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SEPARATION OF NIOBIUM AND TANTALUM - A LITERATURE SURVEY

by

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During recent years increased interest has been shown in the metal niobium. Because niobium has a high melting point (4376° F) and a low neutron cross-section, the Atomic Energy Commission has encouraged research on new methods of winning the metal from its ores with the ultimate hope of finding a process which would yield niobium metal at a reasonable price. The two areas of endeavor in which the principal difficulties in finding such an economic process occur are the separation of tantalum from the niobium and the conversion of purified niobium salts to the metal. This report, then, was written to consolidate the more important information available in the literature on one of these troublesome areas, that of the separation of niobium and tantalum. It is to be noted, however, that all the references available in the literature on niobium-tantalum separation are not included in this report, but only those which appear to have the most usefulness for future endeavors on this problem.

## HISTORY

As was the case with every pair of chemically similar elements, the early history of niobium is really that of the niobium-tantalum earth-acid group. Since niobium-bearing minerals without tantalum are virtually unknown, the first investigators were dealing with mixtures of these two chemically similar elements. The presence of titanium in some cases caused additional complications. Before the final identification of the two elements, seven names had been ascribed to new elements that were thought to be present in these minerals.

Hatchett in 1801 is credited with the first identification of niobium in a heavy black mineral from the British Museum which had been sent from Connecticut a number of years previously. He named it columbium in honor of the country from which the ore came. Ekeberg in 1802 found a new element in some minerals now called tantalite and yttrotantalite. The insolubility of the oxide reminded him of the fable of Tantalus and he named the element tantalum. In 1844 Rose studied a columbite and concluded that two elements were present. He took one of these to be Ekeberg's tantalum and the other he named niobium (from Niobe, the daughter of Tantalus). Later he announced the presence of a third element which he called pelopium (from Pelops, brother of Niobe). Hermann added the names ilminium (from the Ilmen Mountains) and neptunium (from Neptune) while von Kobell completed the list with dianium (from Diana) (1).

Marignac (2) finally identified the two elements in 1866 by separating them as complex potassium fluorides by fractional crystallization. He found that the complex niobium fluoride was isomorphous with the known salts  $K_2TiF_6 \cdot H_2O$  and  $K_2WO_2F_4 \cdot H_2O$ , which indicated that the valence of the niobium was + 5 and the formula of the salt was  $K_2NbOF_5 \cdot H_2O$ .

## OCCURRENCE

The most important source of tantalum and niobium is a ferrous columbate-tantalate,  $Fe(Nb - TaO_3)_2$ . When niobium predominates, the mineral is called columbite, and when the tantalum predominates, tantalite. The iron may be partially replaced by manganese. Columbite and tantalite are heavy minerals, the specific gravity varying from 5.3 to 7.3 with increased tantalum content. They are usually associated with pegmatite dikes. The most important niobium-bearing minerals are listed in Table 1 (1).

During World War II, the world reserves were estimated as equivalent to 15,000,000 pounds of contained niobium and tantalum. Present known reserves are of the order of ten times this figure in columbites and one hundred times including pyrochlores. Pyrochlore is a mineral of the type  $NaCaNb_2O_6F$ , but K, Mg, Fe, Si, Sn, Ta, Ce, V, U, Th, and P may also be present.

The sources of columbite ore are, for the most part, foreign, Nigeria being the most important source. However, large quantities of lower grade ores are known to exist on the North American continent. Table 2 describes most of the known deposits of niobium (3).

## SEPARATION OF NIOBIUM AND TANTALUM

The present commercial method for the opening of columbite-tantalite ores involves a basic fusion with molten caustic at  $650^\circ C$ . In this process the pulverized ore is fused with caustic soda which converts the tantalum and niobium content of the mineral partly into the form of sodium tantalate and niobate, and partly into iron tantalate and niobate. Unlike the mineral itself, iron tantalate and niobate are readily decomposed by mineral acids. Sodium tantalate and niobate are practically insoluble in water containing free alkali, so that, when

Table 1. Niobium-Bearing Minerals\*

Name	Composition	Per cent		Specific gravity	Moh's hardness
		Nb <sub>2</sub> O <sub>5</sub>	Ta <sub>2</sub> O <sub>5</sub>		
<b>Series:</b>					
Columbite (theory)	FeNb <sub>2</sub> O <sub>6</sub>	78.72	--		
Tantalite (theory)	FeTa <sub>2</sub> O <sub>6</sub>	--	86.01		
Columbite	(Fe, Mn) (Nb, Ta) <sub>2</sub> O <sub>6</sub>	26-78	1-48	5.1-6.8	6
Tantalite	(Fe, Mn) (Ta, Nb) <sub>2</sub> O <sub>6</sub>	2-40	42-84	6-7.8	6-6.5
<b>Series:</b>					
Manganocolumbite	(Mn, Fe) (Nb, Ta) <sub>2</sub> O <sub>6</sub>	30-75	5-40	5.2-6.4	6
Manganotantalite	(Mn, Fe) (Ta, Nb) <sub>2</sub> O <sub>6</sub>	4-35	35-82	6-7.8	6-6.5
<b>Stibiocolumbite-stibiotantalite series</b>					
	(Sb, Bi) (Nb, Ta)O <sub>4</sub>	1-48	1-60	5.5-7.5	6-7.3
Pyrochlore	NaCaNb <sub>2</sub> O <sub>6</sub> F	30-64	1-33	4.2-4.4	5-5.5
Yttrotantalite	(Fe, Y, U, Ca) (Nb, Ta, Zn, Sn)O <sub>4</sub>	1-20	37-56	5.5-6.8	5-5.5
Fergusonite	(Y, Er, Ce, Fe) (Nb, Ta, Ti)O <sub>4</sub>	14-46	4-43	5.6-5.8	5.5-6.5
Samarskite	(Y, Er, Ce, U, Fe, Th) (Nb, Ta, Ti) <sub>2</sub> O <sub>6</sub>	27-47	2-27	5.6-5.8	5-6
Eschynite	(Ce, Ca, Fe, Th) (Ti, Nb) <sub>2</sub> O <sub>6</sub>	23-37	?-7	4.9-5.1	5-6
<b>Series:</b>					
Euxenite	(Y, Ca, Ce, U, Th) (Nb, Ta, Ti) <sub>2</sub> O <sub>6</sub>	15-41	1-6	4.7-5	5.5-6.5
Polycrase	(Y, Ca, Ce, U, Th) (Ti, Nb, Ta) <sub>2</sub> O <sub>6</sub>	4-20	?-14	4.7-5.9	5.5-6.5

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Table 2. Location and Estimated Reserves of Niobium Deposits

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Location	Ore Tonnage	Per cent Nb <sub>2</sub> O <sub>5</sub>	Tons Nb <sub>2</sub> O <sub>5</sub>	Remarks
<u>COLUMBITE</u>				
Nigeria	--	--	57,000 proved 13,000 indicated	Partly by-product from tin operations.
Idaho	--	--	8,000 <del>0</del>	Also contains euxenite, samarskite, and fergusonite.
<u>PYROCHLORE</u>				
Africa				
Tanganyika	10,000,000	0.3	30,000	Probably reserves much larger.
Kenya	30,000,000	0.7	210,000	Also contains 3.1 per cent rare earth oxides.
Uganda	200,000,000	0.3	600,000	Also contains 0.8 per cent zircon and 14 per cent P <sub>2</sub> O <sub>5</sub> .
No. Rhodesia	--	--	--	Pyrochlore prospect.
Uganda	--	--	--	Pyrochlore prospect.
Nigeria	140,000,000	0.26	364,000	Pyrochlore in unweathered granite.
Brazil	Possibly very large	2.0	--	Pyrochlore, euxenite and samarskite.
Canada				
Newman Island	5-10,000,000	0.5	25-50,000	Underground mining beneath lake.
Nemegos, Ontario	32,000,000	0.246	78,700	Reserves probably can be enlarged, koppite, pyrochlore and perovskite.
Oka, Quebec	10,000,000 present 100,000,000 ultimate	0.2-0.3 --	200-300,000 --	Pyrochlore and perovskite. Open pit mining.

the fusion mass is treated with water, a large portion of the free alkali is removed by decantation. The mixed iron and sodium tantalates and niobates are finally removed from the fusion extract by filtration and are digested with hot hydrochloric acid which leaves a fairly white mixture of the earth acids. This precipitate is thoroughly washed with water to remove all iron and other soluble impurities (4).

Several investigators have reviewed the various methods for separating niobium and tantalum salts up to about 1945 (5, 6, 7). The process developed by Marignac (2) in 1866, with some variations, is the only method used commercially. In the process niobic and tantalic acids are dissolved in a minimum amount of hydrofluoric acid. The solution is saturated with potassium fluoride, evaporated and cooled. The tantalum is removed as potassium fluotantalate crystals,  $K_2TaF_7$ . After further evaporating and cooling, the niobium is removed as potassium pentafluoniobate crystals,  $K_2NbOF_5 \cdot H_2O$ . This separation is based on the difference in solubility in water of the two double potassium salts. One gram of the tantalum salt dissolves in 150 grams of water while an equal weight of the niobium salt requires only 12 parts water.

A method presented by Schoeller (6) makes use of the different extents to which tantalum and niobium oxalates are affected by hydrolysis. Tannin precipitates the tantalum complex from a slightly acid solution while an excess of tannin precipitates niobium from a neutral solution. This method is commonly used for analytical procedures and it does not appear to be practical for a large scale operation.

Besides the sodium hydroxide fusion mentioned earlier, many other alkaline and acidic fluxes are used to open the ore for analytical determinations. Schoeller (6) has discussed these fluxes in great detail. For analytical determinations Schoeller recommended the use of an acid flux, potassium bisulfate. Alkaline fluxes were rated undesirable because of unavoidable contamination of the melt with the material of the crucible. From an economic standpoint these fluxes are undesirable for commercial operation because of their cost and the high weight ratio of flux to ore (10-20:1) to effect a good recovery. Pierret (8) had adequately reviewed the various fusion processes attempted for the opening of the ore.

Generally the actual separation of tantalum and niobium has been accomplished after most of the impurities have been removed by a fusion and chemical processing procedure. Several investigators have tried separations without involving a fusion step. Jenness (9) obtained tantalum and niobium from ore by chloridizing each metallic element in the ore and raising the temperature to the volatilization point of each chloride.

May, Henderson and Johansen (10) described a method for separating tantalum and niobium by selective chlorination which followed a heat treatment of partially hydrolyzed tantalum and niobium chlorides with ammonia or ammonium chloride. This method was applied only to pure mixtures of the oxides. Separation was not quantitative and, depending upon conditions of hydrolysis, only one purified component of the mixture was obtained. Hiskey and co-workers (11) volatilized niobium and tantalum

by treating the oxide mixture with a chlorinated hydrocarbon. This method was not applicable to ores as the presence of iron caused catalytic decomposition of the chlorinating agent. Recently, however, it was reported (12) that the addition of two to four per cent sodium chloride promotes the formation of stable nonvolatile ferric chloride complexes during the chlorination of niobium-tantalum ores. This particular chlorination method was proven on Western black sand deposits by the Bureau of Mines. The chlorination charge is heated for one hour at  $500^{\circ}\text{C}$  after which titanium tetrachloride first distills, then niobium and tantalum chlorides. These latter are condensed separately and separated by selective hydrolysis as reported above (10). Iron carryover is cut from 60 per cent to practically nil.

Curvelliez (13) patented a process whereby niobium was separated from tantalum by roasting the ore in air at  $800$  to  $1200^{\circ}\text{C}$  with a basic substance which was usually calcium oxide. The residue from the roasting process was heated in a chlorine atmosphere at  $800$  to  $1050^{\circ}\text{C}$  for several hours. Seventy to 80 per cent of the niobium was volatilized as a chloride-oxychloride mixture while practically all the tantalum remained in the residue.

Line and Ingles (14) reported 99.8 per cent recovery of niobium from niobium ores by chlorination with chlorine gas. Either niobium oxychloride or pentachloride was recovered, depending upon the conditions of the reaction. The niobium produced was free from the gangue materials, iron and manganese. No report was made on tantalum.

Ruff and Thomas (15) reported using carbon tetrachloride as a chlorinating agent in the treatment of tantalum and niobium oxides. At  $200$  to  $225^{\circ}\text{C}$  niobium pentachloride was formed, while the tantalum oxide remained unaffected. No sharp separation occurred, however, because in a secondary reaction, niobium pentachloride reacted with tantalum oxide to give niobium oxychloride and tantalum pentachloride.

Kroll and Bacon (16,17) also patented a process for selective chlorination. More niobium oxide than tantalum oxide formed a nitride when the mixture was treated with ammonia at  $600^{\circ}\text{C}$ . When chlorine was passed over this partially nitrided mixture at  $500^{\circ}\text{C}$  niobium was fractionally distilled. In one test 88 per cent of the niobium and less than 5 per cent of the tantalum were removed from a mixture initially composed of 47 per cent niobium oxide and 53 per cent tantalum oxide. Other patents (18,19) suggested treatment of the mixed oxides with hydrogen to reduce selectively the niobium pentoxide to the tetroxide. After this partial reduction, the charge was treated with chlorine gas to distill niobium pentachloride. Only about 20 per cent of the niobium was removed in one cycle.



A French patent (20) described a process for separating tantalum and niobium. Mixed oxides were added to a fused alloy containing tantalum and niobium. Tantalum was displaced with respect to niobium, producing a slag containing tantalum and an alloy containing niobium.

Golibersuch and Young (21) precipitated niobium containing less than 0.2 per cent tantalum by electrolytic reduction of quinquevalent niobium to the quadri- and trivalent states in a 75 per cent sulfuric acid solution. In a similar process (22) niobium was reduced in acid solution with zinc dust. Boiling precipitated the tantalum hydrate, leaving the lower valent niobium in solution. The niobium was then reoxidized and precipitated by boiling.

Yntema (23) reported that an incomplete separation could be effected by electrolytic hydrolysis with different hydrogen ion concentrations. Later investigations by Pierce and Yntema (24) indicated that it was impossible to effect a clean separation by merely regulating the pH. Later, Pierce (25) claimed that niobium could be plated from a saturated solution of ignited niobium oxide in sodium carbonate. A similar solution of tantalum oxide would not plate out tantalum under identical conditions.

Anion-exchange methods have been found effective in separating tantalum and niobium. One method employing a mixed hydrochloric-hydrofluoric acid solution of niobium and tantalum indicated that tantalum was preferentially adsorbed (26,27). Huffman and co-workers (28) found niobium preferentially adsorbed from hydrochloric acid solutions. The niobium and tantalum obtained by these ion-exchange techniques were better than 99 per cent pure, relative to each other. Gillis and co-workers (29) recovered 95 per cent of the niobium free of tantalum by one pass of their oxalic acid solution containing equal weights of the two elements.

Several successful chromatographic separations of niobium and tantalum have been reported. Burstall and associates (30,31) charged cellulose with a fluoride solution of niobium and tantalum and then elutriated this with methyl ethyl ketone. The tantalum was quantitatively transferred to the ketone and was found to contain as little as 0.1 per cent niobium. Addition of hydrofluoric acid to the eluant was required to remove the niobium from the cellulose. Tikhomiroff (32) accomplished this same separation by adsorption on activated alumina from an ammonium oxalate solution. Wood (33) added ammonium ion to the fluoride solutions of niobium and tantalum poisoned with a little titanium. Adsorption on cellulose followed by elutriation showed the ammonium ion completely suppressed the transfer of the titanium. This is a significant factor in the preparation of pure niobium as titanium usually associates itself with the niobium.

Recently, separations have been achieved by liquid-liquid extraction methods. Leddicotte and Moore (34) found that a solution of methyldioctylamine in xylene extracted about 99 per cent of the niobium and only 0.8 per cent of the tantalum from an 8.0 molar hydrochloric acid solution. Scadden and Ballou (35) reported only partial separation by preferentially extracting niobium from a nitric acid solution of their oxalates into a 0.6 molar solution of di-n-propyl phosphoric acid in dibutyl ether. Stevenson and Hicks (36) effected a separation by extraction of tantalum into diisopropyl ketone from mineral acid-hydrofluoric acid aqueous phases. The mineral acids tested were hydrochloric, sulfuric, perchloric and nitric. Hydrochloric acid was reported to be most useful. All of the above extractions were carried out on aqueous solutions containing less than 2.5 grams each of tantalum and niobium per liter.

Ellenburg, Leddicotte and Moore (37) extracted niobium from tantalum as a sulfate with a mixed solvent of tribenzylamine in methylene chloride. Werning and associates (38, 39) obtained 99.85 per cent tantalum and 99.97 per cent niobium with the hydrofluoric acid-hydrochloric acid-methyl isobutyl ketone systems. West (40) has reviewed the above extraction papers in fair detail.

Wilhelm, Kerrigan and Cass (41) reported that many organic solvents were capable of preferentially extracting tantalum from a hydrofluoric acid solution containing high concentrations of niobium and tantalum. In furthering this work Foos and Wilhelm (42) studied the effects of varying the concentration of niobium, tantalum and hydrofluoric acid on the extraction. An aqueous solution containing the equivalent of 517 grams of tantalum and niobium pentoxides per liter was used in these experiments. Single stage extractions of this solution or its water dilutions with many organic solvents indicated that some solvents extracted more than 50 per cent of the total tantalum, analyzing less than one per cent niobium. Although the ketones were most effective for obtaining the separation, many alcohols, amines, aldehydes, ethers, esters, organic phosphates, organic phosphites and mixed organic solvents were useful. A multiple-contact batch extraction carried out on a solution containing 110 grams per liter of total oxides, with diethyl ketone, gave in three stages a 99 per cent recovery of niobium spectrographically free of tantalum and a 96 per cent recovery of the tantalum which analyzed about 0.15 per cent niobium. The most effective separation of tantalum from niobium was obtained when the free hydrofluoric acid content was low.

Other less corrosive systems were also investigated for separating tantalum from niobium by liquid-liquid extraction. Extraction of hydrofluoric acid solutions of niobium and tantalum which had been partially or completely neutralized with aliphatic or aromatic hydroxyamines indicated favorable mass distribution and separation of tantalum from niobium. Tantalum preferred the organic phase in these systems. Although the same general extraction trends were observed for the amine-free and the amine-neutralized hydrofluoric acid solutions of niobium and tantalum, the former system gave higher degrees of separation. Niobium was preferentially extracted by the organic phase from a potassium hydroxide solution of niobium and tantalum. However, in this system the mass transfer and degree of separation were low.

Several counter-current multistage extractions were carried out in which diethyl ketone was employed as the organic phase and amine-neutralized hydrofluoric acid solution of niobium and tantalum as the aqueous phase. In from 10 to 15 stages tantalum and niobium spectrographically free of each other were obtained quantitatively on a continuous basis.

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