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[54] PROCESS FOR PRODUCING WHISKER-REINFORCED METAL MATRIX COMPOSITES BY LIQUID-PHASE CONSOLIDATION

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[52] **U.S. Cl.**..................................**29/419,** 29/420, 29/420.5, 29/527.5, 29/DIG. 47, 164/76, 164/120, 164/319

[51] **Int. Cl.**..**B23p 17/00,** B22f 3/24

[58] **Field of Search**.................29/419 R, 419 G, 420, 527.5, 29/527.7, DIG. 47; 164/120, 319, 76, 321

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[57] **ABSTRACT**

A fiber-reinforced metal composite of desired shape is produced by consolidating a mixture of the metal matrix and the fibers under pressure with the mixture maintained at a temperature in which the matrix system is partly in the liquid phase and partly in the solid phase, utilizing a liquid-phase extrusion die cavity of predetermined volume. With approximately one quarter of the matrix system in the liquid phase, and containing up to approximately 50 percent by volume of oriented fibers or whiskers, the material of the composite billet is extruded into the die cavity in order to consolidate the composite billet to the volume of the die cavity, in which the desired shape is to be formed. Heating is discontinued when the cavity is filled completely by the semi-molten metal fiber composite.

10 Claims, 2 Drawing Figures

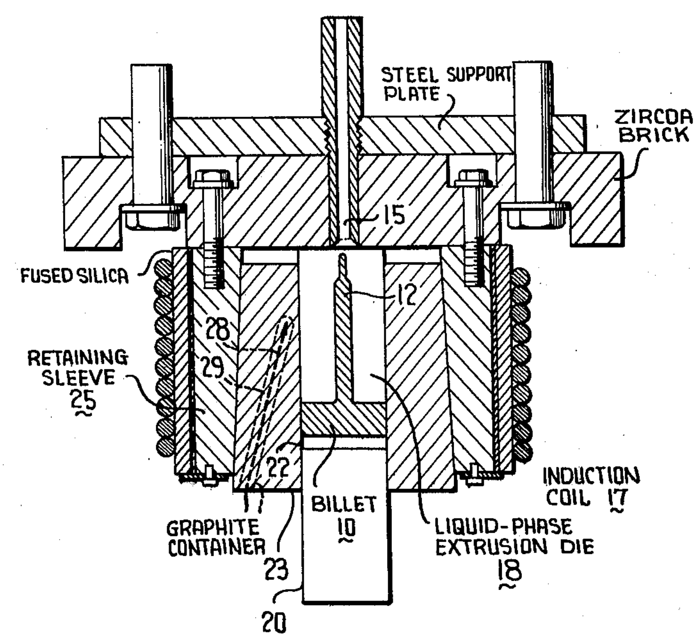

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FIG. 2

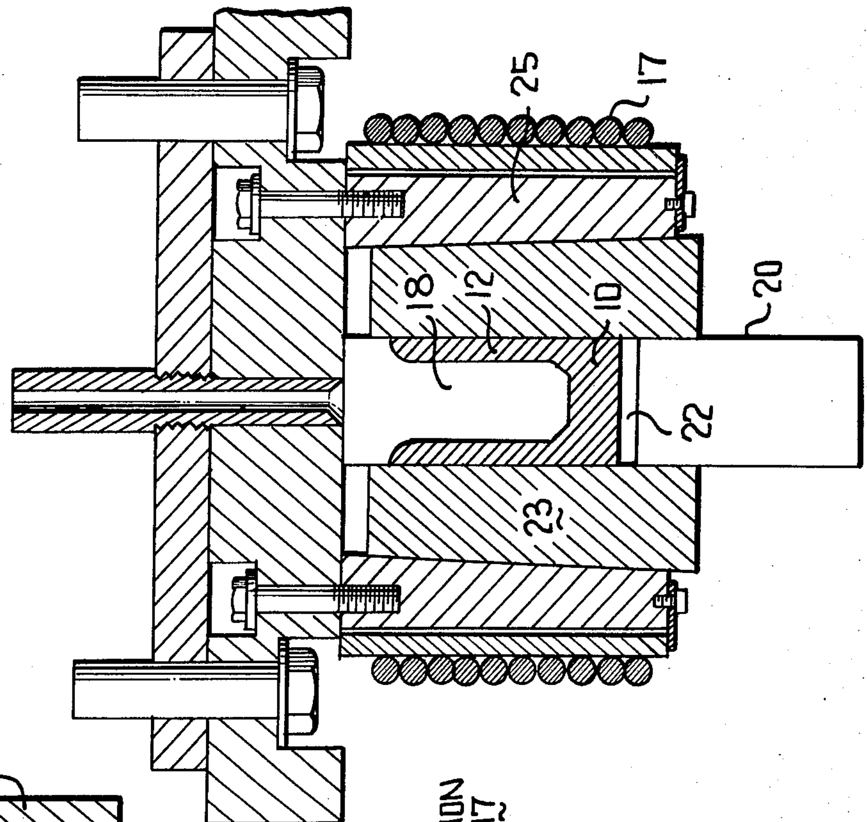
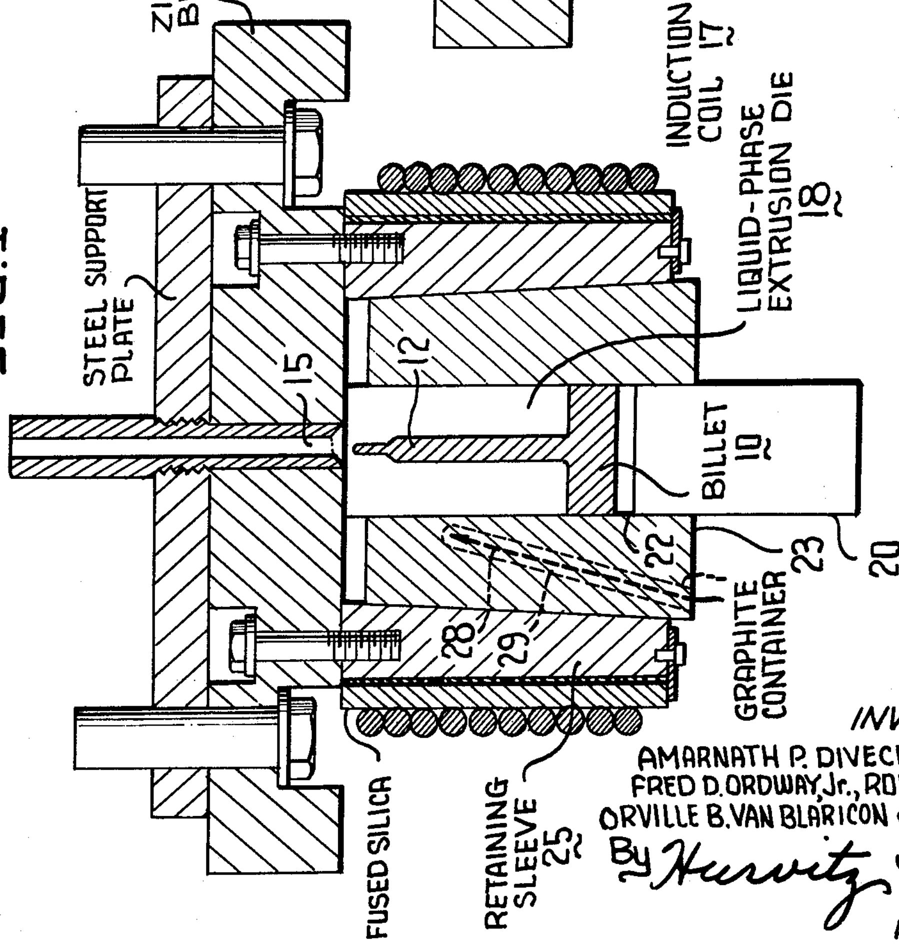


FIG. 1



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PROCESS FOR PRODUCING WHISKER-REINFORCED METAL MATRIX COMPOSITES BY LIQUID-PHASE CONSOLIDATION

BACKGROUND OF THE INVENTION

The present invention relates generally to the reinforcement of metal matrixes with high-strength ceramic fibers (whiskers), coated with suitable metals, or uncoated, and more particularly to apparatus and processes for incorporation of the fibers in a matrix using a method of consolidation and extrusion inherently producing a preferred fiber orientation. The term fiber includes but is not limited to SiC, B, Al_2O_3.

In the application of Divecha et al., Ser. No. 626,190, filed Mar. 27, 1967, entitled "Preparation of Fiber-Reinforced Metal Composites by Compaction in the Liquid-Phase," of common assignee herewith, now U.S. Pat. No. 3,441,392, issued Apr. 29, 1969, several prior art methods of producing fiber-reinforced composites are discussed, and it is observed that one of the principal problems encountered in obtaining fiber-reinforced metal composites has been the lack of suitable processes capable of producing specimens in sizes sufficiently large for practical use and for meaningful evaluation of working characteristics.

One basic requirement in the production of the successfully reinforced composite is the capability of the composite to transfer load from one whisker or fiber to another. Another requirement is that there be a strong bond at the whisker-matrix interface. Wetting of the fibers by the matrix is also essential, and it has been found, for example that pure molten nickel bonds to alumina fibers under prolonged contact of about 30 minutes or more, and that addition of slight amounts of chromium (approximately 1 percent) also enhances wetting of the fibers.

In typical prior art processes the fibers undergo random orientation during formation of the composite, and consequently the compacted and extruded material is of lower strength than could be achieved by selective orientation of the fibers. Moreover, considerable fiber fracture occurs during the consolidation process. In the process disclosed in the aforementioned Divecha et al patent, compaction is performed at temperatures at which the metal matrix is maintained in its semi-molten region. More specifically, the matrix material is mixed in powder form with the coated or uncoated fibers, the latter in loose separated form, and the resulting mixture placed in the die of a conventional hot-pressing apparatus where it is heated to a temperature slightly below the solidus of the system. At that point, a predetermined constant pressure is applied to the metal matrix-fiber system and the temperature of the composite is then raised until the state of the system experiences a cross-over into the semi-molten region of the matrix. Pressure is maintained until desired compaction is achieved, the matrix completely encapsulating the fibers after which the composite is cooled and removed and hot rolled to produce a preferred fiber orientation. Prior to the hot rolling operation, the process produces dense bodies consisting of randomly oriented whiskers in the metal matrix. In order to retain liquid metal within the die cavity while the composite is under pressure at consolidation temperatures, extremely close tolerances are maintained between the solid punch and die cavity of the press mechanism.

While the process as disclosed in detail in the aforementioned Divecha et al. patent is highly advantageous over processes disclosed in the art prior thereto, it is not without its limitations. In particular, while the composites produced are substantially larger than those produced by earlier-employed techniques, they are nevertheless still of relatively small size, necessarily limited by existing mechanical apparatus. For example, such structural shapes as rods, I-beams, tubing, and channels are not readily fabricated by this process because of the usual size requirements of those articles. In addition, a hot rolling step is ultimately necessary to ensure whisker orientation in the preferred direction. The process is best employed to produce solid cylindrical shapes as an initial product prior to further machining or other processing. Other shapes may be produced, but are less desirable economically because of the special die designs involved, as well as material costs and availability.

In the co-pending application of Lare et al., Ser. No. 799,329, filed Feb. 14, 1969 now abandoned, entitled "Process for Consolidation and Extrusion of Fiber-Reinforced Composites," and commonly assigned with respect to this application, there is disclosed a process for producing fiber-reinforced composites in a single continuous operation in which extrusion of the composite into the desired shape immediately follows consolidation of the fiber-matrix system into the reinforced composite, without the requirement of transfer of the composite to another press. In that invention the press includes a die cavity having one or more orifices, each of which is normally closed by a seal of sufficient rigidity to withstand consolidation pressure exerted on the fiber-matrix system, but which seal is itself adapted to undergo extrusion to permit the immediately priorthereto consolidated body to be extruded via the respective die orifice (also termed the extrusion port). After the "green" i.e., unconsolidated fiber/powder mixture is placed into the die, the entire assembly is heated slowly from room temperature to a temperature slightly above the solidus temperature of the mixture, that is, to a temperature at which the matrix is in the liquid phase or in a semi-molten state. During this period, the system is maintained under relatively low pressure in the range from approximately 2,000 to approximately 5,000 pounds per square inch (psi), in a manner similar to that disclosed in the aforementioned Divecha et al patent, until the desired consolidation is achieved. However, in accordance with the Lare et al. invention, upon conclusion of the consolidation cycle the temperature is reduced to a value between the solidus and matrix melting point (a state which may be characterized as a "mushy" phase), and the pressure on the consolidated system or composite is increased to a value sufficient to rupture the seal and to extrude the composite via the extrusion port. This range of temperature values, at which pressure in excess of the consolidation pressure is applied to the composite for extrusion thereof, is critical.

In particular, in the latter invention extrusion is performed at a temperature at which the shear strength and tensile strength of a matrix are sufficiently low to permit ready flow of the matrix material, such that the fibers are merely carried with the matrix in the direction of matrix deformation. While the precise temperature is dependent upon the character and concentration of fibers or whiskers, it resides in a relatively narrow range quite close to the melting point of the matrix.

It has been found that this temperature range near the melting point of the matrix produces some surface tearing of the composite, and additionally that fiber fracture occurs in significant proportions.

It is a principal object of the present invention to provide a process for producing fiber-reinforced metal matrix composites which overcomes one or more of the disadvantages of the prior art processes.

SUMMARY OF THE INVENTION

Briefly, according to the present invention, a process for producing fiber-reinforced metal matrix composites is performed by consolidation entirely in the semi-molten putty-like state. In brief, a composite billet is prepared which incorporates the desired fibers, such as alumina and/or silicon carbide, in a metal matrix, the billet containing up to about 20 percent by volume of oriented whiskers (fibers). The whisker orientation may be controlled by a magnetic orientation system in which the fibers themselves follow the magnetic lines of force, provided they have been coated with magnetizable metal. The entire assembly is heated to approximately 585° C, in the case of a matrix alloy composed of 2.5 percent silicon and the balance aluminum, this temperature being above the solidus temperature to maintain from about 20 to about 25 percent of the metal matrix in the liquid phase. At this point, pressure is applied to the billet for compaction thereof into a closed die cavity, the billet volume being slightly in excess of that formed by the cavity. A ram associated with the press in which the billet is contained is forced against the billet to produce a pressure of from 2,000 to 5,000 psi thereon in order to produce flow of the putty-like material. The ram travel is accurately monitored, and the heating is ceased when the billet has been entirely transferred to the cavity. The extent of travel of the ram is predetermined to insure that the cavity is entirely filled by the semi-molten metal composite. Since the volume of the composite is substantially identically equal to the volume of the closed die cavity, complete densification is achieved.

By use of the process of the present invention many complex shapes, such as nuts, bolts, engine blocks, turbine blades, and so forth, may be formed with reliable composite strength as a consequence of the high aspect ratio and controlled orientation of the fibers and the complete compaction of the final composite member. Only the closed die configuration need be altered according to shape of the composite to be formed.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a simplified schematic diagram of a press employing a closed die cavity for preparation of a rod shaped composite; and

FIG. 2 is a schematic diagram of a press, similar to that shown in FIG. 1, except that the closed die cavity of this embodiment is used for preparation of a tubular or cup-shaped composite.

DESCRIPTION OF THE PREFERRED PROCESS

Suitable fibers (whiskers) for use in reinforcing the metal matrix include alpha alumina (sapphire) and silicon carbide, Carbon, Boron or the like of fiber diameters ranging upwardly to approximately 30 microns and of lengths up to about one-half inch. The fibers may be either coated with suitable metal or uncoated, or both. An example of a suitable coated fiber is sapphire which has been plated with a thin coating of nickel. Such fibers are available from several sources, one of which is General Technologies Corporation, of Reston, Virginia.

In preparation of each fiber-reinforced metal composite by compaction or consolidation in the liquid phase of the matrix, the use of an alloy as the matrix is preferred over the use of a pure metal, since liquid phase hot pressing requires a system having a distinct two-phase region (i.e., liquid plus solid). Either mixture or prealloyed powders of the matrix components may be utilized, but the prealloyed powder has the advantage of a known melting point, whereas the melting point of the mixture must usually be ascertained by experimentation. Moreover with the prealloyed powder there is less probability of producing a non-homogenous structure as a result of the formation of an undesirable intermetallic phase during heating and compaction.

Some of the many suitable mixed and prealloyed powders for preparation of the fiber-reinforced composite include prealloyed atomized powder composed of 10.2 percent by weight of silicon, 0.03 percent magnesium, 0.67 percent iron, and the balance aluminum, of sufficient fineness to pass a standard No. 325 screen, Al–2.5 Si minus 400 mesh prealloyed atomized powder; Al-Cu minus 325 mesh prealloyed atomized powder composed of 4.5 Cu, balance Al; 7075 Al minus 325 mesh prealloyed atomized powder composed of 0.3 Si, 0.6 Fe, 2.1 Cu, 0.2 Mn, 2.2 Mg, 0.2 Cr, 0.10 Ni, 4.7 Zn, balance Al, all of the foregoing powders available from Reynolds Aluminum Company, Richmond, Virginia; Nichrome, 80 Ni-20Cr minus 325 mesh prealloyed atomized powder; Nickel — 2 percent by weight silicon mixed powders; 99.7 percent pure nickel powder, 4–7 microns, prepared by carbonyl process, available from International Nickel Company, of Reston, Virginia; 98 percent pure silicon powder, minus 325 mesh, available from Consolidated Astronautics, New York; and nickel minus 325 mesh prealloyed atomized powder, composed of 1 Si, 1 Mn, balance Ni, available from Hoeganaes Sponge Iron Company of New Jersey.

The following is an example of a process according to the invention, in which an extremely dense silicon carbide fiber-reinforced aluminum-silicon composite is prepared in a desired shape.

After the silicon carbide fibers have been dispersed or separated in known manner, as by agitation is isopropanol, a prealloyed atomized powder or mixed powder, composed of 2.5 weight percent silicon and the balance aluminum, passing a standard No. 400 screen, is added to the resulting slurry in an amount to produce a metal matrix containing up to 50 percent by volume of silicon carbide fibers. In this specific example the volumetric percentage of SiC fibers is 15 percent. Preferably the fibers have diameters in the range from one to three microns and lengths up to about one-half inch. The constituents of the fiber-metal mixture are thoroughly mixed by stirring or agitation for a period of several minutes, during which the mixture may be filtered by aspiration through filter paper.

In order to achieve superior distribution of the composite fibers and to provide higher strength of the ultimate composite material, particularly at the higher fiber concentrations (up to about 40 percent by volume), it is advantageous to use an oriented-fiber matrix mixture. This may be achieved by depositing a thin film of magnetic material such as nickel on the fibers, by an appropriate method such as thermal decomposition of a volatile compound such as nickel carbonyl, $Ni(CO)_4$. The metallized whiskers are then dispersed with the matrix powder to produce a slurry in isopropanol as described earlier. By filtration of the slurry through filter paper in a magnetic field the metallized fibers are deposited on the paper in parallel alignment.

Thereafter, the mixture is dried for compaction or consolidation in a press, such as the hot pressing apparatus shown in FIGS. 1 and 2. The matrix-fiber mixture (billet) **10** is placed adjacent to a die cavity **12** which is closed by a blocking plate aligned with the orifices of the cavity, the latter forming the shape of the final extruded composite. The press is heated by induction coils **17**, the low melting alloy of aluminum — 2.5 percent silicon of the present example being processed using a tool steel die **18**, although a graphite die suitable for higher melting alloys may be employed, if desired. It is desirable, in any event, to employ a plunger or ram **20** and block **22** of graphite to prevent seizing, despite the use of low melting alloys. For a tool steel die, the walls of the die cavity may be coated with suitable lubricant such as molybdenum disulfide to permit rapid ejection and complete extrusion of the composite following consolidation.

The die may be encompassed by a graphite layer **23**, and encased in a fused silica tube **25** as is typical of conventional hydraulic hot pressing apparatus. Induction heating coil **17** is wound about the silica tube **25**, and a temperature sensing element such as thermocouple **28** may extend into a recess **29** within graphite layer **23** to allow monitoring of the temperature of the press at a point in close proximity to the fiber-matrix mixture.

After insertion of the fiber-alloy mixture **10** into the die cavity, the die is heated by the induction unit to approximately 585° C which is above the solidus temperature of the mixture, thereby melting about 20 to about 25 percent of the metal in the case of the aluminum—2.5 percent silicon alloy. When the matrix is heated to a temperature in excess of the solidus temperature, pressure of 2,000 psi to 5,000 psi is applied by the ram to force the billet **10** into the closed die cavity **12**. The billet volume is slightly in excess of that of the cavity. The movement or travel of the hydraulic ram **20** is monitored. When a predetermined position is reached, at which the cavity is entirely filled by the semi-molten metal composite, the heating is discontinued. It should be noted that the billet will not flow under ordinary conditions because the low percentage of liquid matrix and the presence of the fibers therein result in a putty-like consistency.

Only the shape of the die cavity need be changed to conform to the desired shape of the extruded composite. Movement of the punch or ram serves to form the billet into the shape of the die cavity. The composite billet excess is minimal or negligible because the material is almost entirely forced into the container cavity during entry of the ram therein. The effect is formation of the desired shape under total compressive or triaxial force to achieve substantially complete densification, and a porosity much lower than has heretofore been achieved. In addition, there is virtually no waste of composite material under compaction, because it is assured in advance that the billet is of sufficient volume (substantially equal to the volume of the cavity) to be consolidated entirely within the die cavity.

The die assemblies shown in FIGS. **1** and **2** are substantially identical except for the shape of the die (and hence, of the die cavity). In FIG. **1**, die **18** is provided with an elongated axial hole constituting the cavity **12**, to permit formation of rod-shaped composites. In FIG. **2**, die **18**, in conjunction with graphite container **23**, forms a tubular cavity **12** to permit formation of cup-shaped composites.

It is to be emphasized that according to the present invention a fiber-reinforced metal composite is consolidated by the monitored movement of a punch or ram of a press to force the semi-molten putty-like composite billet entirely into a closed die cavity of desired shape. Since the volume of the billet is substantially equal to or slightly in excess of the volume of the cavity, it is assured that essentially the entire billet is transferred to the cavity, and consolidation under total compressive or triaxial forces assures that nearly complete densification is achieved (i.e., voids are virtually absent from the consolidated composite).

A desirable fiber orientation exists in the resulting consolidated composite by virtue of the flow taking place during the process, which tends to align the fibers along the direction of shear. Thus a comparatively complex shape may be readily obtained with reinforcing fibers oriented differently at different points, the various orientations being such as to provide optimum strength of the piece at each point.

We claim:

1. A process for producing a fiber-reinforced composite having oriented fibers by means of a single hot forming operation, said process comprising placing a billet of a matrix of metal and fibers adjacent a liquid-phase extrusion die cavity in a position intermediate said die cavity and a plunger, solely induction heating said billet and die cavity to a temperature only sufficiently in excess of the solidus temperature of said matrix to achieve melting of about 20–25 percent of the metal of said matrix, the volume of material in said billet being only slightly in excess of the internal volume of said die cavity, moving said plunger in a direction and for a predetermined distance selected to extrude material of said billet into said die cavity with only slight overage of matrix material such that shear flow of the matrix material occurs and the fibers become oriented along the direction of shear, and immediately thereafter terminating induction heating of said billet and die cavity.

2. The process according to claim **1**, wherein said billet contains up to **50** percent by volume of ceramic fibers.

3. The process according to claim **2**, wherein said fibers are substantially commonly oriented in said semi-molten billet.

4. The process of producing a fiber-reinforced metal composite having oriented fibers by means of a single hot forming operation, said process including heating a fiber-alloy billet composed of up to 50 percent of fibers in a metal alloy powder to a temperature sufficient to convert not more than 50 percent of the alloy to the liquid phase but less than the melting point of said fibers, while the billet is positioned adjacent to a liquid-phase extrusion die cavity of volume substantially equal to the volume of said billet, and applying pressure to the semi-molten billet thus produced to extrude said billet substantially entirely into said cavity such that shear flow of the matrix material occurs and the fibers become oriented along the direction of shear, and thereafter terminating heating of the billet in said cavity.

5. The process according to claim **4** wherein said metal matrix is up to 25 percent liquid in said semi-molten state of said billet.

6. The process according to claim **5** wherein said matrix is aluminum-2.5 percent silicon alloy.

7. The process according to claim **6** wherein said fibers are silicon carbide.

8. The process according to claim **1** wherein said fibers are composed of a material selected from the group consisting of silicon carbide and alpha alumina.

9. The process according to claim **3** wherein said fibers possess a coating of magnetic material and are oriented in a magnetic field during formation of said billet.

10. The combination according to claim **1**, wherein after solidifying of said billet and die cavity following terminating of said heating, removing fiber-reinforced composite from said cavity.

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