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METHOD OF TREATING METAL POWDERS

Mark N. Fredenburgh, Summit, N. J., assignor to
Radio Corporation of America, a corporation of
Delaware

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The present invention relates to metal powders employed in making an alloy by the powder metallurgy process including at least one refractory metal, and to a method of treating said powders for improving the efficiency of said process.

Alloys including at least one refractory metal in large percentages require recourse to the well known powder metallurgy process for their formation because of the relatively high melting point of refractory metals. It is not feasible to make the alloy by fusing such metals together. According to the powder metallurgy process, powders of the metals to be included in a desired alloy are mixed together, placed in a form of predetermined dimensions and compressed under relatively high pressure to form slugs that are sufficiently coherent for handling. The slugs so formed are then sintered at a temperature sufficiently high to increase their mechanical strength; for it is desirable that the slugs formed by the compression and sintering steps have good coherence and predetermined dimensions for subsequent processing at high temperature. After the high temperature treatment causing the formation of the alloy the slugs of alloy are mechanically worked by swaging after which they are drawn through several successive dies of diminishing aperture size to successively reduce the resultant rods into progressively finer wire formations.

While commercially available metal powders have the desired chemical purity for good results in making the alloy wire, the physical characteristics of such powders, particularly tungsten powder, vary considerably from lot to lot. Since the conditions, such as pressure, temperature and time are usually standardized and fixed in a powder metallurgy process, for the preparation of a specific product variations in the physical characteristics of the powders forming the product result in a sacrifice of efficiency in carrying out the process. One method of measuring the efficiency of a powder metallurgy process involving the manufacture of wire is to compare the ratio of the mass of the finished alloy wire to the mass of the powders employed in making the alloy wire. In many cases, small variations in the physical characteristics of the powders may alter the results to such an extent that the manufacturing efficiency is reduced to a figure of 0-50 per cent.

Weak slugs that fracture during swaging of the alloy may result generally from certain physical characteristics of the metal powders employed in making the alloy by a fixed process.

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One respect in which the physical characteristics of the metal powders vary is in their apparent density. This is distinguished from the absolute density in that it includes in the volume of the powders the pores and spaces between the particles of the powders. As a consequence of this variation in apparent density, when a form or receptacle is used in the practice of the powder metallurgy process having predetermined dimensions for receiving the powders and in which the powders are compressed, the slugs resulting from the application of a definite pressure may be of different sizes and each may have a different apparent density. Also, such variations in apparent density materially affect the efficient loading of the powder receptacle, which is subsequently reflected in the overall efficiency of the process.

Another characteristic of commercial metal powders that varies from lot to lot of such powders is the particle size range and the particle size distribution in the range. Commercial powders such as the powders of tungsten and molybdenum as used in the preparation of alloy wire are usually composed of particles the size of which varies from slightly above 0 to 10 microns in diameter. While this range is satisfactory for a reasonably efficient powder metallurgy process, the particle size distribution in per cent by number sometimes peaks near the lowest limit of the range and sometimes near the highest limit of the range, each of which conditions adversely affects the efficiency of the process referred to.

For example, if the particles peak in size near the lowest limit of the size range in the powder of one metal and peak at a different portion of the range in the powder of another metal to be used in making an alloy, difficulty is experienced in suitably mixing the powders. In order to secure a homogeneous mixture, it is necessary that the particles to be mixed be relatively closely related in size.

Another objection to a peaking of the particle size distribution adjacent the lowest limit of the range is that a slug formed therefrom during the first steps of the powder metallurgy process, is relatively weak and breaks easily during subsequent handling resulting in a waste of metal powder.

On the other hand if the particle size distribution of one metal powder to be mixed with another metal powder, peaks near the highest limit of the range a problem of mixing is also created where the other powder peaks away from the highest limit of the range. Another objection to powders peaking at the highest limit of the

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range is that increased difficulties are encountered during the subsequent stages of the wire process involving swaging and drawing which also result in lowering the efficiency of the process. While it is feasible in the prior art to determine by tests or actual measurements the limits of the particle size range of a powder and the apparent density thereof, there are no simple and satisfactory means available for determining with sufficient accuracy the distribution of particle size fractions so that the performance of the powder can be accurately predicted. The only way this could be determined was by actual performance tests according to which a sample of a powder was subjected to the usual steps of the powder metallurgy process and the value of the powder determined by the efficiency of the process. This is entirely satisfactory for material producing good efficiency but involves considerable cost and required repetition for establishing the value of the material from lot to lot and no disposition of unusable material such as altering or adjusting its physical characteristics to produce satisfactory performance, was available.

This lack in sufficient uniformity in physical characteristics of commercial metal powders, is a serious disadvantage in present manufacturing procedures for making certain alloys by the powder metallurgy process. Such procedures are usually standardized to provide a fixed value of pressures, temperatures and duration of applications. Thus for one value of these manufacturing conditions I have found in making alloys of tungsten and molybdenum that a particle size of the powders ranging from slightly above 0 to 10 microns in diameter and peaking at from 1 to 2 microns is particularly suitable for carrying out an efficient alloying process. It is extremely difficult to obtain commercial powders conforming to a narrow range specification. However, a powder having at least 95 per cent of the particles from slightly above 0 to 3 microns in diameter and with the remainder of the particles not over 10 microns can be readily supplied by the average manufacturer. This powder may or may not be satisfactory in its commercial form for an efficient powder metallurgy process since the particles may actually have a size distribution that peaks at say from slightly above 0 to 1 micron, from 1-2 microns or from 2 to 3 microns. While powders of these characteristics would satisfy the foregoing specification they would not necessarily lend themselves to an efficient alloying and wire making process controlled by the standardized values referred to.

Accordingly it is the object of the invention to improve the efficiency of a powder metallurgy process.

Another object is to provide metal powders of uniform physical characteristics for improving the efficiency of a process for making an alloy therefrom.

A further object is to provide a novel method of treating commercial metal powders composed of definite particle size fractions so as to alter and adjust the particles to produce powders having new definite particle size fractions suitable for use in an efficient alloying process.

Another object is to provide a method of treating commercial metal powders to reduce their particle size range for accommodating the powders to standardized manufacturing procedures with increased efficiency.

A further object is to provide a metal powder that contributes to the formation of coherent

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bodies of increased resistance to fracture during the making of an alloy therefrom by the powder metallurgy process.

Another object is to provide a metal powder having a particle size peaking uniformly in a relatively small portion of the particle size range from lot to lot, for increased economy in the amount of powder employed for making a good alloy therefrom.

A further object is to determine suitability of a metal powder for a standardized alloy making process, by a combination of a relatively simple test and a treatment of said powder based on said test for providing a powder having a particle size peaking at a predetermined portion of the particle size range.

According to one aspect of the invention commercial powders of metal such as tungsten and molybdenum are first tested microscopically to determine the limits of the particle size range thereof. If the test indicates that this range is between 0 and 10 microns, the powder is of a type that can be suitably treated by the method of the invention. No preliminary test is necessary of the apparent density of the powder since the treatment according to the invention renders substantially uniform the apparent density of powders having originally different apparent densities.

Essentially the treatment of commercial metal powders according to the invention for improving the efficiency of a powder metallurgy process for making an alloy of the metals of the powder, includes further limiting the size range of the particles. This further limitation is accomplished by enlargement of the smaller particles and reduction in size of the larger particles. The treatment involves first heating the powders in a reducing atmosphere such as dry hydrogen at a predetermined temperature and for a predetermined time duration. The temperature and time of application are controlled to permit a sintering of the smaller particles to form relatively larger particles. Substantially no change occurs in the larger particles as a result of the heat treatment. The heat treated powder is then subjected to mechanical working to reduce the size of the sintered particles and the size of the agglomerates originally present in the powder. According to the invention a predetermined value for the factors involved in the heat treatment and in the mechanical working will provide a resultant powder having a predetermined particle size range and a predetermined peak of the size distribution in said range.

For best results in a powder metallurgy process involving a specific product it is generally believed the particle size should be limited to a definite range, with the size distribution peaking at some intermediate point in the range. For example, one group of investigators believe a peak at a size of from 4 to 5 microns in diameter is most suitable for making an alloy. Others believe the peaking point should be at other portions of the range. I have found for example that a metal powder having a particle size of from slightly above 0 to 10 microns in diameter and peaking at a particle size of from 1 to 2 microns is well suited for making an alloy with a minimum of powder loss. However, my method of treating commercial metal powders may be practiced with equal advantage no matter what peaking point has been found desirable. For example, a satisfactory powder for improving the efficiency of a powder metallurgy process

may include particles in an overall size range of from slightly about zero to 8 or 10 microns in diameter with the size distribution peaking in a sub range of from 0.5 to 5 microns.

I have practiced my invention in the manufacture of a tungsten molybdenum alloy known as Dowmo and wherein the commercial powders of the metals were supplied in response to a specification calling for a particle size range of from 0 to 10 microns in diameter with 95 per cent of the particles being from 0 to 3 microns in diameter. The 0 value indicates a magnitude that is so small as to be unmeasurable and relatively close to 0 microns.

In carrying out my process for treating commercial powders, the powders supplied in accordance with the foregoing specification are first tested by the use of an electron microscope to determine whether the limits of the size range are as specified.

The next step in the method according to the invention is to heat the metal powders referred to in the preceding paragraph. As actually carried out the heat treatment may involve the use of molybdenum boats 10" long, 1½" wide and 1" deep, into which the metal powders are loaded. The boats are tapped during the loading to pack the powders and the powders are levelled off at the tops of the boats. Each boat contains either tungsten powder, molybdenum

assuming it is desired to mix the powders prior to the heat treatment. As an alternative the powders may be mixed after the heat treatment, in which event, the initial mechanical working is not required.

The resultant powders are found to be of reduced porosity, of increased apparent density and to have a particle size range of from slightly above zero to slightly above 8 microns in diameter with the particle size distribution peaking at from 1 to 2 microns in diameter.

I have found that commercial molybdenum powders are more predictable in their physical characteristics than commercial tungsten powders and consequently satisfactory alloying methods have been practiced in reliance on suppliers' specification for molybdenum powders. In making a Dowmo alloy therefore the treatment according to the invention may be applied solely to the tungsten powder. Since commercial tungsten powders require my treatment to a greater extent than other powders, I have practiced the invention by using tungsten powders from several different sources having a relatively wide difference in their physical characteristics. The following table indicates the particle size range and the peaking point of the particle size distribution in connection with several of such commercial tungsten powders after treatment by my method.

TABLE

Tungsten powders processed according to the invention

Commercial Designation of Tungsten Powder	Particle Size Range in Microns, Percent								
	0-.5	.5-1	1-2	2-3	3-4	4-5	5-6	6-7	7-8
Callite 466H.....	15.6	34.4	37.5	10.8	1.7	-----	-----	-----	-----
N. A. Philips 0-144-1.....	14.9	23.7	46.3	14.9	1.2	-----	-----	-----	-----
Callite 466-D1014.....	9.4	26.3	46.3	14.5	3.0	-----	-----	-----	-----
Callite 482.....	12.0	17.3	54.3	18.5	2.4	.5	-----	-----	-----
N. A. Philips 0-144-2.....	15.8	12.2	44.9	20.8	5.1	.5	-----	-----	-----
R and R 481-1-H12.....	27.2	8.3	22.6	23.9	11.3	.4	.8	-----	-----
R and R 481-1-H2.....	8.4	4.2	28.5	34.6	10.1	4.7	.9	.9	-----
R and R 481-H1.....	7.6	3.3	19.3	36.8	23.1	6.1	3.3	.0	.5

powder, or a mixture of tungsten and molybdenum powders, and the boats are heated individually. Boats containing tungsten powder may be heated for one half hour at a temperature of 1700 degrees C. B. T.±25 degrees C. using an optical pyrometer for temperature measurement. Boats containing molybdenum powder are heated at a lower temperature, to wit 1500 degrees C. B. T. Boats containing mixtures of tungsten and molybdenum powders in the heretofore known relative amounts for providing the alloy known as Dowmo, are heated for one half hour at a temperature of 1600 degrees C. B. T.±25 degrees C. The heat treatments referred to are conducted in dry hydrogen. The resultant product is in the form of slugs of sintered metal.

The next step in the novel method of the invention is to crush the sintered slugs as by means of a jaw crusher so that all of the material is reduced to particle sizes capable of passing through a No. 8 standard testing sieve. The mixture so crushed is then mechanically worked as by ball milling in porcelain jars of one gallon capacity and containing 3000 grams of flint pebbles of from ¾" to 1¼" diameter, rotated at a speed of 60 R. P. M. for about 3 hours.

Before the first step is carried out, the commercial powders may be mechanically worked for about three hours to thoroughly mix the powders,

While the tungsten powders commercially designated Callite 466-H, N. A. Philips 0-144-1, Callite 466-D1014, Callite 482, and N. A. Philips 0-144-2 have a distribution range after treatment according to the invention that peaks at between a particle size of from 1 to 2 microns, which I have found particularly advantageous, the others represent an improvement over the commercial powders. If further improvement is desired in the other powders the heat treatment step temperature may be raised in the direction indicated by the particle size distribution.

I have found that the temperature of the heat treatment according to the invention for tungsten may be carried out with best results from about 1600 degrees C. to about 1750 degrees C. without change in the time of application, although the individual temperatures in this range would change the location of the particle size distribution peak to different points on the size range. An increase in temperature increases the sintering action so that the bond between the particles becomes stronger and the agglomerates are more difficult to break up in the mechanical working step. The temperature therefore should not be high enough to make this bond excessively strong since such particles would be very difficult to break up. Another variable in the heat treatment according to the invention is the time

duration of its application. The time duration may be either increased or reduced from the thirty minute period specified, for either extending or contracting the lower limit of the particle size range of the powder.

While, as has been indicated, it is feasible according to the invention to mix commercial powders of metals which it is desired to alloy, such for example, as the powders of tungsten and molybdenum, prior to the heating step, it is preferable to subject the powders to my novel heat treatment prior to the mixing step. This is for the reason that a more homogeneous mixture is secured when the range of particle size is contracted to reduce the maximum difference in particle size.

The invention may be practiced by employing a relatively wide range in the temperature, and duration of its application, of the heat treatment according to the invention, as well as in the time duration of the subsequent mechanical working operation. For example, a temperature so low and applied for such a short time as to cause only a slight sintering of the powder particles involves a practice of the treatment of the invention. This is for the reason that such slight sintering action accomplishes at least a partial agglomeration of the smaller powder particles and this serves to contract the range of the particle size of the powder. Moreover, a mechanical working may be accomplished according to the invention by subjecting the powder after the heat treatment, or without the heat treatment, to a ball milling operation for a fraction of the three hours specified in the example described in detail before herein. A longer time than three hours may also be used in accordance with the invention.

It is therefore to be understood that the invention in its broader aspects it not to be regarded as limited to the values set forth in the example referred to, but to be directed to a method for reducing the particle size range of metal powders and to cause a predominant portion of the particles to lie in a relatively narrow size sub-range in relation to the overall size range. While, as has been indicated above, I have found a particle size range of from slightly above 0 to 8 microns with the size distribution peaking at a sub-range of from 1 to 2 microns most advantageous in promoting efficiency of a powder metallurgy process, the peaking of the size distribution in another sub-range within the overall size range may also be accompanied by some increase in efficiency in making the alloy over that secured by using untreated commercial powders.

The treatment according to the invention not only results in a desirable size range and size distribution but also in an increase in the apparent density of the particles. This is important as has been previously mentioned in that increased apparent density increases the strength with which the slugs formed of the powder are held together and resist breakage during handling or processing by the powder metallurgy process. According to the invention, the apparent density is first increased during the heating step when a substantial number of the pores in the original particles are closed by the sintering together of the particles. The apparent density is further increased by the mechanical working step during which the particles are rubbed together with an appreciable force. Not only is the apparent density increased in accordance

with the invention but the apparent density of several lots of powders treated by me were found to have a uniform apparent density although the apparent density of the original commercial powders was non-uniform. This uniformity in apparent density is important as has been pointed out before herein.

It will be apparent from the foregoing that I have provided an advantageous treatment as a substitute for a lengthy and uneconomical performance test for assuring suitability of a commercial metal powder for use in an efficient powder metallurgy process.

While I have indicated several ways in which the invention can be carried out it is to be understood that the invention is capable of many modifications as will appear to persons skilled in the art without departing from the spirit thereof and it is desired to include these modifications within the scope of the appended claims.

I claim:

1. In a method of making a relatively strongly coherent tungsten-molybdenum alloy slug from commercial tungsten powder having unpredictable characteristics in respect of particle size distribution and size distribution peak and having relatively small and relatively large particles, and commercial molybdenum powder having a predictable particle size distribution of from slightly above zero to 10 microns in diameter and a size distribution peak of from one-half to five microns in diameter, said method comprising the steps of heating said commercial tungsten powder at a temperature from about 1660° to about 1750° C. for one-half hour to cause said relatively small particles of said tungsten powder to become sintered together in a relatively strongly coherent bond and to cause said relatively large tungsten particles to become sintered in a relatively weakly coherent bond, and subsequently mechanically working said sintered tungsten powder for about three hours to release said larger particles only from said relatively weakly coherent bond, for providing a treated tungsten powder having said predictable particle size distribution and size distribution peak, whereby said treated tungsten powder and said commercial molybdenum powder are adapted to be homogeneously mixed and sintered to provide said strongly coherent slug.

2. Method of making a drawn wire of an alloy of tungsten and molybdenum, from commercial powders of said metals, comprising the steps of separately heating predetermined amounts of said tungsten and molybdenum powders to sinter the particles thereof, separately mechanically working the sintered particles to provide treated powders having smaller size ranges than said commercial powders, mixing said treated powders to form a homogeneous mixture, pressing said mixture to slug form, heating said slug form to sinter the particles thereof to provide a strongly and uniformly coherent body, and drawing said body to relatively fine wire form.

3. Method of making a drawn wire of an alloy of tungsten and molybdenum from commercial powders of said metals, comprising heating a predetermined amount of said tungsten powder at a temperature from about 1660° to 1750° C. for one-half hour, to cause the particles thereof to sinter together in relatively weak bonds, mechanically working the sintered tungsten powder for about three hours, whereby a treated tungsten powder is provided having a reduced amount of

relatively small particles, mixing said treated tungsten powder with a predetermined amount of said molybdenum powder, pressing the resulting mixture to slug form, sintering the slug form to provide a strongly coherent body, and drawing said body to wire form.

MARK N. FREDENBURGH.

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