

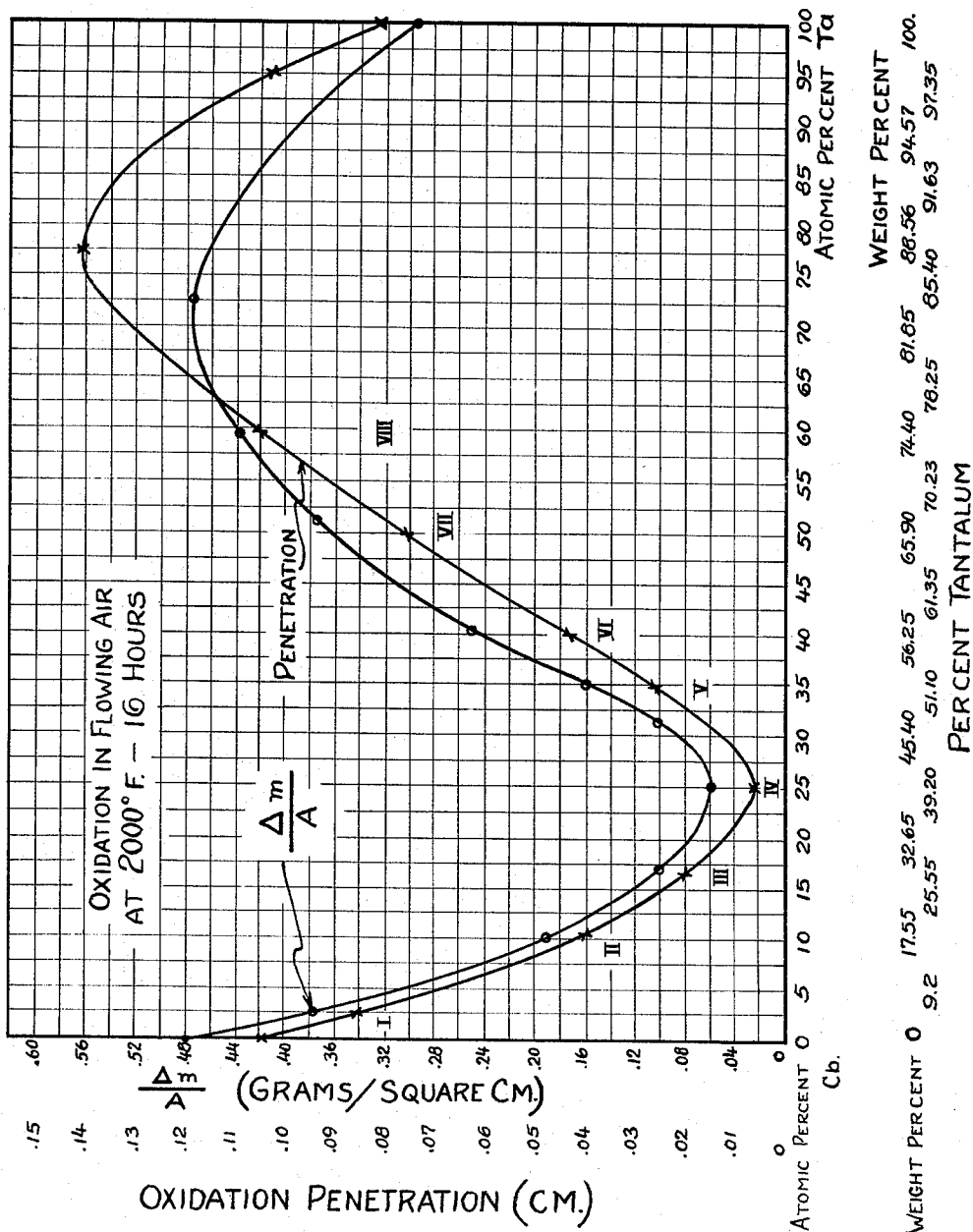
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COLUMBIUM-TANTALUM BINARY ALLOYS

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COLUMBIUM-TANTALUM BINARY ALLOYS

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The present invention relates to metal alloys or inter-metallic compositions and particularly to alloys and inter-metallic compositions of columbium and tantalum.

Tantalum and columbium have been described by many persons skilled in the art as equivalent metal having substantially the same properties and characteristics.

Among the characteristics of tantalum and columbium which were thought to be substantially the same, is the oxidation rate of these metals at elevated temperatures. For example, it has been determined that when substantially pure columbium is subjected to flowing air at temperatures of about 2000° F., the oxidation will penetrate about 0.15 cm. into the surface of the columbium base article in sixteen hours. A substantially pure tantalum article subjected to the same test for the same period of time will have an oxidation of about 0.082 cm. Considered within broad limits, these two values are more or less equivalent. In view of the closeness of these two figures it had been assumed that the oxidation of tantalum and columbium and alloys thereof were all about the same and that the metals and their alloys with each other to be equivalent.

By the present invention, however, there is provided an alloy consisting essentially of tantalum and columbium and which has an oxidation rate substantially less than that of columbium and tantalum, as low as 5% of the oxidation rate of substantially pure columbium.

Alloys of columbium and tantalum embodying this invention have most unusual and unexpected oxidation characteristics and in certain ranges of proportions of these metals in the alloys thereof the alloy will have an oxidation rate less than that of columbium and tantalum, while in other proportions of these metals in the alloy, the alloy will have an oxidation rate which is greater than that of columbium and tantalum.

In testing alloys of columbium and tantalum in accordance with this invention it has been found that the oxidation rate, measured by penetration of oxidation into the surface of the tested alloy object, at 2000° F., in flowing air for sixteen hours, is less than the oxidation rate of substantially pure columbium when the alloy contains tantalum up to 74.40% (60 atomic percent) the balance being substantially all columbium plus minor amounts of impurities. When the alloy contains more than 74.40% by weight of a tantalum, it has an oxidation rate greater than that of columbium. An alloy of columbium and tantalum, with minor amounts of impurities, having about 39.20% tantalum by weight (25 atomic percent) the balance being substantially all columbium, has an oxidation rate that is only about 4% of the oxidation rate of columbium and tantalum.

The oxidation penetration curve for alloys of columbium and tantalum resembles a sine curve as shown on the accompanying drawings by the line indicated "penetration."

The other curve on the graph of the accompanying drawings is designated

$$\frac{\Delta m}{A}$$

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and represents the change of mass of the article per unit surface area of the article measured prior to testing. By this curve it is shown that under the same test conditions as those described above for determining oxidation penetration, the change of mass per unit area for substantially pure columbium is 0.48 gram per square cm. of surface area of the test object. Substantially pure tantalum, when tested under the same conditions, has a change of mass per unit area of 0.30 gram per square cm.

The alloys of columbium and tantalum consisting of about 39.20% by weight tantalum (25 atomic percent) and the balance being substantially all columbium has a change of mass per unit area under the same test conditions of only about 0.08 gram per square cm.

This is a remarkable fact in view of all of the heretofore expounded theory that columbium and tantalum have substantially the same properties and characteristics.

Alloys of this invention have many important advantages and are useful for metal elements to be subjected to high temperatures and oxidizing and corrosive atmosphere such as in gas turbines and rockets etc.

While the oxidation rate of alloys of this invention is substantially less than the oxidation rate of either tantalum or columbium, the other physical properties of the metals are not impaired in the alloy. Indeed, the alloys of this invention are stronger than columbium alone.

As used herein, the term "atomic percent" means the percent of atoms in the alloy which are atoms of the particular metal indicated. For example, the designation "25 atomic percent tantalum" means that 25 percent of the atoms in the alloy are atoms of tantalum. The term "percent" as used herein to designate the proportions of the constituents in an alloy, means percent by weight, unless otherwise indicated.

There are set forth below examples of alloys of tantalum and columbium, illustrative of methods of making these alloys, and results of tests of those alloys. At the outset, it should be noted that alloys embodying this invention may be made by any desired process such as arc melting or sintering, which will be effective to alloy columbium and tantalum.

Example I

An alloy or intermetallic composition of 4 percent (25 atomic percent) tantalum and 96 percent (97.5 atomic percent) columbium was prepared by arc melting an electrode of columbium with 4 percent tantalum therein. The composite electrode consisting of a pressed and presintered powder blend of the required amounts of columbium and tantalum, was arc melted in a protective atmosphere of helium. The electrode was prepared by pressing a blend of powders of the metals at a pressure of about 50 tons per square inch to form a compact which was then presintered at a temperature about 2000° F. for about two hours in a vacuum of about 1 or 2 microns. Other conventional presintering conditions may be used.

The arc melting procedure may be carried out in a vacuum, or in an atmosphere of argon, a mixture of argon and helium, or other inert gases that will not combine with tantalum or columbium at elevated temperatures.

The resulting arc melted alloy when tested for oxidation resistance in flowing air at 2000° F. for sixteen hours had an oxidation penetration of about 0.085 cm. This compares favorably with the oxidation penetration of substantially pure columbium which, under the same test conditions, is 0.105 cm.

The change of mass per unit area of this alloy under these test conditions was about 0.38 gram per square cm. compared to a change of mass per unit area of about

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0.48 gram per square cm. for substantially pure columbium tested under the same testing conditions.

In determining the oxidation rate, i.e., penetration, the dimensions of the test article were determined prior to subjecting the sample to the oxidation test conditions. The oxide film which formed on the surface on the test sample was removed, as by grit blasting or grinding, following which the thickness of the sample was measured. The change in dimension provides the means of measuring the amount of metal lost by oxidation during the test period. The difference between the thickness of the original article and the thickness of the article following exposure to the oxidation test conditions is referred to as "oxidation penetration."

The change of mass per unit area is determined by measuring and weighing the test article prior to subjecting the same to the oxidation test conditions. After the oxidation test, the oxide film is removed and the article is again weighed. The difference in weight in the article divided by the original surface area of the article provides a measure of the amount of material lost due to oxidation and is referred to herein as "change of mass per unit area"

$$\frac{(\Delta m)}{A}$$

Example II

An alloy consisting of 17.55 percent tantalum (10 atomic percent) and 82.45 percent columbium (90 atomic percent) was prepared by arc melting an electrode made from the indicated amounts of the metals, in a mixture of helium and argon, in the manner described in Example I.

The resulting arc melted alloy, when tested for oxidation resistance in flowing air by subjecting a specimen thereof to a temperature of 2000° F. in flowing air for a period of sixteen hours underwent an oxidation penetration of 0.04 cm. and a change of mass per unit area of 0.18 grams per square cm.

These results compare very favorably with an oxidation penetration of 0.105 cm. and a change of mass per unit area of 0.48 grams per square cm. for substantially pure columbium subjected to the same test conditions.

Example III

An alloy consisting of 29.05 percent tantalum (17 atomic percent) and 70.95 percent columbium (83 atomic percent) was prepared by arc melting an electrode made from the indicated amounts of the metals in the manner described in Example I, in a mixture of helium and argon.

The resulting arc melted alloy, when tested for oxidation resistance in flowing air by subjecting a specimen thereof to a temperature of 2000° F. in flowing air for a period of sixteen hours underwent an oxidation penetration of 0.02 cm. and a change of mass per unit of 0.10 grams per square cm.

These results compare very favorably with an oxidation penetration of 0.105 cm. and a change of mass per unit area of 0.48 grams per square cm. for substantially pure columbium subjected to the same test conditions.

Example IV

An alloy consisting of 39.20 percent tantalum (25 atomic percent) and 60.80 percent columbium (75 atomic percent) was prepared by arc melting an electrode made from the indicated amounts of the metals in the manner described in Example I, in a mixture of helium and argon.

The resulting arc melted alloy, when tested for oxidation resistance in flowing air by subjecting a specimen thereof to a temperature of 2000° F. in flowing air for a period of sixteen hours underwent an oxidation penetration of 0.006 cm. and a change of mass per unit of 0.06 grams per square cm.

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These results compare very favorably with an oxidation penetration of 0.105 cm. and a change of mass per unit area of 0.48 grams per square cm. for substantially pure columbium subjected to the same test conditions.

Example V

An alloy consisting of 51.10 percent tantalum (35 atomic percent) and 48.90 percent columbium (65 atomic percent) was prepared by arc melting an electrode made from the indicated amounts of the metals in the manner described in Example I, in a mixture of helium and argon.

The resulting arc melted alloy, when tested for oxidation resistance in flowing air by subjecting a specimen thereof to a temperature of 2000° F. in flowing air for a period of sixteen hours underwent an oxidation penetration of 0.025 cm. and a change of mass per unit of 0.16 grams per square cm.

These results compare very favorably with an oxidation penetration of 0.105 cm. and a change of mass per unit area of 0.48 grams per square cm. for substantially pure columbium subjected to the same test conditions.

Example VI

An alloy consisting of 56.25 percent tantalum (40 atomic percent) and 43.75 percent columbium (60 atomic percent) was prepared by arc melting an electrode made from the indicated amounts of the metals in the manner described in Example I, in a mixture of helium and argon.

The resulting arc melted alloy, when tested for oxidation resistance in flowing air by subjecting a specimen thereof to a temperature of 2000° F. in flowing air for a period of sixteen hours underwent an oxidation penetration of 0.043 cm. and a change of mass per unit of 0.25 grams per square cm.

These results compare very favorably with an oxidation penetration of 0.105 cm. and a change of mass per unit area of 0.48 grams per square cm. for substantially pure columbium subjected to the same test conditions.

Example VII

An alloy consisting of columbium and tantalum, the tantalum being present in an amount of about 65.90 percent (50 atomic percent) and the columbium being present in an amount of about 34.10 percent (50 atomic percent) was prepared by arc melting a presintered electrode made from the indicated amounts of these metals by the method described in Example I, in an inert atmosphere of a mixture of helium and argon.

The resulting arc melted alloy, when tested for oxidation resistance by subjecting a specimen of the alloy to flowing air at 2000° F. for a period of sixteen hours had an oxidation penetration of only 0.075 cm. The change of mass per unit area of this alloy under the same test conditions was only about 0.380 grams per cm. Both of these values compare very favorably with the values of oxidation penetration and change of mass per unit area for substantially pure columbium when tested under the same conditions.

Example VIII

An alloy consisting of 74.40 percent tantalum (60 atomic percent) and 25.60 percent columbium (40 atomic percent) was prepared by arc melting a presintered electrode made from the indicated amounts of these metals by the method described in Example I in an atmosphere consisting of a mixture of helium and argon.

The resulting arc melted alloy when tested for oxidation resistance by subjecting a specimen of the alloy to flowing air at 2000° F. for a period of sixteen hours had an oxidation penetration substantially the same as the oxidation penetration of substantially pure columbium under the same test conditions, i.e., about 0.105 cm. However, the change of mass per unit area of this alloy under these test conditions was only about 0.44 grams

per square cm. compared to 0.48 grams per square cm. for substantially pure columbium.

With alloys of tantalum and columbium made by the method described in Example I, but having tantalum therein in an amount in excess of 74.40 percent (60 atomic percent), the oxidation resistance is not substantially improved over that of pure tantalum and columbium under the same test conditions as those described above. For example, an alloy of about 86.5 percent by weight tantalum (about 77 atomic percent) made as described in Example I and tested as described in Example I had an oxidation penetration of about 0.14 cm. This is to be compared with an oxidation penetration of substantially pure columbium, about 0.105 cm., under the same test conditions.

While the change of mass per unit area curve does not arise as sharply as the penetration curve at the high tantalum end thereof, it shows a change of mass per unit area of about 0.49 grams per square cm. when an alloy of tantalum and columbium containing about 83.25 percent tantalum (72.5 atomic percent), balance columbium, was tested under the test conditions described above.

The foregoing detailed description is given for clearness and for understanding of the invention, and no unnecessary limitations should be understood or implied therefrom, since some modification will be obvious to those skilled in the art.

I claim:

1. An alloy or intermetallic composition consisting essentially of columbium and tantalum, the tantalum being present in an amount in the range of from about 17

percent by weight to about 74.40 percent by weight and the balance being substantially all columbium and having a greater resistance to oxidation at elevated temperatures than substantially pure columbium.

2. An alloy or intermetallic composition consisting essentially of columbium and tantalum, the tantalum being present in an amount in the range of from about 17 percent by weight to about 51 percent by weight, the balance being substantially all columbium and having a greater resistance to oxidation at elevated temperatures than substantially pure columbium.

3. An alloy or intermetallic composition consisting essentially of columbium and tantalum, the tantalum being present in an amount in the range of from about 32 percent by weight to about 45 percent by weight, the balance being substantially all columbium and having a greater resistance to oxidation at elevated temperatures than substantially pure columbium.

4. An alloy or intermetallic composition consisting essentially of columbium and tantalum, the tantalum being present in an amount of about 39.2 percent by weight of the alloy, the balance being substantially all columbium and having a greater resistance to oxidation at elevated temperatures than substantially pure columbium.

References Cited in the file of this patent

"Initial Investigation of Niobium and Niobium-Base Alloys," Saller et al. United States Atomic Energy Commission, BMI-1003. May 23, 1955.

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