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OTHER REFERENCES

PRODUCT ENGINEERING, May 30, 1960, p. 61 relied on copy in 75/200 F.

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[54] PRODUCTION OF REINFORCED COMPOSITES 6 Claims, 2 Drawing Figs.

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 75/200, 75/208; 164/62, 164/119, 164/41, 164/138,
 164/125, 164/97, 164/100, 164/105; 29/191.2
 [51] Int. Cl. **B22d 17/26**
 [50] Field of Search 75/200F,
 208(Inquired); 29/191.2; 164/61, 62, 119, 41, 138,
 125, 97, 100, 105

[56] References Cited

UNITED STATES PATENTS

3,084,421 4/1963 McDaniels et al. 75/200F

ABSTRACT: Alumina whiskers or other fibrous reinforcing agents can be completely infiltrated by an aluminum matrix without the necessity for special whisker precoating operations. The infiltration is accomplished by packing the fibrous skeleton material into a mold open at one end which, in turn, is placed in an impervious alumina housing likewise open at one end. A body of the aluminum infiltrant metal is positioned inside the alumina housing in line with the opening in the mold. The alumina housing is evacuated at a temperature of at least 500° C. to facilitate outgassing of the aluminum and the contents of the mold. The temperature is then increased to at least 950° C. and gas pressure is applied to the molten aluminum metal to introduce such under pressure into the evacuated mold opening to infiltrate the fibrous skeleton therein. Finally, the material within the mold is solidified progressively moving in a direction opposite that in which the molten metal moved to infiltrate the fibrous skeleton.

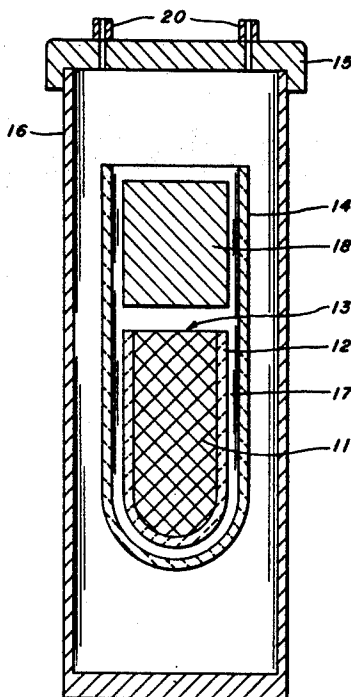


FIG. 1.

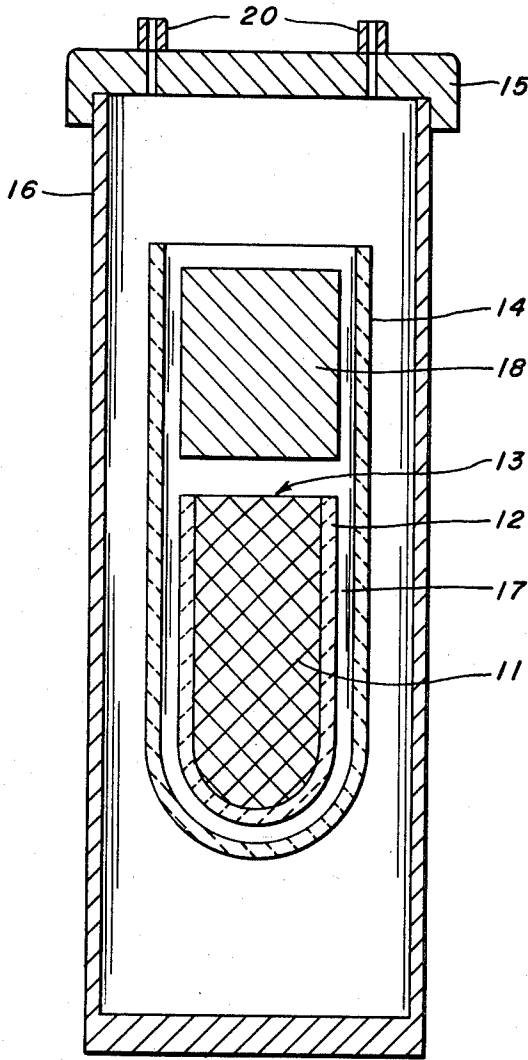
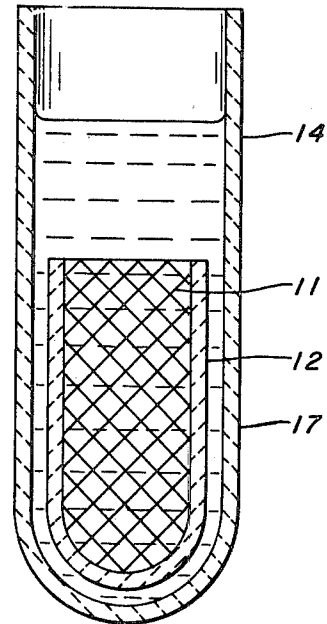


FIG. 2.



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PRODUCTION OF REINFORCED COMPOSITES

BACKGROUND OF THE INVENTION

It has been proposed to provide composites featuring a matrix of one material reinforced by some other material of much higher strength. An example of such is an aluminum metal matrix reinforced by whiskers of alumina or other suitable reinforcing additions. The whiskers here may be almost perfect crystals having one dimension very much greater than the other dimensions. Such whiskers, as is known, are characterized by rather high tensile strength levels, as high as 1,000,000 p.s.i., or even higher. Other fibrous reinforcing materials are wool, yarn and the like known in the art. Important considerations in composites of the type described are that utilizing the enormous strength capabilities of the reinforcing whiskers requires that the load imposed on the composite be transferred to the whiskers and that the load be exerted along the length of the whiskers so that the load can be transferred from whisker to whisker through the matrix where the ends of the whiskers overlap. The latter requires that for maximum reinforcement the whiskers be properly oriented so that their lengths coincide with the direction in which the load will be applied, which itself, raises somewhat of a problem in that the whiskers are of minute size and orienting them in one direction calls for an exercise of considerable care. The whiskers, whether unidirectionally oriented or otherwise, must have the structural loads imposed upon the composite transferred to them. This requires a very high level of adhesion between the matrix and the reinforcing whiskers.

One desirable matrix, because of its high strength to weight ratio, is aluminum along with its alloys. It is particularly desirable to provide composites of aluminum reinforced with alumina whiskers or sapphire whiskers as they are often termed. Sapphire wool, consisting of a mat of sapphire whiskers, is often employed for this purpose. The reinforced aluminum composite, where imparted with a sufficient volume fraction of sapphire whiskers, can exhibit tensile strengths up to 100,000 p.s.i. and is extremely useful in many applications where a high strength to weight ratio, especially at elevated temperatures, is desired.

One problem inherent in producing aluminum-sapphire wool composites is achieving an adequate amount of matrix-whisker adhesion. According to prior practice, achieving good adhesion between the aluminum matrix and the sapphire whiskers generally requires that the latter first be coated with nickel, titanium, or the like, prior to the addition of the aluminum in order to promote an efficient bond. This coating operation adds considerably to the cost and effort involved in producing aluminum composites reinforced with sapphire whiskers. Another serious problem is porosity in the aluminum which inherently impairs the strength of the composite. Achieving both good adhesion and substantial freedom from porosity without recourse to expensive and cumbersome measures such as precoating forms an object of this invention.

STATEMENT OF THE INVENTION

The present invention contemplates infiltration by molten aluminum metal of a fibrous skeleton which has previously been placed in a mold open at one end. The fibrous material may be sapphire whiskers or wool. The open ended mold is placed in an impervious alumina housing which may be of cylindrical configuration. A body of the aluminum infiltrant metal is positioned inside the alumina housing with access to the mold entrance. The alumina housing, the aluminum, the contents of the mold and the mold itself are desirably evacuated, preferably at a temperature of at least 500° C., to remove gas that can cause porosity in the composite. The temperature is increased to at least 950° C. and then pressure is applied to the molten aluminum to force such into the mold opening so as to infiltrate the fibrous skeleton contained therein. The mold contents are then solidified, preferably progressively

solidified, moving in a direction opposite that in which the molten metal moved to infiltrate the fibrous skeleton. One satisfactory arrangement is to position the aluminum body just above the mold entrance so that the aluminum can flow downwardly into the mold, urged by the pressure application above the molten aluminum. The resulting composite exhibits very good adhesion between the sapphire wool reinforcement and the aluminum matrix and also exhibits substantial freedom from porosity. These characteristics tend to maximize the strength achieved with a given volume fraction of reinforcement.

DETAILED DESCRIPTION

In the ensuing detailed description, reference is made to the drawings in which:

FIG. 1 is an elevation view in cross section showing an arrangement suitable for carrying out the invention and

FIG. 2 is likewise a partial elevation in cross section showing the aluminum matrix in the molten condition.

Referring to FIG. 1 mold 12 is positioned inside alumina housing 14 all of which are situated inside a pressure vessel 16. Above the mold and inside the alumina housing is placed a body of aluminum infiltrant metal 18.

The aluminum infiltrant metal may be substantially pure aluminum or an alloy of aluminum such as an alloy containing a small amount of magnesium or manganese which alloys are known to be strengthened by the solid solution effects of the alloying additions. Also, aluminum alloys of the so-called heat treatable or the artificially ageable alloys may be employed. Because of the added strength inherent in these alloys, the matrix can contribute significantly to the total strength of the composite.

The reinforcement may be provided by alumina, or sapphire, whiskers as they are often designated. Sapphire whiskers are essentially elongated single crystals of extremely small cross section normal to their long axis. A typical dimension transverse to the whisker length is 2 microns. A typical whisker length is several hundred microns. Also useful is a sapphire wool material which is a mat comprising a large number of whiskers. The mat may be compressed so that substantially all the whiskers have their length oriented in a single flat plane. They may also be compressed into other basic shapes which may orient many of the whisker axes along some desired direction. Also, a multistrand yarn fashioned from whiskers might be employed. The whiskers may also be suspended in plastic materials which can be extended to orient the whiskers. The plastic is burned away in air to leave a yarn of the oriented whiskers for incorporation into the matrix.

While the invention is particularly suitable in connection with sapphire whiskers other materials may also serve as the reinforcement phase. For instance, boron nitride whiskers are known to exhibit very good strength and have served as reinforcement in a number of composites and would be suitable in practicing the invention.

In the practice of the invention, the whiskers or the whisker mat is placed inside a mold. It is preferably packed by the application of some sort of force in order to increase the volume fraction of the reinforcing media since, generally speaking, the greater the volume fraction, the greater becomes the strength in the composite. Thus, the mold is substantially filled to increase the volume fraction in the preferred practice of the invention. A volume fraction of 0.3 provides for very good strength in a composite featuring an aluminum matrix although very significant strength improvements, two to threefold or more, are realized with a lower volume fraction, for instance a volume fraction of 0.1. The whiskers may be packed in such a way as to arrange their respective lengths along so some preferred direction although this can be rather troublesome. The known advantage here is concentrating all of the reinforcement in one direction and making possible more efficient packing to achieve higher volume fraction.

However, these aspects are known by those practicing the art and are not further elaborated upon. Regardless of how the whiskers are packed into the mold, they form a network which is referred to herein as a fibrous skeleton, that is, a skeleton of fibrous reinforcing material.

The mold is preferably fashioned from a material which is not easily wetted by molten aluminum. This facilitates the easiest possible mold release of the solidified composite. For instance, referring to FIG. 1, the mold 12 may be of a split design and fashioned from a compacted powder of boron nitride which also contains some boron oxide powder. After solidification of the composite, the mold can be separated therefrom by parting the sections. If the aluminum adheres to the boron nitride mold, as sometimes may occur, the mold sections can still be parted without severe difficulty since, in the areas of mold adhesion, a thin mold residue of boron nitride or oxide particles releases with the casting. This residue is readily removed from the composite casting by filing or scraping. The mold is provided with an entrance opening 13 to permit molten metal to enter and infiltrate the mold contents.

As shown in the drawing, mold 12 is situated inside an impervious alumina housing 14 which extends beyond the mold entrance 13. The alumina housing may be provided in the form of a tube closed at one end as shown in the drawing. A slug or body 18 of the aluminum infiltrant metal is positioned in the alumina housing 14 but outside of the mold 12 although it is positioned in access relationship with the mold entrance 13. By access relationship is meant that it is positioned such that when it melts, it can be caused to flow into the mold opening. One way of facilitating such is to position the body above the upwardly facing mold opening as shown in the FIG. The aluminum body, where the alumina housing has a circular opening, may conveniently be a cylindrical slug. Of course, it should be of ample size to fill the mold cavity and the space 17 between the mold and the alumina housing.

The impervious alumina housing 14 is placed inside a vessel 16 which can withstand several hundred pounds of pressure at temperatures up to 1,000° C. and even much higher. One such suitable vessel is a stainless steel tube 16 closed at its bottom end. The stainless steel tube is shown provided with one or more nozzles or connections 20, suitably positioned in cap 15 for the removal or addition of gas. Heating means, not shown, are provided in any way appropriate for heating the contents of the vessel tube 61. The heating means, suitably induction heating means, are desirably movable along the outside of vessel 16 to facilitate directional cooling which, in such an arrangement, is effected merely by progressive withdrawal or relative movement of the heating means with respect to the mold.

In operation, the system is heated to at least 950° C. although it is preferable that the outer vessel or tube 16 first be evacuated as the temperature is increased to 550° C. and higher. The vacuum level desired here is about 1 Torr absolute pressure. Lower absolute pressures, 1×10^{-5} Torr are preferred. The vacuum causes outgassing of the contents of the tube 16 and particularly as to the alumina housing 14 and its contents. Outgassing usually entails about 1 hour at 550° C. Since the process, from the standpoint of the aluminum matrix and the fibrous skeleton, is all contained within the alumina housing, tube 16 could theoretically be omitted although alumina housing 14 would then have to serve as a pressure vessel. Obviously, this places very severe conditions on the alumina housing which are alleviated by the use of tube 16. However, in this description, and in the claims appended hereto, the alumina housing is described as being evacuated since the purpose of evacuating is to evacuate and outgas the alumina housing and its contents. As indicated earlier, it is preferred to outgas the system to minimize porosity in the composite casting.

After outgassing has been effected, the alumina housing and its contents are heated to a temperature of at least 950° C., and preferably of at least 1,000° C. If outgassing is not employed, the system is simply brought to the 950° C. minimum

temperature without any evacuation. This heating causes the molten aluminum to wet the alumina housing and flow into the space 17 between the mold and the housing as shown in FIG. 2. Since the aluminum wets the alumina housing, it provides a seal against and across the housing which seals off the contents of the alumina housing below the aluminum pool from any gas which may be applied above the pool. This seal facilitates forcing the molten aluminum into the mold by the application of pressure which desirably is gas pressure. When gas pressure is applied above the molten aluminum pool, it forces the molten aluminum into the mold and into and throughout the interstices of the alumina whiskers or other fibrous reinforcing agents constituting the fibrous skeleton within the mold. 150 desirable gas pressure is at least 150 p.s.i. although pressures of 300 p.s.i. and higher are preferred.

A preferred practice of the invention, especially suited where the reinforcing media is degraded by molten aluminum at 950° C. or higher, contemplates, after heating to at least 950° C. and effecting wetting of the alumina by the molten aluminum, but before applying gas pressure, the additional step of cooling to a lower temperature of 10° to 100° C. higher than the liquidus temperature of the matrix. Lowering the temperature at this point retains the advantage of the higher temperature exposure in that the alumina housing is still wetted by the molten aluminum while keeping to a minimum any degradation of the reinforcing media. Another advantage is further minimizing any sticking which may occur between the matrix and mold materials.

The gas employed to apply pressure should be one which does not react excessively with the molten aluminum so as to excessively contaminate such. A highly suitable gas in this connection is argon and the other gases from Group VIII of the Periodic Table. If the exposure to molten aluminum is not excessively long, for example, less than 1 hour, nitrogen may be employed. The application of the gas pressure is maintained for a sufficient time before freezing to allow thorough infiltration of the fibrous skeleton in the mold with the molten aluminum. A typical range for time is 1 to 60 minutes depending on the fineness of the individual filaments or whiskers. A time of 10 minutes at pressures of 200 p.s.i. or greater is preferably allowed for infiltration of 0.1 volume fraction of 2 micron diameter whiskers in a two-dimensional random array although the infiltration is reasonably complete in less than 1 minute.

After infiltration is completed, the mold contents are solidified preferably by directional freezing to minimize porosity. The freezing should proceed in a direction opposite that of the molten matrix metal infiltration. That is, referring to FIG. 1 where the molten matrix metal infiltrated the mold contents by moving downwardly into the mold, the directional freezing is accomplished in an upward direction, i.e. with the bottom of the mold being cooled first. Where the heat is provided by a movable induction coil outside tube 16, directional solidification can be provided by simply moving the coil upwardly in a gradual manner. During solidification, it is desirable that the gas pressure be maintained to assist feeding of molten metal to compensate for shrinkage, to minimize porosity and to maintain wetting of the reinforcing agent with molten metal up to the moment of freezing. After solidification, the mold contents are removed and subjected to any desired further processing such as machining.

A better understanding of the invention evolves from the following illustrative example. A tensile strength of 17,500 p.s.i. was obtained for a composite sample containing 0.112 volume fraction of sapphire paper, 1—3 micron diameter single crystal alumina whiskers, 500—5,000 microns long, aligned randomly in a single plane, incorporated into high purity aluminum with 1.4 percent porosity. The tensile strength of a comparable blank of high purity aluminum was only 6,600 p.s.i. Using the appropriate formula for random whisker alignment is a single plane, the tensile strength of the sapphire whiskers in this matrix actually utilized is calculated as being:

$$\frac{3[17,500 - (1 - 0.112 - 0.014)6,600]}{0.112} = 315,000 \text{ p.s.i.}$$

While this strength level is somewhat less than the often discussed exotic strength levels of 1,000,000 p.s.i. or thereabouts, it must be realized that these exotic strength levels are of a more theoretical than practical nature and are seldom realized in actual composites. The above-calculated utilized whisker strength does compare very favorably with the calculated utilized whisker strength reported by others who generally found it necessary to first coat the whiskers with another metal before the incorporation into an aluminum matrix. Metallographic examination indicated a very good bond between the matrix and sapphire whiskers with no noticeable porosity and no discernible reaction zone at the bond interface.

We claim:

1. The method of infiltrating, with an infiltrant metal of aluminum or an alloy thereof, a skeleton of a fibrous reinforcing material to provide a reinforced composite comprising the steps:

- a. providing a skeleton of said fibrous material in a mold open at one end, the mold being positioned inside an alumina housing with a body of said infiltrant metal positioned in access relationship with the mold opening;
- b. heating the alumina housing, the infiltrant metal and the mold and its contents to a temperature of at least 950° C. to cause said infiltrant metal to wet said alumina housing;
- c. urging, by the application of pressure, said infiltrant metal into said mold and through the interstices of said fibrous skeleton; and
- d. solidifying said infiltrant metal.

2. The method of infiltrating, with an infiltrant metal of aluminum or an alloy thereof, a skeleton of a fibrous reinforcing material to provide a reinforced composite comprising the steps:

- a. providing a skeleton of said fibrous material in a mold open at one end, the mold being positioned inside an alumina housing with a body of said infiltrant metal positioned in access relationship with the mold opening;
- b. heating and evacuating to effect substantial outgassing of the alumina housing contents together with the aluminum

metal;

- c. further heating to a temperature of at least 950° C. to cause said infiltrant metal to wet said alumina housing;
- d. introducing said aluminum metal under pressure into said mold and through the interstices of said skeleton; and
- e. progressively solidifying the contents of the mold by freezing in a direction opposite that in which the skeleton was infiltrated.

3. The method of infiltrating, with an infiltrant metal of aluminum or an alloy thereof, a skeleton of a fibrous reinforcing material to provide a reinforced composite comprising the steps:

- a. providing a skeleton of said fibrous material in a mold having an entrance opening at its upper end, the mold being positioned inside an alumina housing with a body of said infiltrant metal positioned within said housing and above and adjacent the mold entrance opening;
- b. evacuating at a temperature of at least 500° C. to an absolute pressure of not more than 1 Torr to effect outgassing of the alumina housing contents;
- c. further heating to a temperature of at least 950° C. to cause said infiltrant metal to wet said alumina housing;
- d. urging said aluminum metal downward into said mold and into and throughout the interstices of said skeleton by the application of a gas pressure of at least 150 p.s.i. above said aluminum metal, the gas being selected from the group consisting of N and the inert gases in Group O of the Periodic Table of the elements; and
- e. progressively solidifying the contents of said mold by freezing progressively upwardly from the mold bottom to its top.

4. The method according to claim 3 wherein there is disposed between steps (c) and (d) the added step wherein the temperature is lowered to a level of from 10° to 100° C. above the liquidus temperature of the infiltrant metal.

5. The method according to claim 1 wherein the interior surface of the mold comprises compressed boron nitride particles.

6. The method according to claim 1 wherein the skeleton of fibrous reinforcing material comprises sapphire whiskers.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,547,180 Dated December 15, 1970

Inventor(s) Charles N. Cochran and Richard C Ray

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Col. 1, line 20 delete "erted along the length of the whiskers so that the load be ex-".

Col. 2, line 4 delete "above the mold entrance so that the aluminum body just".

Col. 2, line 72 delete "so" after "along".

Col. 3, line 25 insert --14-- after "housing".

Col. 3, line 46 change "61" to --16--.

Col. 3, line 66 change "16" to --18--.

Col. 4, line 14 change "150" to --A--.

Col. 4, line 73 change "is" to --in--.

Col. 5, line 6 delete "utilized".

Col. 5, line 28 delete "and" after ";".

Col. 6, line 5 delete "and" after ";".

Col. 6, line 28 delete "and" after ";".

Signed and sealed this 23rd day of March 1971.

(SEAL)
Attest:

EDWARD M. FLETCHER, JR.
Attesting Officer

WILLIAM E. SCHUYLER,
Commissioner of Paten