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SINTERING OF LOOSE PARTICULATE ALUMINUM METAL

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ABSTRACT OF THE DISCLOSURE

A rigid sintered aluminum metal article is made by the method of heating a charge of loose, particulate aluminum metal in a mold in a closed gas-impervious container to a temperature and for a time at which a predetermined portion of the charge, in the range of 15–70 percent by weight, becomes molten. The particulate metal is substantially free of oxide films thicker than 700 Å. Formation of a rigid integral article is substantially assured if the metal is protected from rapid oxidation during at least two of the steps of making the particulate metal, storing the same and sintering charges thereof.

This invention relates to the sintering of loose particulate aluminum metal to form a rigid article. It is more particularly directed to the preparation of highly porous aluminum metal articles, such as articles useful in the form of oil filters and air filters for internal combustion engines, as well as more impervious solid articles having higher structural strength.

For the purposes of the specification and claims, aluminum and aluminum-base alloys containing at least 70 percent by weight of aluminum are hereinafter referred to as aluminum metal.

Conventional preparation of sintered articles from powdered metals such as brass or bronze powder is carried out by first cold pressing the loose particulate metal to form a compact and heating the compressed particulate metal just sufficiently to obtain sintering with substantially no melting of the metal. However, the conventional methods have been singularly unsuccessful when applied heretofore to the sintering of particulate aluminum.

It is therefore a principal object of the invention to provide a method for sintering loose particulate aluminum metal.

Another object of the invention is to provide a method of sintering loose particulate aluminum metal which avoids the necessity of making a compact, as by a cold pressing operation, before sintering.

Another object of the invention is to provide a method of avoiding heavy oxide films in the preparation and sintering of particulate aluminum metal.

Yet another object of the invention is to provide a method of making a sintered article of aluminum metal having a preselected degree of porosity.

A further object of the invention is to provide a method of obtaining an improved, rigid, porous article of aluminum metal having desirably high strength properties.

Another object of the invention is to provide a method of preparing a rigid, coherent substantially fully densified article in the sintering of particulate aluminum metal.

Still another object of the invention is to provide a method of preparing a sintered article of aluminum metal having a reproducible, preselected degree of porosity and yet uniformly good structural strength from article to article.

These and other objects and advantages of the present invention are obtained by providing substantially or sufficiently oxide-free, loose, particulate aluminum metal pre-

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pared under properly controlled conditions, as herein-after more fully described; placing a charge of such particulate aluminum metal in a mold in a container which is closed to exclude oxidizing atmosphere; and heating the charge of loose particulate metal in a dry, substantially inert atmosphere and to a temperature and for a time that at least about 15 percent by weight of the charge becomes molten, but complete melting of the charge does not take place; and thereafter permitting the partially melted metal to cool and solidify, thereby forming a strong, rigid, aluminum metal article.

The presence of an oxide layer on the surface of particulate aluminum metal interferes with the sintering thereof by forming a barrier layer between the particles which is difficultly bridged even by molten metal during the sintering process. Although it is desirable to minimize oxidation to insure optimum sinterability, appreciable oxidation can be tolerated without destroying the sinterability of the aluminum pellets. The critical oxide layer thickness which must not be exceeded if good sintering is to be obtained consistently appears to be about 700 Å. Since oxidation can occur during each of the steps of preparing, storing and sintering the particulate metal, it is imperative that each step be controlled so that the final oxide thickness at the moment fusing and bonding is to occur is below the critical thickness.

Aluminum metals which may be desirably transformed into sintered articles by the present process include aluminum and any of the aluminum-base alloys containing at least 70 percent by weight of aluminum. Preferably, the alloy has a fairly wide solidification range, e.g., a range of at least 25 Fahrenheit degrees. Examples of aluminum alloys having a suitable melting range and desirably employed in the present process are the binary aluminum alloys containing, respectively, about 5 percent of copper (AL-5Cu), about 2 percent of silicon (AL-2Si) and about 8 percent of magnesium (AL-8Mg).

The foregoing aluminum alloys all exist as homogenous melts at conventional casting or atomization temperatures (1250 to 1400° F.). A different type of aluminum metal advantageously sintered according to the invention is the aluminum alloy which does not exist as a homogenous melt at conventional casting or atomization temperatures. Such alloys form separate phases, the molten phase, and a discrete solid phase. Since these alloys settle or stratify during treatment of the melt before casting, they are not castable as such and particulate metal containing the intended constituents cannot be prepared by this route.

To bring such an alloy to particulate form without loss of alloying constituents, it is therefore necessary to atomize the alloy at a temperature at which the alloy is homogenous.

These alloys are hereinafter referred to as normally heterogeneous alloys. The normally heterogeneous alloys advantageously treated according to the invention consist of at least 20 percent by weight, and more preferably at least 50 percent by weight of aluminum or homogenous aluminum alloy, and the balance a component which is insoluble at normal casting temperatures, or the balance may be an intermetallic compound with aluminum which is insoluble in the melt at normal casting temperatures. Examples of insoluble components which may be used are silicon and beryllium. Examples of other suitable elements and the intermetallic compounds they form are listed as follows:

| Element: | Intermetallic compound |
|----------|------------------------|
| B | AlB ₂ |
| Fe | FeAl ₃ |
| Mn | MnAl ₆ |
| Zr | ZrAl ₃ |

Examples of specific suitable alloys of this type are: (1) 30 percent by weight of silicon, the balance aluminum, and (2) 20 percent by weight of manganese, and the balance aluminum.

Homogenous aluminum metal may be obtained in particulate form by machining solidified massive aluminum metal, or, by disc or jet atomizing the molten metal, as well understood in the art, in a dry, preferably non-oxidizing atmosphere. Examples of a suitable atmosphere are dry natural gas, air having a dewpoint of 20° F. or lower (i.e., containing less than 0.3 percent by volume water vapor), or a vacuum. The machined chips or atomized pellets may be of any size or fineness practical for making a useful sintered product. Atomized pellets are usually finer than 20 mesh when formed. Generally, the machined chips should not have a maximum dimension greater than about 1/2 inch. Disc or jet atomizing is the preferred method of preparing particulate aluminum for the method of the present invention.

The alloys which are only moderately active, such as the aluminum-copper or aluminum-silicon alloys, may be prepared in particulate form either by machining the massive metal, i.e., by mechanical comminution, or, by one of the atomizing processes. It is highly desirable that aluminum alloys containing constituents such as magnesium or lithium are obtained in particulate form by disc or jet atomizing the molten metal in a dry, substantially inert atmosphere.

As indicated above, atomizing is the preferred method of preparing particulate homogeneous aluminum alloys, although mechanical comminution methods may be employed if oxidation is properly controlled. Al-5Cu and Al-8Mg were found to sinter much more readily when the particulate metal was obtained by atomizing molten metal rather than by machining solidified metal. Atomized pellets of both said alloys sintered well even after exposure to the ambient atmosphere for several months at room temperature. Machined chips of Al-5Cu which had been encapsulated in glass shortly after preparation sintered quite adequately. Machined chips of the same alloy which were not encapsulated were found not sinterable after exposure to the ambient atmosphere for one week at room temperature. Chips formed of Al-8Mg and machined in air were found not to sinter at all regardless of storage conditions. Mechanical comminution methods are more acceptable when carried out in an inert atmosphere.

The atomizing atmosphere employed in disc atomizing or the stream of gas employed in jet atomizing should be as inert as possible. Contamination of the atmosphere or jet with oxygen is not per se serious, but the presence of water vapor is to be avoided as much as reasonably possible. Aluminum pellets prepared in an atmosphere or jet of natural gas or argon, or even in air, can be readily sintered if the water vapor content of the gas in each case is kept quite low. In each case, to assure sinterability, the water vapor content should desirably be kept below about 0.15 percent by weight water. Collecting the pellets in water during the atomizing process also tends to impair sinterability. Pellets obtained by atomization in air containing 0.44 percent water were sinterable when the pellets were collected dry, but were not sinterable when the pellets were collected in water.

A highly inert gas is used for atomizing when spherical pellets are desired, while air is used for producing irregularly shaped particles. Because greater oxidation occurs when air is used as the atomizing gas, somewhat closer control is required during subsequent storage and sintering steps than would be required for pellets atomized in a highly inert gas such as argon.

In atomizing the normally heterogeneous alloys, the temperature of the melt is maintained sufficiently high for the melt to be entirely molten and homogenous during the atomizing operation. Temperatures of 1400 to 2000° F. are employed, though still higher temperatures could be used if container materials resistant to the action of high

temperature molten aluminum were available. At temperatures above about 1600° F. the reactivity of the molten aluminum is greater so that it is desirable, when atomizing molten aluminum at temperatures above about 1600° F. to take greater care to avoid water vapor or other reactive gas in the atomizing gas and environment.

In each of the particle making, storing, compacting and sintering operations described herein, it is particularly important to avoid reaction of the aluminum with water vapor. The effect is not completely understood, but it may be due to the formation of a thicker bulkier oxide than is had on reaction with oxygen.

If the particulate metal is not used immediately after its preparation to make a sintered article, the metal is stored in a dry air environment to minimize oxidation. The particulate metal may be stored more protectively in a dry inert gas atmosphere, if desired. Aluminum pellets can be successfully sintered several years after being prepared by atomizing if they are properly stored. Storage in air is satisfactory if low humidity is maintained. Al-2Si pellets atomized in natural gas have been found not sinterable if exposed at 100° F. for more than about seven days in air saturated with water vapor. The sinterability of Al-2Si pellets atomized in natural gas is not adversely affected on exposure of the Al-2Si pellets to the atmosphere for 20 days if the storage temperature does not exceed about 75° F., or, if most of the water vapor is removed from the air. If the Al-2Si pellets have been atomized in air, the critical exposure time under conditions of 100° F. temperature and about 100% relative humidity is reduced to less than one day. Pellets of Al-8Mg exposed to the atmosphere at 75° F. and 100% relative humidity are not sinterable after four days' exposure. Pellets of Al-5Cu have been successfully sintered after being held in storage in an air conditioned room for a month under conditions of about 40 to 60% relative humidity, but the same alloy pellets usually become unsinterable in about five to seven days on being exposed to 90% relative humidity or higher at room temperature as found in an open warehouse on a rainy day.

Desirably, the particulate metal is stored in a dry atmosphere of natural gas, nitrogen, helium or argon containing less than about 0.5 percent by volume of oxygen and less than 0.3 percent by volume of water vapor, the atmosphere being substantially free of gases such as carbon dioxide and chlorine. Metal in the form of atomized pellets may be stored in dry air containing less than about 0.3 percent by volume of water vapor. Generally, it is sufficient and practical to simply store the pellets in a closed receptacle such as a fiberboard drum. The use of a desiccating agent and an inner plastic liner for the receptacle provides additional protection from ambient atmospheric moisture.

In carrying out the sintering process of the invention, loose particulate aluminum metal is provided which has been prepared so as to be substantially free of thick oxide films, as described above, and which, if not freshly made, has been properly stored. The loose particulate metal is placed in a mold which is desirably sealed off and placed in a furnace. If desired, the mold may be placed in a container which is adapted to be filled and/or flushed continuously with an inert gas during the sintering period. The container is placed in a furnace of appropriate size and heating capacity. It is also possible to use a properly closed furnace itself as the container and to flush the interior with an inert gas and to maintain an inert atmosphere in the furnace during the sintering step.

A critical aspect of the sintering step is the selection of the material for construction of the mold, since most materials give off oxidizing gases, e.g., H₂O vapor and O₂, when heated, and since molten aluminum has a strong tendency to alloy with or bond to many materials. The mold may be constructed of material which adsorbs gases but which can be readily degassed as by heating and/or subjecting the material to a high vacuum. Glass; steel

sprayed with refractories such as $\text{MgO} \cdot \text{Al}_2\text{O}_3$, ZrO_2 , Cr_2O_3 and ZrSiO_4 ; "Pyroceram" brand ceramic; steel coated with porcelain enamels which are not significantly attacked by molten aluminum; and pyrolytic graphites have been found to be suitable for making molds. Oxidized steel, steel sprayed with Al_2O_3 , and conventional graphite tend to liberate oxidizing gases and may be used as mold material only if they are properly degassed. Stainless steel and freshly abraded steel do not cause oxidation of molten aluminum but are unsuitable as mold materials because they tend to alloy with molten aluminum and, consequently, the sintered article becomes firmly bonded to the mold.

For the purposes of the specification and claims, the term non-bonding is used in connection with the tendency, or the lack thereof, of molten aluminum metal to bond to the mold walls.

Of course, if it is desired to form a sintered metal section or element adhering to a surface of an article as an integral part thereof, e.g., a filter formed within a tubular article, the mold selected is, at least in part, the surface or surfaces of the article itself, and the article selected is formed of, or coated with, a material or metal or alloy to which the particulate aluminum metal is capable of alloying or otherwise bonding during the sintering step referred to hereinabove. Thus, a filter permanently bonded to a surrounding tubular matrix may be prepared by the present method.

The sintering atmosphere must be carefully controlled to avoid excessive oxidation of the particulate aluminum metal because the metal is maintained at a high temperature for a relatively long time in the sintering process. Such gases as carbon dioxide, oxygen, water vapor and chlorine are to be avoided. While natural gas may be used with proper precautions, the gases of choice for filling the sintering chamber are helium, argon, hydrogen, nitrogen, methane, ethane and mixtures thereof. Or the chamber may simply be evacuated to a pressure below about 10 millimeters Hg and preferably below about 1 millimeter Hg. Water vapor in the sintering atmosphere is particularly deleterious and if present should always be removed to a level below about 0.3%, by volume.

So long as the sintering atmosphere used is substantially inert to the charge of particulate aluminum metal, the atmosphere need not be static, but may be a flowing atmosphere if desired. Thus, if it is not feasible to seal the container during sintering, it is sufficient to maintain a positive pressure, inside the container, of substantially inert gas.

If it is necessary or advantageous for some other reason to carry out sintering in the presence of a somewhat reactive oxidizing gas such as air, the particulate aluminum may still be satisfactorily sintered if the container is preselected to provide a limited amount of atmosphere, viz., less than about 40 percent by volume headspace above the charge of metal being sintered. Al-2Si pellets, for example, can be satisfactorily sintered even in an oxygen atmosphere maintained at ambient atmospheric pressure if the ratio of the volume of the pellet bed to the free space above the pellets in the container is 5:3 or greater, but cannot be sintered if the ratio is smaller, for example, 1:1. The amount of oxidation which can be tolerated in the sintering step depends on how much oxidation has already occurred during preparation of particulate metal and in storage.

If it is necessary to carry out the sintering in the presence of an oxidizing gas, the particulate aluminum can be sacrificially protected by a more active metal which serves as a "getter." Thus, it has been found possible to sinter atomized pellets of aluminum metal in either a loosely or tightly covered large container which contains magnesium pellets in one or more separate compartments or receptacles. Care must be taken to avoid contact between the magnesium and aluminum pellets which readily fuse together at sintering temperatures.

A highly important aspect of the invention is maintaining control over the extent of melting of the charge during sintering. Preparation of the sintered particulate aluminum metal article of the invention requires some concurrent melting during the sintering step in order to obtain an article with desirably high strength properties. In preparing a porous article, the particulate metal in the sintering container is heated to a temperature and for a time that only partial melting will take place. Melting a portion of the pellets equal to at least 15 percent of the weight of the pellets is required to obtain good bonding between the pellets so as to form an article with good structural strength.

The degree of porosity of the sintered article is controlled by the extent to which the particulate metal is melted during sintering. In the preparation of highly porous articles, as in the preparation of filters, the desired amount of melting is in the range of about 15 to 30 percent. If somewhat less porosity is desired, or if higher strength is required, the amount of melting may be increased to as high as about 70 percent by weight of the pellets. More than this amount of melting is not particularly harmful if the material employed is a homogenous alloy but there is no particular advantage in completely melting the charge. In the case of the heterogeneous alloys, melting more than 70% by weight of the pellets is disadvantageous or deleterious because of the tendency for any unmelted metal to segregate as by floating or settling in one part of the melt and also because of the tendency for compositional changes to take place in the molten phase during subsequent solidification of the molten metal on cooling.

More preferably, the volume of metal melted during sintering does not greatly exceed the volume of the void spaces between the particles of metal in the charge prior to sintering. Thus, the amount of metal melted in a charge of uncompressed substantially spherical pellets should not greatly exceed about 35 percent by weight of the charge, while the amount of metal melted in an uncompressed charge of either machined chips or irregularly shaped pellets should not greatly exceed about 50 percent by weight of the charge.

The time and temperature required for the requisite amount of partial melting can be determined from an appropriate phase diagram, or the determination may be made by direct experiment. Typical temperatures employed are: 1120° F. for Al-2Si; 1100° F. for Al-5Cu; and 1080° F. for Al-8Mg. Sintering times of about 4 hours are suitable at these sintering temperatures. Adequate sintering may be carried out for shorter times at somewhat higher temperatures. Generally, temperatures above about 1250° F. are to be avoided because of the increased tendency for oxidation or excessive melting to take place. If an oxidizing gas is present in the atmosphere within the sintering container and if melting is not carried out quickly, oxidation may give rise to sufficient barrier material between particles to prevent adequate bridging and the sintered metal will not be coherent, i.e., will not have sufficient structural strength to be useful.

To make temperature control simpler, i.e., to permit rapid and adequate melting without causing melting of the entire charge, it is often desirable to use an aluminum alloy having a melting range of at least 25 Fahrenheit degrees and more preferably at least 50 Fahrenheit degrees. Then on maintaining a temperature within the melting range, but at least about 3 to 10 degrees below the upper end of the melting range, one can be reasonably sure that not all of the metal will become molten.

The amount of melting should be readily controllable, particularly in the preparation of a filter having a preselected degree of porosity. Too little melting will not provide adequate bonding while too much melting will not provide adequate porosity in the sintered article. It should be understood that the selection of an aluminum alloy with a wide melting range is not mandatory because it is

possible to sinter unalloyed aluminum with the desired degree of porosity on maintaining close control of temperatures and times.

After liquid phase sintering of particulate aluminum metal according to the invention, the temperature of the sintered article should be decreased below the solidus temperature for a time sufficient to completely solidify the molten phase before the sintered article is removed from the mold.

If a high density, high strength, substantially non-porous article is desired, the present method is readily adapted to the purpose by the inclusion of a pressing step in which the particulate material is compressed prior to the sintering step. Such pressing may be carried out at temperatures classified as cold or warm.

Cold pressing is generally carried out at room temperature and especially at a temperature below about 100° F.

Warm pressing is carried out at temperatures in the range of about 300 to 500° F. In compressing the particulate metal at temperatures below about 500° F., it is usually desirable to employ particulate material having irregular shapes as contrasted to spherical particles since the spherical particles are difficultly compressed to cause sticking together. Achievement of a specific gravity of at least 80% of the theoretical value for aluminum is desirable.

Since the preferred method of preparing particulate aluminum material according to the invention is by atomizing the alloy, when irregularly shaped particles are desired for cold or warm pressing, it is necessary to carry out the atomizing process in such a manner as to provide particles of irregular shape. This is accomplished by atomizing the aluminum metal with a gas jet, in the case of jet atomizing, or in an environment, in the case of disc-atomizing, wherein the gas jet or environment used contains sufficient oxygen to cause the pellets to have an irregular shape. It is still highly desirable that the atomizing gas or environment is dry, i.e., contains less than about 0.3 percent by volume water vapor.

Sintering may be omitted altogether upon using a hot pressing operation. In carrying out hot pressing, the particulate material is pressed under the requisite conditions of temperature and compression pressure to obtain a substantially fully densified article during the hot pressing step. During such hot pressing, a fully bonded integral article is obtained and a subsequent step of sintering is not employed. Such hot pressing is carried out by pressing a heated charge of the particulate aluminum metal in a press, e.g., in the preheated container of an extrusion press having the die opening blanked off by a cover plate and preferably under an inert atmosphere. Sufficient pressure is applied to bond the particles together and to substantially eliminate the void spaces. Temperatures of the order of 900 to 1000° F. are generally suitable, although some of the softer alloys may be hot pressed at a temperature as low as 800° F. Temperatures at which the alloy normally melts are to be avoided in hot pressing. Sufficient pressure is employed in hot pressing to bring the specific gravity of the charge to at least about 2.43 (90% of theoretical) and more preferably to at least 2.6 grams per cubic centimeter.

Because of the tendency of aluminum metal to bond to other metals at elevated temperatures, it is generally desirable in preparing the more fully densified article to subject the metal charge first to a cold or warm pressing operation in which most of the desired increase in density is achieved. By so doing, most of the movement of the charge within the container takes place at a temperature where lubricity is no problem. The partly compressed material is then subjected to a hot pressing operation in which the desired densification is completed under conditions at which the charge and the ram of the press are subjected to a small amount of movement so that lubri-

cants have a chance to operate satisfactorily at the more elevated temperature.

Generally, in carrying out both hot and cold pressing, the specific gravity of the charge of particulate metal is brought to at least about 2.16 in the cold pressing step, and the further densification is carried out by hot pressing, usually at about 900° F.

One of the big advantages of any pressing operation according to the present invention is that the densified material is obtained either directly in pressing or subsequently on sintering and since any substantial reduction in cross-section, which is a necessary consequence of an extrusion step, is avoided, it is easier to make large articles according to the present method.

Particulate material employed in making a high density article must be protected from oxidation as indicated hereinabove during the preparation of the particulate metal, during any intermediate storage and certainly during sintering following cold pressing or in a hot pressing operation after cold pressing. Hot pressing can be carried out in air but in the case of the more active alloys, it is preferred to provide a protective atmosphere during the pressing operation and more critically while heating the charge before the compression step.

The sintered or hot pressed article of the invention may be worked, if desired, to obtain a product having the advantages of pellet metallurgy. Methods such as rolling, extruding, forging, impact extrusion, sizing and coining may be employed. Generally, in making a worked product, the higher density metal is used to avoid cracking, especially if the material is to be rolled or forged. A surprising property of the sintered or hot pressed article, however, is the ease with which voids therein are removed on working the article.

On sintering or hot pressing a particulate heterogeneous aluminum alloy according to the invention, a rigid article is obtained which consists of an aluminum or aluminum alloy matrix having very intimately, finely and uniformly dispersed therethrough, a volumetrically coextensive insoluble phase consisting of an insoluble constituent or an insoluble aluminum intermetallic compound. The matrix phase comprises at least 20 percent by weight of the rigid article and consists of aluminum or aluminum base-alloy as defined hereinabove.

Such articles tend to be brittle and very hard, and being resistant to wear and having a high compression yield strength modulus, are desirably used for bearing inserts.

For the purposes of the specification and claims, the term aluminum metal is to be understood to include the heterogeneous alloys described hereinabove.

An important and surprising aspect of the present invention is the further discovery that good sintering may still be obtained if careful control is exercised to avoid oxidation of the aluminum metal in at least one of the two steps (1) making particulate aluminum metal and (2) sintering, where the particulate aluminum metal is sintered within 24 hours after its preparation.

Good sintering may also be obtained if careful control is exercised to avoid oxidation of the aluminum metal in at least two of the three steps of (1) making particulate aluminum metal, (2) storing the particulate aluminum metal, and (3) sintering the particulate aluminum metal. The careful control to avoid oxidation in the several steps is already described hereinabove.

It is especially important to avoid oxidation by contact of the aluminum metal with water or water vapor. Oxidation by reaction with H₂O is more deleterious to good sintering than an equivalent amount of oxidation by reaction with oxygen or air.

The minimum requirements of care, to avoid too much oxidation at the several stages or steps of the present method, which still permit good sintering when the operation falls below this standard of care in only one step

are illustrated as follows, it being understood that equivalent conditions may also be employed:

Jet atomizing: A stream of molten aluminum metal is broken into fine particles by hitting it with a jet of natural gas which contains less than 500 parts per million (by weight) oxygen and less than 0.3 percent by volume water vapor.

Storage: The particles of aluminum are stored in a closed container at an ambient room temperature below 100° F. and at a relative humidity less than about 90%.

Sintering: The particulate aluminum metal is placed in a sealed glass container which is either filled with an inert gas such as argon or evacuated. The container and contents are heated for 4 hours at a temperature which will bring about melting of 15 to 70 percent of the metal.

The following examples of the invention are illustrative thereof and the invention is not to be considered limited thereto.

Example 1

A melt of aluminum alloy consisting of about 5 weight percent of copper and the balance commercial aluminum was prepared by melting the requisite proportions of aluminum and copper together in a gas-fired setting. The molten alloy was fed through conventional jet atomizing

lets were collected in a dry atmosphere while a separate additional part of the pellets were chilled by collection in water. The pellets collected in water were separated from the water within less than an hour and the pellets were then dried, carefully washed with a volatile chlorinated hydrocarbon and dried.

In each case, the atomized pellets were stored in a closed container at ambient room temperature and under atmospheric conditions until sintering was attempted.

Sintering in each case was attempted by sealing the charge of pellets in a glass container having less than 40 percent by volume headspace, the headspace being evacuated or filled with an inert gas, usually argon. The sealed container was heated for 4 hours at a temperature calculated to produce 15 to 30 percent melting of the charge. Whether or not adequate sintering occurred to produce a strong, rigid porous article was determined by visual and microscopic examination of the sintered article. Sintering is considered adequate if the pellets are bonded to each other sufficiently that the outer layer of the article cannot be rubbed off by hand. Another practical test is to attempt to break, by hand, a 3/4-inch diameter rod shaped test bar about 3 to 4 inches long. The article is strong enough if it cannot be broken on trying to bend it by hand.

The conditions of atomizing the metal and collecting the pellets and the results of the sintering tests are listed in the following Table I.

TABLE I

| Test No. ¹ | Atomizing Gas | Impurities Added | | Collection | | Sinterable |
|-----------------------|---------------|----------------------------------|---------------------|------------|---------------------|------------|
| | | Vol. percent of H ₂ O | P.p.m. (wt.) Oxygen | Dry | In H ₂ O | |
| 1 | Natural Gas | 0.028 | 100 | X | | Yes. |
| 2 | do. | 0.44 | 400 | X | | Yes. |
| 3 | do. | 0.44 | 400 | | X | Yes. |
| 4 | Argon | 0.44 | 2,000 | X | | Yes. |
| 5 | do. | 0.44 | 2,000 | | X | Yes. |
| 6 | do. | 0.7 | 5,000-10,000 | X | | No. |
| 7 | do. | 0.7 | 5,000-10,000 | | X | No. |
| 8 | Air | 0.14 | (2) | X | | Yes. |
| 9 | do. | 0.44 | (2) | X | | Yes. |
| 10 | do. | 0.44 | (2) | | X | No. |
| 11 | do. | 0.8 | (2) | X | | No. |
| 12 | do. | 0.8 | (2) | | X | No. |

¹ Tests numbered 6, 7, 10, 11 and 12 are comparison tests.

² None added.

equipment and pellets of the aluminum alloy were formed in a dry, natural gas atmosphere in an enclosed container. The pellets of aluminum alloy were then stored in a dry atmosphere for about ten days before 25 grams of pellets were placed in a glass tube 10 millimeters in diameter and sealed at one end. The glass tube containing the metal charge was evacuated, flushed with argon and evacuated, before the glass tube was sealed. The glass-enclosed charge was then placed in a heating oven and heated at 1100° F. for 4 hours. After the charge had cooled, it was removed from the glass tube. The loose, particulate aluminum metal had been converted to a rigid, porous cylindrical article displaying substantial mechanical strength properties (about 1500 to 7000 pounds per square inch tensile yield strength), typical of a well sintered article of aluminum-base alloy.

Example 2

A series of experiments was carried out illustrating the leeway in control of oxidation in one step (here, atomizing), so long as the minimum conditions or better are met in the other two steps of the process. Aluminum-silicon binary alloy containing about 2 percent by weight of silicon, was melted and jet atomized with a stream of natural gas, argon or air. In the several runs, various concentrations of water vapor and/or oxygen were added to the atomizing gas. In most atomizing runs, part of the pel-

Examination of the results shows that substantial amounts of water vapor and oxygen may be tolerated in the atomizing step if at least minimum conditions are maintained during storage (if any) and sintering.

In a similar series of experiments, in which there was employed the aluminum-magnesium binary alloy containing about 8 percent by weight of magnesium, the sensitivity of this aluminum-magnesium alloy to atomizing conditions was found to be substantially the same as for the above aluminum-silicon binary alloy.

Example 3

A series of experiments was carried out illustrating the leeway in control of oxidation in one step (here, storage), so long as the minimum conditions or better are met in the other two steps of the process. Aluminum-silicon binary alloy containing about 2 percent by weight of silicon, was melted and jet atomized with a stream of natural gas containing less than 400 p.p.m. oxygen and less than 0.15 percent by volume water vapor. The pellets were collected in a dry atmosphere. The pellets were divided into several portions which were stored under various conditions and lengths of time before sintering was attempted. The conditions employed in the sintering attempts were those described in Example 2.

A portion of pellets stored in an open-top container for 20 days in a room in which dry flowing air was maintained

at 95° F. was sinterable. Another portion of pellets which was stored in an open-top container for 3 days in a room maintained at 100° F. and 100% relative humidity was still sinterable. However, a portion of pellets stored in an open-top container for 7 days under conditions of 100° F. and 100% relative humidity was not sinterable.

In additional experiments, the aluminum-silicon binary alloy containing 2 percent by weight of silicon was jet atomized in a stream of air containing 0.14 volume percent water vapor. One portion of the pellets was stored for 20 days in an open container in a room in which dry flowing air was maintained at 95° F. Another portion was stored 24 hours in an open container in a room maintained at 100° F. and 100% relative humidity.

Sintering of each portion was attempted under the sintering conditions described in Example 2. The pellets stored in dry flowing air were sinterable, showing these conditions for this length of time to be the equivalent of storage in a closed container.

The pellets stored at 100° F. and 100% relative humidity for one day were not sinterable, whereas other pellets of the same alloy atomized by a jet of dry natural gas and stored at 100° F. and 100% relative humidity for 3 days were sinterable.

Similar tests carried out with the aluminum-copper binary alloy containing about 5 percent by weight of copper yielded substantially the same results.

Example 4

A series of experiments was carried out illustrating the leeway in control of oxidation in one step (here, sintering) so long as the minimum conditions or better are met in the other two steps of the process. Aluminum-silicon binary alloy containing about 2 percent by weight of silicon was melted and jet atomized with either a stream of natural gas or of substantially dry air. The so-obtained pellets were stored in a closed container at ambient room temperature and atmospheric conditions until sintering attempts were made.

Portions of the atomized pellets were placed individually in respective glass containers and the containers sealed. In the several experiments, the headspace volume was varied, and differing gases were employed in the headspace. The approximate pellet bed volume was determined after tapping the container lightly and the volume measured included the voids between the pellets. The gas in the headspace, the relative headspace volume and the sinterability are listed for each experiment in Table II as follows:

TABLE II

| Test No. | Gas In The Container | Ratio, Pellet Bed Vol. to Headspace | Sinterable |
|-----------------|--|-------------------------------------|------------|
| 13 | None (Vacuum) | | Yes. |
| 14 | Argon | 3:1 | Yes. |
| 15 | Argon plus 3% by volume H ₂ O vapor | 3:1 | Yes. |
| 16 | Natural Gas | 3:1 | Yes. |
| 17 | Air | 3:1 | Yes. |
| 18 | CO ₂ | 3:1 | Yes. |
| 19 | O ₂ | 7:1 | Yes. |
| 20 | O ₂ | 3:1 | Yes. |
| 21 | O ₂ | 5:3 | Yes. |
| 22 ¹ | O ₂ | 1:1 | No. |

¹ Test number 22 is a comparison test.

The data shows that a headspace volume up to 40 percent of the total container volume, the headspace being filled with a gas no more active than oxygen, may be suitably employed so long as at least minimum conditions are met in atomizing and storing the aluminum alloy pellets.

In a similar series of tests using the aluminum-copper binary alloy containing about 5 percent by weight of copper, similar results were obtained, sensitivity to oxidation being very little different than the aluminum-silicon binary alloy.

The method of the invention having been thus described, modifications thereof will at once be apparent to those skilled in the art, and the scope of the invention is to be considered limited only by the hereinafter appended claims.

We claim:

1. The method of making a rigid aluminum metal article which comprises: heating a charge consisting essentially of loose, particulate aluminum metal in a mold in a closed gas-imprevous container to a temperature and for a time at which from about 15 to 70 percent by weight of the charge becomes molten, the particles of said loose particulate aluminum metal being substantially free of oxide films thicker than about 700 Angstroms.

2. The method as in claim 1 in which the temperature and time are pre-selected whereby a predetermined extent of melting is obtained and a porous article is formed.

3. The method as in claim 1 in which the mold employed is formed of material which bonds to the aluminum metal during said heating.

4. The method as in claim 1 in which the mold employed has an inner surface formed of non-bonding material with respect to aluminum metal, and said inner surface is a gas-free surface.

5. The method as in claim 1 in which a sealed glass container is used which serves as both the said mold and the said closed container.

6. The method as in claim 1 wherein the loose particulate aluminum metal comprises at least one aluminum-base alloy having a melting range of at least 25 Fahrenheit degrees.

7. The method as in claim 1 in which the loose particulate aluminum metal comprises a heterogeneous alloy, said heterogeneous alloy consisting of at least 20 percent by weight of homogeneous aluminum alloy and the balance a metal phase which is insoluble at normal casting temperatures, said insoluble metal phase being completely miscible with said homogeneous aluminum alloy at an elevated temperature above the normal casting temperatures.

8. The method as in claim 1 in which said loose particulate aluminum metal (1) is prepared in particulate form in a substantially non-oxidizing atmosphere, (2) is thereafter stored substantially free from contact with active oxidizing gases and (3) is substantially protected from oxidation during subsequent sintering.

9. The method as in claim 8 in which each of the steps of preparing in particulate form, storing and sintering of the particulate aluminum metal is carried out in an atmosphere containing less than 0.3 percent by volume of water vapor.

10. The method as in claim 1 in which protection from contact with active oxidizing gases is carried out in only two of the three steps of preparing the loose particulate metal, storing such particulate metal and heating the said charge thereof.

11. The method as in claim 1 in which the charge of loose particulate aluminum metal is subjected to a compacting pressure at a temperature below about 500° F. prior to said heating.

12. The method as in claim 1 in which the atmosphere in the container is selected from the group consisting of nitrogen, hydrogen, helium, argon, natural gas, methane, ethane and mixtures thereof, any of the foregoing gases in addition to oxygen or air in an amount which does not occupy more than 40 percent by volume head space above the charge in the container, and any of the foregoing gases at a pressure below about 10 mm. of mercury pressure.

13. The method as in claim 1 in which the said particulate metal is heated in the presence of a charge of loose particulate magnesium in the said container, both said particulate metals being exposed to the same atmosphere.

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