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3,597,158

METHOD OF MAKING DIAMONDS

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ABSTRACT OF THE DISCLOSURE

Synthetic diamonds are made by mixing a carbonaceous material with an alloy consisting of about 95% nickel and 5% beryllium or 45–96% nickel and 55–4% zirconium, and heating the mixture at a pressure of 50,000–60,000 atmospheres to a temperature of 1200–1350° C.

This invention relates to an improved method for converting non-diamond carbon into the diamond form.

U.S. Pats. Nos. 2,947,609, 2,947,610, 2,947,611 and 2,992,900 describe processes for transforming non-diamond carbon into diamond wherein a carbonaceous material such as graphite is mixed with a metal or metal alloy, the mixture placed in a suitable apparatus and subjected to high pressures, and the temperature raised to the melting point of the metal or its alloy. If the mixture contains a metal having a catalytic effect and the temperature and pressure conditions are such that diamond is the thermodynamically stable form of carbon, the non-diamond carbon may convert to diamond.

The rate of breakage of the costly tungsten-carbide anvils generally used to bring the graphite-metal mixture to the required pressures increases exponentially with pressure. Thus, in order to minimize wear and breakage of the anvil and otherwise reduce wear and tear on the apparatus, it is desirable that the pressure at which diamond conversion is effected be as low as possible. Alloys are particularly useful in this respect since they generally melt at a temperature lower than that of the pure metal, and the melting point of an alloy depends somewhat on the pressure of the system. Use of an alloy having the capability to yield satisfactory diamonds at a temperature even only 100° lower, and hence at final pressures 3,000 atmospheres lower, can greatly extend the working life of the anvil.

While in certain cases the use of alloys will not impair diamond production, it is most often found in practice that the dilution of a metal having the desired catalytic activity with another metal results in an alloy of activity inferior to that of the parent catalyst. I have now discovered an improved method for converting non-diamond carbon to diamond utilizing lower melting alloys which permit operation at relatively low temperatures and pressures and, at the same time, result in an increased yield of a superior diamond product.

It is, therefore, an object of the present invention to provide an improved method for converting non-diamond carbon into the diamond form at temperatures and pressures lower than those customarily employed.

It is a further object of the invention to provide a method that gives rise to increased yields of larger diamonds than can otherwise be obtained by operation at such lower temperatures and pressures.

It is yet another object of the present invention to provide a method that results in diamond crystals of more perfect shape at these milder conditions.

In accordance with these objects, the present invention is a method for making diamonds which comprises mixing a carbonaceous material with an alloy consisting of about 95% nickel and 5% beryllium or 45–96% nickel

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and 55–4% zirconium, heating the mixture at a pressure in the range 50,000–60,000 atmospheres to a temperature in the range 1200–1350° C., and recovering the diamonds formed. The synthetic diamonds produced by the method of the present invention have the same utility as natural diamonds, e.g., in ornamental articles and in cutting and abrasive tools.

Graphite remains the most convenient starting material for conversion to diamond. In addition, the process of the present invention is applicable to other carbonaceous materials which undergo this conversion, e.g., amorphous carbon, carbon-containing products of nature, carbon-containing compounds of known and indeterminate structure, and the like.

The process of the present invention may be effected using any suitable apparatus capable of producing the conditions of pressure and temperature required for conversion of the carbonaceous starting material to diamond. A particularly useful apparatus is the hydraulic press described in U.S. Pat. 2,947,610, which description is incorporated herein by reference. In the experiments below, a powdered alloy mixture was prepared and used to fill a hollow graphite cylinder which was then inserted into the apparatus and subjected to the indicated pressure. The temperature was raised to the indicated value by passage of an electric current through the reaction system.

As noted above, diamond making procedures require that the metal or metal alloy be raised at least to its melting point. However, bringing the carbonaceous starting material to the diamond stable region shown in U.S. Pat. 2,947,609, even in contact with a molten alloy of a metal taught by the patent as effective for the purpose, may not always result in the desired conversion. For example, diamonds were not formed from graphite when the following alloys were heated to melting at the indicated conditions:

Approx. alloy composition (percent)	Pressure (atmos.)	Temperature (degrees C.)
33–88 Ni, 67–12 Ge.....	63,000	1,250
60 Mn, 40 Ge.....	60,000	1,250
60 Ni, 30 Ge.....	63,000	1,500
63–80 Ni, 37–20 In.....	60,000	1,390
84 Ni, 16 Mg.....	61,000	1,390
72 Mn, 28 Y.....	60,000	1,320
65 Ni, 35 Pr.....	63,000	1,450
60 Ni, 40 Sn.....	60,000	1,320
70 Ni, 30 Th.....	60,000	1,450
57 Ni, 43 Y.....	61,000	1,250
60 Ni, 40 Zn.....	63,000	1,450

While the diamond stable region in U.S. Pat. 2,947,609 is taught as extending above 50,000 atmospheres and 1200° C., nearly all of the working examples of the patent show pressures upwards of 63,000 atmospheres and temperatures upwards of 1350° C. Only manganese-nickel alloy (60:40 eutectic) appears effective as a conversion catalyst in the 50,000–60,000 atmosphere pressure range and at temperatures below 1350° C.

Using the following nickel-beryllium or nickel-zirconium alloys:

Approx. alloy composition:	Melting point, ° C.
95% Ni, 5% Be	1200
45–96% Ni, 55–4% Zn	1100–1250

I am able to produce diamonds at pressures of 60,000 atmospheres or below and at temperatures of 1350° C. or below. Further, utilization of these alloys in the conversion of carbonaceous material to diamond at such milder conditions unexpectedly gives rise to an increased yield of larger and more perfectly shaped diamonds than obtained by using an operative alloy of U.S. Pat. 2,947,609, i.e., manganese-nickel.

Mixtures of graphite and the test alloy were placed in the graphite cylinder, brought to the indicated pressure

in the hydraulic press, and the alloy melted by passing an alternating current longitudinally through the graphite-alloy mixture. After cooling and depressurization, the diamonds produced were separated and examined. The results are summarized below:

Approx. alloy composition (percent)	Pressure (atmos.)	Temperature (degrees C.)	Run time (min.)	Diamond yield (mg.)
60 Mn, 40 Ni-----	56,000	1,350	6	3.7
95 Ni, 5 Be-----	56,000	1,350	6	32.0
60 Mn, 40 Ni-----	60,000	1,200	3	3.5
50 Ni, 50 Zr-----	60,000	1,200	3	25.8

As shown, the use of nickel-beryllium and nickel-zirconium alloys result in the formation of larger quantities of diamond than obtained with manganese-nickel alloy under comparable conditions. It was also found that use of these alloys produced more nearly perfect diamond crystals as determined visually by comparing the many excellent crystalline faces produced with these alloys contrasted with the few clearly defined crystal faces produced with manganese-nickel. It was further characteristic of these alloys that their use resulted in diamonds larger than those obtained using manganese-nickel alloy as summarized below:

Approx. alloy Composition (percent)	Temperature (degrees C.)	Pressure (atmos.)	Average crystal diameter (mm.)
60 Mn, 40 Ni-----	1,320	56,000	0.2
95 Ni, 5 Be-----	1,320	56,000	0.5
60 Mn, 40 Ni-----	1,200	60,000	0.2
50 Ni, 50 Zr-----	1,200	60,000	0.7

The above-offered non-limiting examples are illustrative of the present invention. Other variations falling within the scope of the invention will suggest themselves to those skilled in the art and my invention is as claimed.

I claim:

1. A method of making diamonds which comprises mixing a carbonaceous material with an alloy consisting of about 95% nickel and 5% beryllium, heating the mixture at a pressure in the range 50,000-60,000 atmospheres to a temperature in the range 1200-1350° C., and recovering the diamonds formed.

References Cited

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