vol. 217, p. 108963, 2024, doi: https://doi.org/10.1016/j.mineng.2024.108963.

[13] C. Subramanian and A. Suri, "Recovery of niobium and tantalum from low grade tin slag-A hydrometallurgical approach," Environmental and Waste Management, pp. 100–107, 1998, [Online]. Available: https://eprints.nmlindia.org/2842/1/100-107.PDF.

[14] E. A. Brocchi and F. J. Moura, "Chlorination methods applied to recover refractory metals from tin slags," *Miner. Eng.*, vol. 21, no. 2, pp. 150–156, Jan. 2008, doi: https://doi.org/10.1016/j.mineng.2007.08.011.

[15] D. M. C. Machaca, T. C. de Carvalho, J. A. S. Tenório, and D. C. R. Espinosa, "Advancements in the extraction of niobium and tantalum: Innovative strategies in hydrometallurgical processes," *Miner. Eng.*, vol. 222, 2025, doi: https://doi.org/10.1016/j.mineng.2024.109125.

[16] A. Shikika, M. Sethurajan, F. Muvundja, M. C. Mugumaoderha, and S. Gaydardzhiev, "A review on extractive metallurgy of tantalum and niobium," *Hydrometallurgy*, vol. 198, Dec. 2020, doi: https://doi.org/10.1016/j.hydromet.2020.105496

[17] O. Rodríguez, F. J. Alguacil, E. E. Baquero, I. García-Díaz, P. Fernández, B. Sotillo, and F. A. López, "Recovery of niobium and tantalum by solvent extraction from Sn-Ta-Nb mining tailings," *RSC Adv.*, vol. 10, no. 36, pp. 21406–21412, Jun. 2020, doi: https://doi.org/10.1039/d0ra03331f.

[18] M. Nete, W. Purcell, and J. T. Nel, "Separation and isolation of tantalum and niobium from tantalite using solvent extraction and ion exchange," *Hydrometallurgy*, vol. 149, pp. 31–40, 2014, doi: https://doi.org/10.1016/j.hydromet.2014.06.006.

[19] O. S. Ayanda and F. A. Adekola, "A Review of Niobium-Tantalum Separation in *Hydrometallurgy*," Journal of Minerals & Materials Characterization & Engineering, vol. 10, no. 3, pp. 245–256, 2011, doi: https://doi.org/10.4236/jmmce.2011.103016.

[20] M. Nete and W. Purcell, "Separation and Purification of Niobium and Tantalum from Synthetic and Natural Compounds," *Doctoral dissertation*, University of the Free State, South Africa, 2013.

[21] F. Wulaningsih, "Isolasi Katekin dari Daun Teh Hijau (Camellia sinensis) dengan Metode Maserasi dan Ekstraksi," *Unpublished undergraduate thesis*, Universitas Indonesia, 2008.

[22] M. Kumamoto, T. Sonda, K. Nagayama, and M. Tabata, "Effects of pH and metal ions on antioxidative activities of catechins," *Biosci. Biotechnol. Biochem.*, vol. 65, no. 1, pp. 126–132, 2001, doi: https://doi.org/10.1271/bbb.65.126.

[23] D. Botten, G. Fugallo, F. Fraternali, and C. Molteni, "Structural Properties of Green Tea Catechins," *Journal of Physical Chemistry B*, vol. 119, no. 40, pp. 12860–12867, Oct. 2015, doi: https://doi.org/10.1021/acs.jpcb.5b08737.

[24] K. M. Bark, J. E. Yeom, I. J. Yang, O. H. Park, C. H. Park, and H. R. Park, "Studies on the interaction between catechin and metal ions," *Bull. Korean Chem. Soc.*, vol. 33, no. 12, pp. 4235–4238, Dec. 2012, doi: https://doi.org/10.5012/bkcs.2012.33.12.4235.

[25] I. I. Lungu, D. Marin-Batîr, A. Panainte, C. Mircea, C. Tuchiluș, A. Ștefanache, F. A. Szasz, D. Grigorie, S. Robu, O. Cioancă, and M. Hăncianu, "Catechin-Zinc-Complex: Synthesis, Characterization and Biological Activity Assessment," *Farmacia*, vol. 71, no. 4, pp. 755–763, 2023, doi: https://doi.org/10.31925/farmacia.2023.4.11.

[26] I. I. Lungu, O. Cioancă, C. Mircea, C. Tuchiluș, A. Ștefanache, R. Huzum, and M. Hăncianu, "Insights into Catechin–Copper Complex Structure and Biologic Activity Modulation," *Molecules*, vol. 29, no. 20, Oct. 2024, doi: https://doi.org/10.3390/molecules29204969.

[27] D. L. Pavia, G. M. Lampman, G. S. Kriz, and J. R. Vyvyan, *Introduction to Spectroscopy (5th ed.)*, Cengage Learning, 2015. W. Nofriandani, "Studi Pendahuluan Interaksi Ekstrak Katekin (Camellia sinensis) dengan Tantalum dan Niobium Hasil Leaching Bertahap TTB 2," *Unpublished undergraduate thesis*. Universitas Indonesia, 2017.

[28] H. Li, Z. Zhang, J. Chen, M. Sun, and H. Tang, "Effect of Cu2+ on the binding of catechins to zein through multi-spectral and in silico analyses," Food Chem., vol. 463, 2025, doi: https://doi.org/10.1016/j.foodchem.2024.141547

[29] F. A. Cotton, G. Wilkinson, C. A. Murillo, and M. Bochmann, *Advanced Inorganic Chemistry (6th ed.)*, John Wiley & Sons., 1999.

[30] S. Permana, J. W. Soedarsono, A. Rustandi, and A. Maksum, "Other Oxides Pre-removed from Bangka Tin Slag to Produce a High Grade Tantalum and Niobium Oxides Concentrate," in *IOP Conference Series: Materials Science and Engineering*, Institute of Physics Publishing, Jun. 2016, doi: https://doi.org/10.1088/1757-899X/131/1/012006.

[31] J. Meija, T. B. Coplen, M. Berglund, W. A. Brand, P. De Bièvre, M. Gröning, N. E. Holden, J. Irrgeher, R. D. Loss, T. Walczyk, and T. Prohaska, "Atomic weights of the elements 2013 (IUPAC Technical Report)," *Pure and Applied Chemistry*, vol. 88, no. 3, pp. 265–291, 2016, doi: https://doi.org/10.1515/pac-2015-0305.