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METHOD OF FORMING URANIUM CARBIDE

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This invention relates to uranium-carbon compounds and more particularly to uranium-carbon alloys having a low carbon content. More specifically, the invention is concerned with the formation of uranium monocarbide.

Broadly stated, it is well-known in the art that when uranium and carbon are heated in contact with each other at an elevated temeprature for a sufficient period of time, a compound or alloy may be formed having a uranium-carbon composition. The structural composition of the alloy or compound thus formed is dependent upon the stoichiometric amounts of carbon and uranium present and upon the temperature at which the alloying occurs.

When the uranium-carbon alloy is heated to a temperature in excess of 2300° C., approaching 2400° C., there is formed an alloy, uranium sesquicarbide (U_2C_3) which will disproportionate upon cooling to uranium dicarbide (UC_2). The art does not reveal the existence of any other characteristic alloy of the uranium-carbon system, other than uranium dicarbide and uranium sesquicarbide.

In accordance with a specific embodiment of this invention, it is herein contemplated to form uranium monocarbide (UC) in distinct proportions from uranium dicarbide (UC_2) and uranium sesquicarbide (U_2C_3).

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

It has been found in accordance with the method of this invention that specific uranium compounds or alloys containing carbon may be prepared by heating uranium metal in contact with methane at an elevated temperature.

The solubility of carbon in comminuted uranium metal is extremely low when the uranium is in the beta state having a temperature between 660 and 770° C. The solubility of the carbon in the beta-uranium is of the order of approximately 0.03% (0.6 atom percent) C. while in the uranium-alpha phase, less than 660° C., the solubility is of the order of 0.01% (0.2 atom percent) C. As the temperature of the uranium metal is increased, the solubility of the carbon approaches a maximum in the liquid uranium phase, greater than 1125° C. It shall be noted that carbon is slightly soluble in uranium metal when the uranium metal is in the alpha phase, and that a slight solubility of carbon even occurs when the uranium metal is heated at a temperature of about 400° C.

In examining the existence of uranium-carbon alloys over a temperature range from about 400°

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C. to a temperature in excess of about 2375° C., there is evidence for the existence of a high temperature uranium-carbon alloy having the structural formula of U_2C_3. Upon cooling, this compound (U_2C_3) disproportionates into uranium-carbon compounds, primarily UC_2. In order to form uranium-carbon compounds having at least two atoms of carbon per one atom of uranium, it is found that the carbon content must exceed 4.8% by weight and that the temperature must be in excess of the melting point of the uranium metal at about 1125° C. In addition, it is found that by increasing the temperature to about 1800° C., a greater proportion of uranium dicarbide is formed.

In examining the carbon-uranium system, the proportion of carbon to uranium must exceed 4.8% by weight to form the dicarbide and less than 4.8% carbon to form the uranium monocarbide.

Uranium-carbon alloys may be prepared by contacting a hydrocarbon gas, such as methane, with comminuted uranium metal and allowing the methane gas to be in continuous contact with the uranium in an amount proportional to a stoichiometric amount to form the desired uranium-carbon alloy. In addition, it is noted that after forming the uranium-carbon alloys, primarily uranium dicarbide at an elevated temperature, the alloy may be quenched to a temperature less than the melting point of the uranium metal, less than 1125° C., so that uranium-carbon alloy thus formed disproportionates into another specific uranium-carbon alloy, primarily uranium monocarbide. The uranium-carbon alloy produced by quenching exhibited a dendritic structure identifying a preponderance of uranium monocarbide. The uranium monocarbide thus produced did not exhibit an extremely high purity, since a small percentage of uranium dicarbide was still present; another method for producing uranium monocarbide was therefore devised.

In accordance with another embodiment of the invention uranium monocarbide of high purity may be prepared by reacting a hydrocarbon, such as methane, in contact with uranium metal at a temperature less than the melting point of the uranium metal. In this mode of preparation the tendency toward the formation of uranium-carbon alloys other than uranium monocarbide is considerably reduced. In addition, by introducing the carbon content, by means of methane, and the uranium content in stoichiometric relationships, the tendency to specifically form uranium monocarbide is increased.

As an example, uranium monocarbide may be

formed by contacting comminuted uranium metal of high purity in an enclosed vessel with hydrocarbon gas, such as methane, of high purity, and then increasing the reaction temperature to a temperature in excess of about 400° C. The reaction takes place as shown in the following formula:

$$U+CH_4 \rightarrow UC+2H_2$$

The preferred mode of operation embodies the use of a continuous flow system wherein purified methane is allowed to flow and contact a heated comminuted uranium mass in a stoichiometric amount wherein the amount of carbon supplied by the methane gas shall not be in excess of approximately 4.8% by weight. The operating temperature for forming the uranium monocarbide, in this particular process, is considered to be between 400° and the melting point of uranium which is about 1125° C.; although the preferred operating temperature was found to be about 900° C.

As another example of the invention, uranium monocarbide may be prepared by contacting purified methane in a dynamic flow system with uranium hydride at a temperature in excess of the decomposition temperature of the uranium hydride which is at about 435° C.

In this process a very reactive form of powdered uranium is probably formed in the first decomposition reaction wherein uranium hydride may be decomposed by heat to uranium and hydrogen. The reaction may be considered as follows:

(1) $$2UH_3 + \text{heat} \xrightarrow[>435^\circ C.]{\Delta H} 2U + 3H_2$$

The uranium metal then reduces the methane and combines with the carbon of the methane, and this may be illustrated by the following equation:

(2) $$U + CH_4 \xrightarrow[>400^\circ C.]{\Delta H} 2UC + 2H_2$$

The combined reaction may be illustrated in the following manner:

(3) $$2UH_3 + 2CH_4 \xrightarrow[>400^\circ C.]{\Delta H} 2UC + 7H_2$$

The reaction shown in Equation 3 is considered to be exothermic; and although the initial decomposition temperature may be approximately 435° C. there will be a rise in temperature proportional to the heat of reaction.

It has been found that a relatively high proportion of uranium monocarbide is formed when the amount of reactants is held to the required stoichiometric proprotions relative to the amounts required to form uranium monocarbide and when the temperature of the reaction is maintained at about 900° C.

It shall be also noted that uranium carbon alloys may be formed by using this process wherein the stoichiometric amount of carbon exceeds the 4.8% relationship to the uranium content and when the temperature exceeds the melting point of the uranium metal. It has been found that in using this mode of reaction the uranium carbon alloys having an atomic carbon content greater than one will disproportionate upon cooling or quenching to form uranium monocarbide. This method does not produce relatively pure uranium monocarbide but produces a mixture of dendrites including uranium monocarbide and uranium dicarbide.

Thus, I have been able by following the process described and claimed herein to consistently produce a mass of uranium monocarbide having a predetermined carbon content proportional to the stoichiometric amount required to form the uranium monocarbide.

While the foregoing embodiments of the invention have been discussed as performed at atmospheric pressure in a dynamic system involving the continuous flow of methane through the reactive mass of uranium or uranium hydride, a batch process has also been found suitable wherein stoichiometric amounts of uranium and carbon have been introduced into the system and the reaction carried out in a similar manner. Although methane is preferred, particularly for obtaining uranium monocarbide, it shall be noted that the invention shall not be limited to methane and that other compounds may be used to introduce the carbon content; such compounds may be ethane, propane, and other similar hydrocarbon compounds.

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 the invention as defined in the following claims.

The composition, uranium monocarbide, and a process of manufacturing it by the reduction of uranium oxide with carbon, are the subject matter of the Patent No. 2,448,479 granted to Harley A. Wilhelm and Adrian H. Daane on August 31, 1948.

What is claimed is:

1. A process for producing uranium monocarbide which comprises heating uranium metal in contact with methane in stoichiometric amounts at a temperature about 400° C. and less than 1125° C. wherein the carbon content thus introduced is equal to about 4.8% to the uranium metal content.

2. A method of preparing uranium monocarbide, comprising heating stoichiometric quantities of uranium with methane at substantially 900° C.

3. A process for producing uranium monocarbide, which comprises heating uranium metal in contact with a quantity of aliphatic hydrocarbon corresponding to about 4.8% carbon with regard to the uranium metal at a temperature above 400° C. and less than 1125° C.

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REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
560,291	Acheson	May 19, 1896
2,380,008	Abrams et al.	July 10, 1945

OTHER REFERENCES

Friend, Textbook of Inorganic Chemistry, vol. VII, Part III, pages 333–334 (1926), pub. by Charles Griffin and Co., London; copy in Sci. Lib.

"Fuels and Their Combustion" by Haslam et al,. McGraw-Hill Book Co., Inc. (1926), p. 185. (Copy in Division 59.)

Mellor, Inorganic and Theoretical Chemistry, vol. XII, pp. 10 and 32 (1932), Longmans, Green and Co., London.

Hansen, Aufbau der Zweistofflegierungen, p. 384 (1943), Edwards Bros., Ann Arbor, Mich. (Copy in Division 3.)