Patented Sept. 25, 1951 2,569,225

UNITED STATES PATENT OFFICE

2,569,225

METHOD OF FORMING URANIUM MONOCARBIDE

James H. Carter, Blacksburg, Va., and Adrian H. Daane, Ames, Iowa, assignors to the United States of America as represented by the United States Atomic Energy Commission

No Drawing. Application March 26, 1947, Serial No. 737,430

10 Claims. (Cl. 23—14.5)

1

This invention relates to a uranium-carbon alloy having a low carbon content. More specifically our invention is concerned with the formation of uranium monocarbide from uranium metal and carbon.

It is an object of this invention to provide an efficient process for the preparation of uranium monocarbide.

Other objects and advantages of this invention will be readily apparent from the following description of preferred embodiments of our invention.

Broadly stated, it is well-known in the art that uranium and carbon may be heated in comminuted contact with each other at an elevated temperature for a sufficient time to form uranium-carbon compounds or alloys. These temperatures are in excess of the melting point of uranium; and the compound thus produced contains a composite mass of uranium dicarbide (UC_2) and uranium sesquicarbide (U_2C_3).

In accordance with an embodiment of the present invention the existence of another specific uranium carbide, uranium monocarbide, is established by metallographic examination of a mass produced by contacting uranium and carbon in stoichiometric proportions at a temperature less than the melting point of the uranium metal.

The solubility of carbon in solid uranium is less than in the liquid phase which occurs at a temperature in excess of 1125° C. The solubility of carbon in solid uranium has been experimentally determined. The solubility of carbon in the gamma phase, wherein the temperature of the solid metal is in excess of 770° C. is of the order of 0.05 percent (1 atom percent carbon). The uranium metal containing 0.05% carbon shows a substantial dissolution of the carbide when annealed approximately 96 hours at 1000° C. The upper limit of the solubility of carbon in the beta-uranium within a temperature range of 660° C. to 770° C. is of the order of 0.03 percent (0.6 atom percent carbon); while in the alpha phase wherein the uranium metal is at a temperature less than 660° C. the solubility is not much greater than 0.01 percent (0.2 atom percent carbon).

In examining the metallographic crystal structure of the carbide formation in the uranium mass wherein the reaction temperature was kept below 1125° C. (melting point of uranium), a dendritic structure existed in the form of cubic faced-centered crystals (probably NaCl-structure), isomorphous with those of UN and UO. According to observations, the carbon in uranium

2

monocarbide can be replaced by oxygen or nitrogen to give a slight decrease in the lattice constant (as low as 4.936 Å. when UO_2 is found); and therefore the most reliable value of the lattice constant is the highest one obtained, primarily 4.951 Å. The lattice constant of uranium monocarbide does not markedly depend upon the presence of an excess amount of uranium or carbon, thus indicating low solubility of both components in the carbide.

In examining the uranium carbon system, it shall be noted that evidence for the existence of a high temperature compound is somewhat slight, although Widmanstatten patterns of alloys between uranium sesquicarbide (U_2C_3) and uranium dicarbide (UC_2) have been interpreted as indicating the existence of solid solutions of αU_2C_3 and ρUC_2. No carbides with more than two carbon atoms per uranium atom seem to exist although there is some X-ray evidence, however, for solubility of carbon in the UC_2 lattice, particularly at temperature near the melting point (2375° C.) of the uranium dicarbide alloy.

The system diagram shows that where the stoichiometric proportions of carbon to uranium exceed 4.8% by weight of carbon there is a greater tendency to form the UC_2 structure and probably the U_2C_3 structure, depending on the temperature. When the stoichiometric percentage of carbon is reduced to less than 4.8% of carbon, the tendency of the reactive mass of uranium and carbon to form uranium monocarbide is enhanced; that is, at a temperature less than the melting point of the uranium metal (1125° C.).

A mode of preparing uranium monocarbide consists in mixing powdered uranium metal with powdered carbon in stoichiometric proportions wherein the carbon content is less than 4.8% of the composite mass content, then pressing the mixture into small blocks under high pressure, 20–50 tons p. s. i., for example, and then heating the pressed blocks in a temperature range less than the melting point of uranium, preferably at about 900-1000° C. for about 100 hours.

Another method of forming uranium monocarbide is to compositely mix comminuted uranium and carbon in stoichiometric proportions to form uranium monocarbide, and then pressing the mixture into small blocks under high pressures, about 50–100 tons p. s. i., for example, then heating the pressed blocks in a temperature range above the melting point of the uranium metal and less than the melting point of the uranium carbide produced, 2250° C., whereby a mixture of UC_2 and U_2C_3 is formed, and then

quenching the composite mass to a temperature less than the melting point of the uranium metal at about 900° C. The carbon alloy thus formed will disproportionate into uranium dicarbide containing uranium monocarbide and carbon, independently of an excess component in the melt.

In the foregoing process of producing uranium monocarbide, another embodiment consists of heating uranium metal of high purity content with carbon in a stoichiometric amount of about 4.8 per cent at subatmospheric pressure at a temperature in excess of 1125° C. and then quenching the uranium-monocarbide composition to a temperature of about 900° C.

In another embodiment of the present invention, it was found that uranium monocarbide may be produced by heating uranium metal of high purity content with carbon in a stoichiometric amount of about 4.8 per cent by weight in a carbon dioxide atmosphere at a temperature between 900° C. and 1125° C.

Numerous variations and modifications in the preferred methods and examples described will be readily apparent and may be made without departing from the spirit and scope of our invention as defined in the following claims.

This application is a continuation-in-part of our co-pending application, Serial No. 549,282, filed August 12, 1944, now Patent No. 2,526,805.

The method of forming uranium monocarbide by heating uranium oxide and carbon and the product are the subject matter of Patent No. 2,448,479, granted to Harley A. Wilhelm and Adrian H. Daane on August 31, 1948.

What is claimed is:

1. The process of producing uranium monocarbide which comprises heating uranium metal in contact with carbon in approximately stoichiometric amount at a temperature sufficiently high to react but below the melting point of uranium.

2. The process of producing uranium monocarbide which comprises heating uranium metal of high purity with carbon in a stoichiometric amount of about 4.8% by weight in a carbon dioxide atmosphere at a temperature at between approximately 900° C. and 1125° C.

3. A process of producing uranium monocarbide which comprises heating uranium in contact with carbon at a temperature in excess of the melting point of uranium so as to form a uranium-carbon mass consisting essentially of uranium, UC_2 and U_2C_3 and then quenching and maintaining said mass at a temperature below the melting point of uranium but sufficiently high to form uranium monocarbide.

4. The process of producing uranium monocarbide which comprises heating uranium metal of high purity in contact with carbon at a temperature in excess of the melting point of the uranium metal so as to form a uranium carbon alloy consisting essentially of uranium, UC_2 and U_2C_3, and then quenching and maintaining said alloy at a temperature above about 900° C.

5. The process of producing uranium monocarbide which comprises heating uranium metal of high purity in contact with carbon in a stoichiometric amount at a temperature in excess of the melting point of the uranium metal so as to form a uranium-carbon alloy consisting essentially of uranium, UC_2 and U_2C_3, and then quenching and maintaining the alloy at a temperature of about 900° C.

6. The process of producing uranium monocarbide which comprises heating uranium metal of high purity with carbon in a stoichiometric amount of about 4.8% at a temperature in excess of the melting point of uranium metal whereby a mixture of uranium, UC_2 and U_2C_3 is obtained and then quenching and maintaining at a temperature of about 900° C.

7. The process of producing uranium monocarbide which comprises heating uranium metal of high purity with carbon in a stoichiometric amount of about 4.8% at sub-atmospheric pressure at a temperature in excess of 1125° C. whereby a mixture of uranium, UC_2 and U_2C_3 is obtained and then quenching and maintaining at a temperature of about 900° C.

8. A process for producing uranium monocarbide, comprising reacting a uranium- and carbon-containing mixture selected from the group consisting of a mixture of uranium plus carbon in about stoichiometric amounts and a mixture of uranium and UC_2 plus U_2C_3 in which the total uranium and the total carbon contents are present in approximately stoichiometric proportions by heating the mixture to a temperature sufficient to bring about reaction but below the melting point of metallic uranium and maintaining said temperature for reaction and formation of uranium monocarbide.

9. A process for producing uranium monocarbide, comprising reacting a uranium- and carbon-containing mixture selected from the group consisting of a mixture of uranium plus carbon in which uranium is present in at least stoichiometric amounts and a mixture of uranium, UC_2 and U_2C_3 in which the total uranium is present in at least stoichiometric proportions as to uranium monocarbide by heating the mixture to a temperature sufficient to bring about reaction but below the melting point of metallic uranium, and maintaining said temperature for reaction and formation of uranium monocarbide.

10. A process for producing uranium monocarbide, comprising reacting a mixture of uranium, UC_2 and U_2C_3 in which the total uranium is present in at least stoichiometric proportions as to uranium monocarbide by heating the mixture to a temperature sufficient to bring about reaction but below the melting point of metallic uranium, and maintaining said temperature for reaction and formation of uranium monocarbide.

JAMES H. CARTER.
ADRIAN H. DAANE.

No references cited.