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B. C. GARDNER ET AL

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METHOD OF PRODUCTION OF REFRACTORY METAL LEAF

Filed Oct. 14, 1932

Fig. 1.

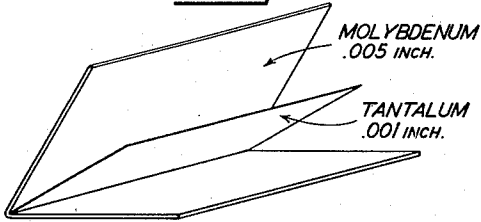


Fig. 2.

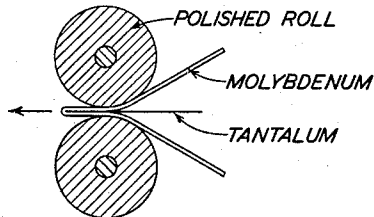


Fig. 3.

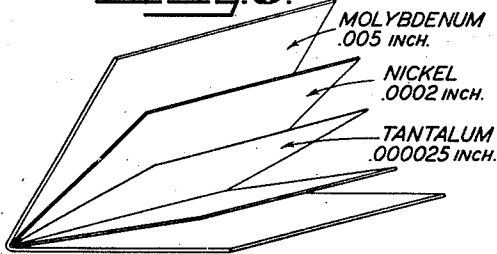


Fig. 4.

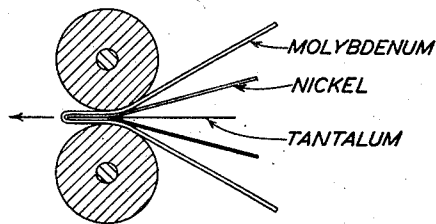


Fig. 5.

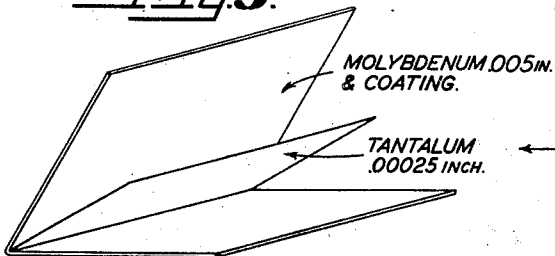


Fig. 6.

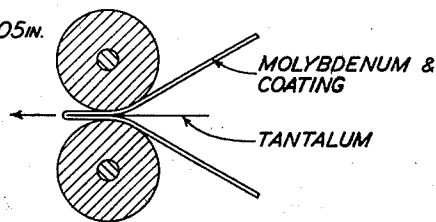


Fig. 7.

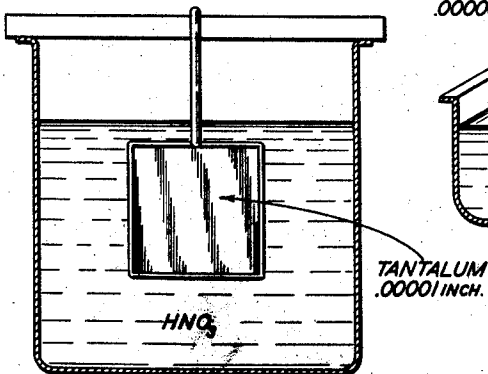
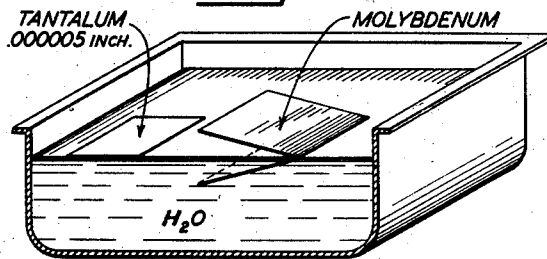


Fig. 8.



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METHOD OF PRODUCTION OF REFRACTORY METAL LEAF

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7 Claims. (Cl. 29-18)

Our invention relates to a refractory metal leaf and method of production, and more particularly to a method of producing a tantalum leaf having a substantially uniform thickness and a high
5 polish.

Among the objects of this invention are: to produce a refractory metal leaf having a uniform thickness of less than .00025 inch; to produce a refractory metal leaf having a high polish imparted by a rolling process; to produce a tantalum leaf having a thickness less than .00001 inch; to provide means of handling such thin metal leaf; to provide methods whereby refractory metal may be rolled to extreme thinness; to provide a material for use as a lubricant in a rolling process which produces thin refractory metal leaf; and to provide a method of annealing thin sheets of refractory metal.

Other objects of our invention will be apparent or will be specifically pointed out in the description forming a part of this specification, but we do not limit ourselves to the embodiment of the invention herein described, as various forms may be adopted within the scope of the claims.

Referring to the drawing, which diagrammatically illustrates various steps of our invention:

Figure 1 shows the formation of a book, or container for refractory metal.

Figure 2 shows the book of Figure 1 passing through polished rolls.

Figure 3 shows the formation of another type of book.

Figure 4 shows the book of Figure 3 passing through the rolls.

Figure 5 shows an alternative form of book for extremely fine leaf.

Figure 6 shows the book of Figure 5 passing through the rolls.

Figure 7 shows a method of mounting the fine
40 leaf.

Figure 8 shows a method of floating extremely fine leaf on water.

While refractory metals, such as tungsten, molybdenum and tantalum may be drawn into fine wire, they have not, as far as we are aware, been fashioned into leaf form. Ordinary rolling processes will produce sheet with a minimum of perhaps .0005 inch although a sheet .001 inch is the minimum supplied by most commercial
50 producers.

In its broadest sense our invention comprises a method of rolling refractory metal such as tantalum for example to a substantially uniform thickness of less than .000005 inch. The
55 method comprises repeated rollings of a book

formed of sheet metal, the book containing the metal to be reduced to leaf form. We also have invented a lubricant to be placed between the leaf and the book so that the leaf may not stick to the book covers, or we may remove the
60 book material with an acid which does not attack the leaf. These methods produce a leaf having a desirable high polish.

While we employ certain differing procedures in order to obtain different thicknesses of leaf
65 we prefer to start, in a method wherein it is desired to produce tantalum leaf, with a book formed from molybdenum sheet .005 inch thick, by folding in the center to form two covers, as shown in Figure 1.

A square of .001 inch tantalum sheet preferably about $1 \times \frac{1}{2}$ inch is first thoroughly degassed and annealed in vacuo as nearly as possible to its melting point or, in practice to about 2400° C. This is preferably done by enclosing the tantalum in a tungsten cylinder of small diameter, and heating the cylinder to incandescence by using it as a filament in an evacuated container, while the vacuum pumps
70 are operating.

The tantalum is then removed and placed in the book, which is cut to 1 inch across the fold by about 2 inches length of each cover. The inside of the molybdenum covers are polished to an extreme fineness and the tantalum placed so
85 that its 1 inch width corresponds with the fold width of 1 inch, leaving $1 \frac{1}{2}$ inches toward the open end of the covers for expansion of the tantalum. The tantalum is placed with one edge directly against the fold.

Closing the book, it is then fed through polished jewelers' rolls repeatedly, always starting the book through fold first, as shown in Figure 2.

The book is rolled until the tantalum has nearly doubled its area, i. e., to about 1×1 inch.
95 There is practically no expansion laterally of the rolls, the tantalum enlarging toward the open end of the book.

After few rollings, the polished surface of the molybdenum is examined, and if it appears cloudy or otherwise damaged, the tantalum is removed and placed in another book having a properly polished surface. This precaution is necessary, as the polish is removed by rolling, and if rolling is continued after the book polish
100 has deteriorated, leaf cannot be produced.

The repeated rollings are then continued until a thickness of about .0002 inch is obtained, when it is desirable to re-anneal, and cut to size again. New polished books are preferably
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provided at thicknesses of approximately .00007 inch, and .00004 inch, until a thickness of .000025 inch is reached, which is as far as we prefer to go using only the polished molybdenum books. Where a leaf of this thickness is desired, the above process is sufficient, and the leaf may be readily removed from the book.

An examination of the leaf thus produced shows a substantially uniform thickness, no thin spots, and the leaf has a highly polished surface.

When thinner leaf is desired, it may be produced by several methods. In one such method we roll nickel sheet to .0002 inch in exactly the same manner as above described for tantalum. We then form a book of the nickel, place a piece of the .000025 inch tantalum therein as described for the start of the original process, and then place the nickel book inside of a polished molybdenum book, with the fold of one against the fold of the other, as shown in Figure 3. The nickel, having been rolled between polished molybdenum, will have a high polish also.

The combined books are inserted in the rolls as shown in Figure 4 and the repeated rollings started. The nickel book increases in area, and decreases in thickness, as does the tantalum inside. The molybdenum covers are replaced frequently, but the nickel book is not disturbed, until the increase in area indicates that the tantalum has decreased to .00001 inch.

The nickel-tantalum book now cannot be opened, as the rolling has caused them to stick together, so the entire book is spot welded to a frame, usually a frame in which the leaf is intended to be used. This welding is preferably performed under water.

The nickel is then dissolved away in HNO_3 as shown in Figure 7, and as the frame is made of a material not soluble in the acid, the tantalum remains. It is washed and dried, and is then ready for use as a leaf having a high polish and a uniform thickness of .00001 inch.

We have found, in the welding process, that the weld spots are not attacked by the acid, even though nickel is between the tantalum and the frame. It is our belief the nickel has melted away directly at the spot, allowing the tantalum to come in direct contact with the frame. At least, the acid does not loosen the welds.

We have found it desirable to discontinue the nickel-tantalum book after a thickness of .00001 inch is reached, and if a still thinner leaf is desired, to utilize still another method.

A highly polished molybdenum book is provided with a lubricated inner surface. This coating may be formed by exposing the highly polished inner surfaces of the book to the fumes of magnesium burning in air, which appear to deposit on the surfaces an extremely fine, impalpable, almost evanescent coating of magnesium oxide. It is however amply sufficient to prevent the tantalum from sticking to the metal of the covers. Other fumes, e. g., those of aluminum or even soot, which will deposit an exceedingly fine coating are satisfactory, as we believe the lubricant to be purely physical in character.

We then place a cut and annealed piece of .00025 inch tantalum leaf, produced by one of the above described methods, in the fold of the coated molybdenum book and roll to .000005 inch.

The molybdenum book may then be carefully opened, the leaf will be free, and will float out on a water surface as shown in Figure 8. The

finished leaf may then be welded to a frame, or utilized in any manner desired.

The methods as above described have been utilized to produce a refractory metal leaf of extreme thinness, and when such leaf is produced by those methods, a high polish is carried all the way through to the final product. Even the thinnest leaf produced, that having a thickness of less than .000005 inch is remarkably uniform in thickness, being free both from thin spots, and from wedging or other gross dimensional changes.

While the method has been described as applied to the sizes and materials which we have found most suitable to our own work, the usefulness of the method is not thus limited. We have substituted "stainless steel" for nickel and even for the molybdenum with a fair degree of success, although the process cannot be carried as far, in a single stage, when this is done. Nickel can be rolled very thin between molybdenum or tantalum leaves, for example.

In general, best results are obtained by forming the book or casing of harder and less ductile material than the metal of the leaf. Added rigidity can, in some degree, be imparted by greater thickness and by cold-working and polishing, as is evidenced by the use of nickel for rolling the harder and less ductile tantalum, but this procedure is limited in the amount of attenuation which may be obtained. There appears to be no definite lower limit, however, to the process of drawing out nickel between stainless steel, or stainless steel between molybdenum. The use of a less desirable casing material may in some degree be compensated for by imparting to it a higher polish, and by more frequent casing changes, as the breakdown of the foil is always preceded by the deterioration of the inner surface of the book; as soon as this surface begins to have an etched appearance the casing should be changed.

As far as is known the above described method is the only way by which refractory metals may be formed into leaf of extreme thinness, but it should be understood that the processes described will produce leaf equally well from metals which have hitherto been reduced to leaf form by other methods well known in the art. All metal leaves, however, produced by the processes as described, will be characterized by their high polish.

We claim:

1. The method of producing a metal leaf which comprises enclosing said metal in a polished sheet metal container, repeatedly rolling said container until said polish is impaired, inserting said metal in a newly polished container, and repeating said rolling.

2. The method of producing a metal leaf which comprises enclosing said metal in a polished container, repeatedly rolling said container, removing the partly rolled leaf, enclosing said partly rolled leaf in a newly polished sheet metal container, coating the interior walls of said last mentioned container with a deposit from the fumes of metal burned in air, and repeatedly rolling the book thus formed.

3. The method of producing a metal leaf which comprises enclosing said metal in a polished container, repeatedly rolling said container, removing the partly rolled leaf, enclosing said partly rolled leaf in a newly polished sheet metal container, coating the interior walls of said last mentioned container with a deposit from the fumes of metal burned in air, repeatedly rolling the

book thus formed, opening said book, and allowing the finished leaf to float free of said book on the surface of a liquid.

4. The method of producing a refractory metal leaf which comprises enclosing said refractory metal in a polished refractory container, repeatedly rolling said container, removing the partly rolled leaf, enclosing said partly rolled leaf in a polished non-refractory metal container, enclosing said non-refractory container in a polished refractory container, repeatedly rolling the book thus formed, removing said non-refractory container, and dissolving said non-refractory metal in a liquid which will not attack the resultant refractory metal leaf.

5. The method of producing a refractory metal leaf which comprises enclosing said refractory metal in a polished refractory container, repeatedly rolling said container, removing the partly rolled leaf, enclosing said partly rolled leaf in a

polished non-refractory metal container, enclosing said non-refractory container in a polished refractory container, repeatedly rolling the book thus formed, removing said non-refractory container, and dissolving said non-refractory metal in acid which will not attack said leaf.

6. In the method of producing a delicate metal leaf by rolling between metal covers joined at one edge, the step of opening said covers in a liquid and allowing the contained leaf to float out of said covers on the surface of said liquid.

7. The method of producing a refractory metal leaf which comprises enclosing said refractory metal in a sheet metal container, said container being formed of a refractory metal slightly softer than the metal to be leafed, and rolling said container to produce expansion of both materials.

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