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CARBIDE DEPOSITION ON TANTALUM

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The present invention relates to a method of forming a carbide coating on metal and, more particularly, forming a surface layer of tantalum carbide on tantalum metal.

Liquid fuel reactors possess many advantages over solid fuel reactors such as reprocessing during operation and rapid change from a critical configuration to a non-critical configuration. Utilization of plutonium as a reactor fuel is advantageous when utilized in a breeding cycle. Consequently, a great deal of effort has been invested in the development of a molten plutonium reactor. It has been found quite difficult to find a material capable of containing the molten plutonium without excessive corrosion. Perhaps the most promising material is carbide coated tantalum metal. Since this coated tantalum is to be used as a structural material, it is highly desirable that the metal substrate retain its ductile characteristics.

Tantalum powders have been coated with tantalum carbide by the intimate admixture of tantalum powder with carbon black under dry hydrogen between 1400 to 1600° C. From this prior art, it might be expected that massive tantalum could be coated with carbide at a higher temperature. Although this method forms a tantalum carbide coating on a tantalum powder, it has been found that the carbide diffuses into the tantalum substrate thereby causing the tantalum to assume an unwanted hardness. Carburization has also been effected by heating in a methane atmosphere. However, this method is disadvantageous in that the methane spontaneously pyrolyzes thereby depositing an interfering coat of carbon on the surface. Such a coating is spotty and incapable of protecting the tantalum metal as would a carbide coating.

The present invention permits the formation of a very thin coherent layer of tantalum carbide on tantalum metal when it is heated in contact with lampblack in an atmosphere of hydrogen. Temperatures of 800 to 1160° C. permit the retention of the tantalum substrate softness.

It is accordingly an object of the present invention to provide a tantalum carbide coated tantalum metal wherein the tantalum metal substrate retains its ductile characteristics.

It is a further object of this invention to provide a thin coherent coating of tantalum carbide on tantalum metal thereby protecting the tantalum from oxidation.

According to the present invention a piece of tantalum is packed in lampblack and the system is evacuated to about 10^{-5} mm. of mercury. The system is heated to the desired temperature (between 800 and 1160° C.) and held at this temperature for between 15 and 50 minutes in order to outgas the tantalum metal and lampblack. Hydrogen is then added to a pressure above 100 mm. of mercury, preferably between 200 and 300 mm. of mercury. Hydrogen is added after attaining the desired temperature in order to minimize hydride production and to permit outgassing. It is assumed, although this assumption forms no part of the invention, that methane produced at the desired temperature reacts with tantalum to form a light surface coating of carbide. This assumption is based on the fact that there is no carburization if hydrogen is not present.

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It should be noted that lampblack is the only carbon source presently known which is efficacious in the above method. All sizes and purities of graphite have been utilized, but have uniformly failed to achieve a satisfactory coating. Furthermore, the lampblack packing may only be used one time effectively. This tends to indicate that the contaminants in lampblack are beneficial and permit the satisfactory coating to be obtained under the reaction conditions specified. Lampblack of average particle size 90 angstroms, can yield a coating as thin as one micron when a temperature of 800° C. is utilized. If a less finely divided lampblack is used, e.g., less than one micron particle size and approximately $\frac{1}{10}$ micron particle size, a coating of between 2 and 3.5 microns thickness is achieved. Temperatures of 1200° C. and above are not useful in the above-described method since these temperatures will result in hardening of the tantalum substrate.

The method of the invention can best be illustrated by the following examples which are given for this purpose only and not for the purpose of limiting the spirit and scope of the present invention.

Example 1

A tantalum metal piece is packed in a 95-angstrom average particle size lampblack. The metal and lampblack are placed in a quartz tube furnace and heated to 800° C. at a pressure of about 10^{-5} mm. of mercury. These conditions are held for 45 minutes for outgassing purposes and then hydrogen is admitted bringing the pressure up to 200 mm. of mercury. These conditions are held for two hours and then cooled to room temperature. The tantalum carbide coating found on the tantalum metal piece is coherent and one micron thick.

Example 2

A tantalum metal piece is packed in a less finely divided lampblack (i.e., approximately $\frac{1}{10}$ micron average particle size) placed in a quartz tube furnace. The system is heated to 1100° C. at a pressure of about 10^{-5} mm. of mercury and held at these conditions for about 15 minutes. Subsequently, hydrogen is admitted to bring the pressure up to 300 mm. of mercury and the 1100° C. temperature and 300 mm. hydrogen pressure is maintained for two hours and 15 minutes. A $3\frac{1}{2}$ micron tantalum carbide coating results.

Example 3

Lampblack as in Example 2 is used in packing a tantalum metal in a quartz tube furnace. A temperature of 1100° C. at a pressure of about 10^{-4} to 10^{-5} mm. of mercury is maintained for 50 minutes, after which hydrogen is admitted bringing the pressure up to 300 mm. of mercury. The 1100° C. temperature is maintained for one hour and a coating of two to three micron thickness results.

It will be seen from the above discussion that a method is described which coats tantalum metal with tantalum carbide at unexpectedly low temperatures, thereby yielding a beneficial high ductility tantalum substrate. Lampblack is the only known carbon source which will permit the above described reaction and each lampblack packing may be used only once effectively. The method is independent of surface treatment. Some samples have been pickled in acid solution and yielded identical coatings under identical reaction conditions as those utilizing an unpickled metal piece.

What is claimed is:

1. The method of forming a tantalum carbide on tantalum metal comprising heating the tantalum metal in

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contact with lampblack in an atmosphere of hydrogen at a temperature between 800 and 1160° C.

2. A method as in claim 1 wherein the hydrogen is added after the desired temperature is maintained for a period between 15 and 50 minutes.

3. A method as in claim 2 wherein the reaction system is held in a vacuum of about 10^{-5} mm. of mercury before the addition of hydrogen.

4. A method as in claim 3 wherein a sufficient quantity of hydrogen is added to bring the reaction system pressure to above 100 mm. of mercury.

5. A method as in claim 4 wherein the pressure after addition of hydrogen is between 200 and 300 mm. of mercury.

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6. A method as in claim 1 wherein the lampblack is of less than one micron average particle size.

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