



(12) **EUROPEAN PATENT APPLICATION**
published in accordance with Art. 158(3) EPC

(43) Date of publication:
06.02.2002 Bulletin 2002/06

(51) Int Cl.7: **C22C 47/10**

(21) Application number: **00971764.6**

(86) International application number:
PCT/JP00/07745

(22) Date of filing: **02.11.2000**

(87) International publication number:
WO 01/46486 (28.06.2001 Gazette 2001/26)

(84) Designated Contracting States:
**AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU
MC NL PT SE TR**

(30) Priority: **21.12.1999 JP 36246599**

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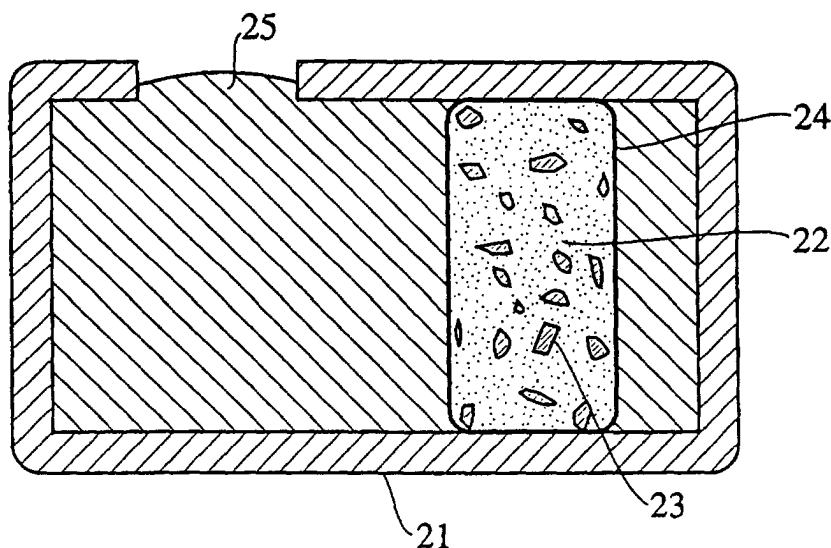
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(54) **METHOD FOR PRODUCING METAL-BASED COMPOSITE MATERIAL**

(57) A method for producing a metal based composite material comprising a matrix metal containing a reinforcing filler in at least a part thereof, characterized as comprising (1) charging at least one reinforcing filler in the form of fibers or particles and at least one penetration-enhancing metal into an aluminum container, (2)

submerging the aluminum container in the state of having an oxygen-containing gas remaining therein into a melt of a matrix metal comprising an aluminum alloy or a magnesium-aluminum alloy, (3) dissolving the aluminum container in the matrix melt, to thereby render the melt of the matrix metal to penetrate into the reinforcing filler, and then (4) solidifying the melt of the matrix metal.

Fig. 2



Description

FIELD OF THE INVENTION

[0001] The present invention relates to a method for producing a metal-based composite material comprising an aluminum alloy, etc., as a matrix, particularly to a method for producing a metal-based composite material by pressureless impregnation.

PRIOR ART

[0002] Metal-based composite materials are composed of a combination of matrix metals having basic properties such as strength, ductility, toughness, etc., and various reinforcing fillers. In many cases, because they contain reinforcing fillers such as long or short fibers, particles, etc. of ceramics, having moldability and thermal conductivity of the matrix metals themselves and rigidity, wear resistance, low thermal expansion coefficient, etc. of the reinforcing fillers, they are widely used in various applications such as parts of transportation vehicles that are becoming lighter in weight, and circuit boards, etc. of electronics parts requiring low thermal expansion coefficients.

[0003] Among the metal-based composite materials, there are dispersion-strengthened composite materials having reinforcing fillers such as ceramics, etc., dispersed in alloy matrices such as aluminum, magnesium, etc. Particularly aluminum alloys are widely used as matrices for the composite materials because of their lightness in weight and low cost.

[0004] Dispersion-strengthened composite materials comprising light alloys such as aluminum alloys, magnesium alloys, etc. as matrices not only are light in weight because of the light alloys, but also have various improved properties such as strength, rigidity, wear resistance, thermal expansion coefficient, density, high temperature strength, etc. because of the inclusion of the reinforcing fillers. How much their properties are improved largely depends on the proportions of alloy components and reinforcing fillers, the shapes and sizes of reinforcing fillers, and treatment methods for producing the dispersion-strengthened composite materials.

[0005] There have conventionally been various methods for producing composite materials comprising light alloys as matrices, and these production methods are mainly classified into a powder metallurgy method, a melt-stirring method and an infiltration method.

[0006] The powder metallurgy method comprises mixing a powdery matrix metal with a reinforcing filler such as long or short fibers, particles, etc., molding the resultant mixture at room temperature, and then sintering or hot pressing the resultant green body to produce a composite material. Because the powder metallurgy method usually comprises press molding, it is not suitable for products having complicated shapes, and thus inevitably directed to products having relatively simple

shapes. Also, because the molding is carried out at high temperature and high pressure, it is disadvantageous in high cost for particularly large products.

[0007] The melt-stirring method comprising adding a reinforcing filler to a molten metal, and stirring the mixture at a high speed for a long period of time to disperse the reinforcing filler in the matrix metal melt, thereby producing a composite material. Though this method can produce large amounts of composite material ingots at a relatively low cost, it is extremely difficult to provide partially composite material products.

[0008] The infiltration method comprises infiltrating a molten metal into space between reinforcing fillers, thereby producing a composite material. This method can produce composite materials having complicated final shapes. However, to infiltrate an aluminum alloy into space between SiC particles, for instance, the SiC particles should have good wettability with the aluminum alloy. "Good wettability" is a characteristic that a liquid has a small contact angle to a solid, meaning that the liquid is easily spread on a solid surface. However, because SiC particles have poor wettability with the aluminum alloy, the infiltration of the aluminum alloy into gaps between SiC particles does not occur spontaneously at atmospheric pressure. Namely, SiC particles are repelled by the aluminum alloy melt, failing to provide a homogeneous composite material.

[0009] The infiltration methods include a pressurized infiltration method in which a matrix metal melt is infiltrated into a porous green body of a reinforcing filler such as ceramics, etc. at high pressure to produce a composite material, and a pressureless infiltration method in which a matrix metal melt is infiltrated into gaps of a reinforcing filler without pressure to produce a composite material.

[0010] In the pressurized infiltration method, because a melt of a light alloy such as aluminum is infiltrated into a green body of a reinforcing filler at high pressure, a partially composite material can be obtained. For instance, Japanese Patent Publication No. 6-38968 proposes a method for producing a metal-reinforcing filler composite material comprising compression-molding a mixture of fine pieces of an oxide of the first metal (for instance, NiO powder) and a reinforcing filler (for instance, silicon carbide whisker) to form a porous body, and causing a melt of the second metal (matrix metal), for instance, an aluminum melt, having a larger tendency to form an oxide than the first metal to infiltrate into the porous body under pressure. However, this method is disadvantageous in that because a uniform mixture of fine pieces of an oxide of the first metal and the reinforcing filler is compression-molded to form a porous body, and causing a matrix metal melt to infiltrate into the porous body by using a high-pressure casting apparatus, it is disadvantageous in high production cost. Thus, this method cannot produce a composite material without pressure, the casting method being limited to a high-pressure casting method or a centrifugal casting

method.

[0011] As an example of a pressureless infiltration method, Japanese Patent 2,801,302 discloses a so-called PRIMAX™ method in which a matrix metal of aluminum, etc. is infiltrated into a reinforcing filler without pressure to produce a metal-reinforcing filler composite material. Specifically, this method comprises placing a mixture or its preform of an infiltration accelerator such as magnesium powder, etc., and reinforcing filler of a silicon carbide, etc. in a non-reactive container; disposing a matrix metal adjacent to the mixture or its preform; placing both in a furnace; filling the furnace with a nitrogen gas atmosphere; keeping the furnace at about 675-1200°C to spontaneously infiltrate a molten matrix metal into the reinforcing filler, thereby producing a metal-reinforcing filler composite material. It is considered that the infiltration accelerator functions to react with a nitrogen gas to improve the wettability of a reinforcing filler surface, thereby accelerating the spontaneous infiltration of the molten matrix metal into the reinforcing filler.

[0012] However, because the composite material is produced in a nitrogen atmosphere in the method of Japanese Patent 2,801,302, a furnace with a gas-substituting means is needed, and the heating temperature should be about 675°C or higher, the production cost of the metal-reinforcing filler composite material is extremely high, making it difficult to put the metal-reinforcing filler composite material into practical use. Further, Japanese Patent 2,801,302 discloses only specific examples of matrix metal-reinforcing filler composite materials in which the reinforcing filler is dispersed substantially entirely, failing to provide any specific examples of partially composite materials,

[0013] Japanese Patent 2,905,519 proposes a method for producing a metal matrix composite body comprising charging SiC powder, etc., as a reinforcing filler and magnesium powder as an infiltration accelerator into a copper mold; sealing one end of the copper mold with a copper foil; substituting the atmosphere inside the copper mold with nitrogen; and immersing the copper mold in a molten matrix metal (for instance, magnesium-containing aluminum alloy melt) at about 750°C for about one hour to produce a metal matrix composite material. During immersion in the molten aluminum alloy, the copper mold and the copper foil are melted, resulting in the spontaneous infiltration of the molten aluminum alloy in the filler.

[0014] In this method, too, like Japanese Patent 2,801,302, the infiltration accelerator appears to improve the wettability of a reinforcing filler surface, accelerating the spontaneous infiltration of the molten aluminum alloy into the reinforcing filler, thereby producing the composite material. However, the copper mold and the copper foil are melted in the matrix metal melt, the composition of the matrix metal melt undesirably varies. Also, an apparatus for substituting the atmosphere inside the copper mold and the copper foil with nitrogen

is needed, and the heating temperature should be at least about 675°C. Accordingly, the resultant metal-reinforcing filler composite materials suffer from high cost, making their practical use difficult. Further, Japanese Patent 2,905,519 discloses only examples for obtaining matrix metal-reinforcing filler composite materials having substantially overall reinforcing fillers dispersed therein, failing to disclose examples of partially composite materials at all.

[0015] U.S. Patent 3,364,976 discloses a method comprising immersing a mold having at least one opening and containing a gas (for instance, a gas containing oxygen and nitrogen such as air) having reactivity with molten magnesium in a melt of magnesium as a matrix metal; causing the magnesium melt to react with oxygen and nitrogen existing in a mold cavity to generate a reduced pressure state inside the mold; and filling the mold with a matrix metal melt by suction effects by the reduced pressure. U.S. Patent 3,364,976 further discloses that this method is applicable to Al-Mg alloys.

[0016] However, in the method of U.S. Patent 3,364,976, the matrix metals infiltratable into reinforcing fillers are substantially limited to magnesium or Al-Mg alloys, with only specific examples using an Al-5%Mg alloy melt to produce composite materials. Further, because the reinforcing filler is charged into a mold made of steel, graphite, glass, etc., and because the mold is immersed in a molten matrix metal, when composite materials partially reinforced with fillers are sought, the mold remains in a boundary between the matrix metal and the metal-reinforcing filler composite material, unsuitable for practical use.

OBJECT OF THE INVENTION

[0017] Accordingly, an object of the present invention is to provide a method for producing a metal-based composite material at a low cost without using a nitrogen atmosphere, by infiltrating a matrix metal melt into space between reinforcing fillers without pressure and at a relatively low temperature.

[0018] Another object of the present invention is to provide a method for producing a metal-based, partially composite material by bringing part of the product composite with reinforcing fillers by pressureless infiltration.

SUMMARY OF THE INVENTION

[0019] As a result of intense research in view of the above objects, the inventors have found that by charging a reinforcing filler in the form of fibers or particles and an infiltration-accelerating metal in the form of powder, chip, foil, plate or bulk into an aluminum container; introducing this container into a matrix metal melt or disposing this container in a die or a casting mold; pouring a matrix metal melt thereinto; and bringing the inside of the container into contact with the matrix metal melt in a state where the inside of the container is shut off from

the outside atmosphere, the infiltration-accelerating metal is caused to react with an oxygen-containing nitrogen gas in the container, thereby spontaneously generating a reduced pressure state in the container, such that the matrix metal melt is sucked into the container to infiltrate the matrix metal melt into space between reinforcing fillers at a relatively low temperature without control of atmosphere and pressure. The present invention has been completed based on this finding.

[0020] Thus, the method for producing a metal-based composite material comprising a matrix metal containing at least partially a reinforcing filler according to the present invention comprises (1) charging at least one reinforcing filler in the form of fibers or particles and at least one infiltration-accelerating metal into an aluminum container; (2) immersing the aluminum container in a melt of a matrix metal made of an aluminum alloy or a magnesium-aluminum alloy in a state that an oxygen-containing nitrogen gas remains in the aluminum container; (3) melting the aluminum container in the matrix metal melt, so that the matrix metal melt is infiltrated into the reinforcing filler, and then (4) solidifying the matrix metal melt.

[0021] In a preferred embodiment of the present invention, the aluminum container is introduced into a matrix metal melt in a crucible. In another preferred embodiment of the present invention, the aluminum container is disposed in a cavity of a die or a mold at a predetermined position in advance, and the matrix metal melt is charged into the cavity.

[0022] In a preferred embodiment of the present invention, an aluminum foil is used as the aluminum container, and a mixture of the reinforcing filler and the infiltration-accelerating metal is completely wrapped by the aluminum foil. In a further preferred embodiment of the present invention, an aluminum can with a lid is used as the aluminum container, and after charging a mixture of the reinforcing filler and the infiltration-accelerating metal into the aluminum can, the aluminum can is closed by the lid.

[0023] By disposing the aluminum container in the matrix metal melt at a predetermined position, it is possible to provide a metal-based composite material whose predetermined portion is only composite with the reinforcing filler. Instead, it is also possible to uniformly disperse the reinforcing filler in the matrix metal melt by stirring the matrix metal melt after the matrix metal melt is infiltrated into the reinforcing filler.

[0024] The oxygen-containing nitrogen gas is preferably air. The space percentage of the container [(volume of oxygen-containing nitrogen gas / inside volume of container) x 100%] is preferably 30-70%.

[0025] The infiltration-accelerating metal is preferably at least one metal selected from the group consisting of magnesium, calcium, zirconium and alloys containing these metals, particularly pure magnesium or magnesium alloy. Also, the infiltration-accelerating metal is preferably in the form of at least one selected from the group

consisting of powder, chip, foil, plate and bulk.

[0026] The matrix metal is preferably an aluminum alloy or a magnesium-aluminum alloy. The temperature of the matrix metal melt is preferably between T_m and $T_m + 40^\circ\text{C}$, wherein T_m is the liquidus point of the matrix metal. The reinforcing filler is preferably constituted by ceramics, particularly SiC particles.

BRIEF DESCRIPTION OF THE DRAWINGS

[0027]

Fig. 1 is a schematic view showing an apparatus used for the method for producing the metal-based composite material in EXAMPLE 1;

Fig. 2 is a schematic view showing an apparatus used for the method for producing the metal-based composite material in EXAMPLE 4;

Fig. 3 is a schematic view showing an apparatus used for the method for producing the metal-based composite material in EXAMPLE 5;

Fig. 4 is a schematic view showing an apparatus used for the method for producing the metal-based composite material in EXAMPLE 6;

Fig. 5 is a schematic view showing an apparatus used for the method for producing the metal-based composite material in EXAMPLE 7;

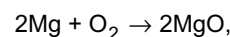
Fig. 6 is a graph showing the variations of pressure and temperature inside the aluminum container in EXAMPLE 1; and

Fig. 7 is a photomicrograph (x 200) showing a cross section structure of a matrix metal-reinforcing filler composite portion of the metal-based composite material produced in EXAMPLE 4.

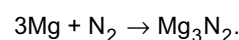
DESCRIPTION OF THE BEST MODE

[1] Principle of the present invention

[0028] With an aluminum container filled with a reinforcing filler and an infiltration-accelerating metal (for instance, magnesium) in a sealed state, keeping this container in a matrix metal melt results in the sublimation of magnesium as an infiltration-accelerating metal by the temperature of the matrix metal melt, causing the following reaction between magnesium and an oxygen-containing nitrogen gas (for instance, air) in the aluminum container, forming oxide and nitride and consuming oxygen and nitrogen (air):



and



Because the aluminum container in which this reacting takes place is immersed in the matrix metal melt, it is not communicating with the outside atmosphere, free from the air entering from outside, spontaneously generating reduced pressure in the aluminum container. When the aluminum container is melted in the matrix metal melt in this state, the matrix metal melt is infiltrated, namely is caused to penetrate, into space or gaps between the reinforcing fillers, even though there is substantially no pressure applied from outside. Further, because the resultant Mg_3N_2 has good wettability with the aluminum melt, the infiltration of the matrix metal melt is accelerated when the matrix metal is an aluminum alloy.

[0029] Oxygen contained in a gas in the aluminum container has higher reactivity with magnesium than nitrogen. Accordingly, the degree of evacuation in the aluminum container is higher when the atmosphere in the aluminum container is an oxygen-containing nitrogen gas than when it is a nitrogen gas atmosphere. Because the present invention uses an oxygen-containing nitrogen gas such as air, the degree of infiltration of the matrix metal melt is higher in the present invention than in Japanese Patent 2,905,519 in which a nitrogen gas is used, with lower production cost of the composite body.

[0030] When the reinforcing filler is in the form of particles, pressure difference ΔP for initiating the infiltration of the matrix metal melt into the reinforcing filler particles is represented by the following Laplace equation [1]:

$$\Delta P = \frac{-4\sigma \cos\theta}{\alpha D} \quad [1]$$

σ : surface tension of the matrix metal melt,

θ : contact angle ($180^\circ > \theta > 0^\circ$) of the matrix metal melt to the reinforcing filler,

α : constant, and

D: particle size of the reinforcing filler.

[0031] The equation [1] indicates that in order that the wettability of the reinforcing filler with the matrix metal melt is improved by surface improvement, etc. of the reinforcing filler particles, the infiltration of the matrix metal melt occurring spontaneously, the condition of $\Delta P < 0$, namely, $\cos \theta > 0$ should be met. However, in a combination of ceramics and an aluminum alloy melt, in general, it is extremely difficult to achieve the condition of $\cos \theta > 0$, with $\Delta P > 0$ in a usual temperature range producing metal-based composite materials. When the condition of $\Delta P < 0$, namely, $\cos \theta > 0$ is achieved, the smaller D (particle size of the reinforcing filler) in the equation [1], the larger the absolute value of ΔP , meaning larger likelihood of the infiltration of the matrix metal melt.

[0032] However, the inventors have confirmed that the larger the particle size of the reinforcing filler (the larger the space between the reinforcing filler), the more easily the infiltration of the matrix metal melt takes place.

This means that what the method of the present invention utilizes is not restricted to improvement in wettability. Accordingly, the generation of reduced pressure in the container greatly contributes to the spontaneous infiltration of the matrix metal melt into the reinforcing filler. Namely, in the present invention, by synergy effects of (a) reacting the infiltration-accelerating metal in the container with oxygen and nitrogen to generate a reduced pressure state in the container, and (b) improving the wettability of the reinforcing filler with the matrix metal melt, the matrix metal melt is spontaneously infiltrated into the reinforcing filler.

[2] Materials

(1) Reinforcing filler

[0033] The reinforcing filler may be selected from ceramics of SiC, Al_2O_3 , SiO_2 , AlN, TiC, TiO_2 , ZrO_2 , Si_3N_4 , sialon, etc. The reinforcing filler may be used in various forms of long or short fibers, particles, etc., and various shapes of reinforcing fillers may be combined. When the reinforcing filler is in the form of particles, its average particle size is preferably about 1-1000 μm , more preferably about 10-100 μm . Also when it is in the form of fibers, its fiber diameter is preferably about 0.1-1000 μm , more preferably about 1-100 μm . The reinforcing filler, when dispersed in the matrix metal, imparts excellent properties such as high rigidity and high wear resistance, etc. to the matrix metal.

(2) Infiltration-accelerating metal

[0034] The infiltration-accelerating metal mixed with the reinforcing filler is preferably pure magnesium or a magnesium alloy, though calcium, zirconium or alloys containing these metals may also be used. The infiltration-accelerating metal is preferably in at least one shape selected from the group consisting of powder, chip, foil, plate and bulk.

(3) Aluminum container

[0035] With an aluminum alloy or a magnesium-aluminum alloy as a matrix metal melt, the use of an aluminum container is preferable because the composition variations of the matrix metal melt do not occur unlike Japanese Patent 2,905,519. Though there is an aluminum oxide film on a surface of the aluminum container, which is not melted at a temperature of a matrix metal melt, this oxide film is reduced to easily meltable aluminum, when the infiltration-accelerating metal is oxidized in the container. Accordingly, holes are formed in the aluminum container in the matrix metal melt, so that the melt enters into the aluminum container.

[0036] The aluminum container may have any structure, as long as (a) it can hold a reinforcing filler and an infiltration-accelerating metal, (b) when immersed in the

matrix metal melt, it is substantially sealed to the outside atmosphere, and (c) it can keep its initial shape until the inside of the aluminum container reaches sufficiently reduced pressure to cause the matrix metal melt enter into the reinforcing filler. Specifically, the aluminum container may be a cylindrical can with a lid, a sealable capsule, or a foil capable of completely wrapping a mixture of the reinforcing filler and the infiltration-accelerating metal.

[0037] In the present invention using an aluminum container meltable in a matrix metal melt, container materials do not remain in boundaries between the matrix metal and the composite material unlike U.S. Patent 3,364,976 using a steel mold, a graphite mold, a glass mold, etc.

(4) Matrix metal

[0038] The matrix metal, to which the pressureless infiltration method of the present invention is applicable, is preferably an aluminum alloy, though a magnesium-aluminum alloy can also be used. In the case of the magnesium-aluminum alloy, the percentage of aluminum is preferably 1% by weight or more, more preferably 3% by weight or more to minimize the variations of a matrix alloy composition due to the melting of the aluminum container.

[0039] By using an aluminum container as a container for holding a mixture of the reinforcing filler and the infiltration-accelerating metal, and by making the atmosphere in the container an oxygen-containing nitrogen gas, the present invention utilizes not only the nitriding reaction of the infiltration-accelerating metal but also the oxidation reaction (exothermic reaction) thereof, resulting in holes formed in the aluminum container even at a relatively low temperature of the matrix metal melt. Accordingly, the temperature of the matrix metal melt may be substantially a casting temperature. Specifically, the temperature of the matrix metal melt is preferably about a temperature between T_m and $T_m + 40^\circ\text{C}$, wherein T_m is the liquidus point of the matrix metal. Because the matrix metal melt at such a relatively low temperature can be used, the metal-based composite material can be obtained at a low cost.

(5) Atmosphere in aluminum container

[0040] To cause a reaction between the infiltration-accelerating metal and oxygen and nitrogen in the container by the temperature of the matrix metal melt, the atmosphere inside the aluminum container should be a gas containing oxygen and nitrogen gas.

[0041] When the oxygen partial pressure is low, the oxidation reaction of the infiltration-accelerating metal is insufficient to reduce oxide film on the aluminum container, making it difficult to melt the aluminum container in the matrix metal melt at a usual temperature and thus making the infiltration of the matrix metal melt less likely. Accordingly, an oxygen gas should exist in the alumi-

num container in a sufficient amount as to oxidize the infiltration-accelerating metal in the aluminum container.

[0042] A nitrogen gas reacts with the infiltration-accelerating metal such as magnesium, etc. in the aluminum container to form nitrides having good wettability with the matrix metal melt. Because the nitrides are attached to the surface of the reinforcing filler, the matrix metal melt is easily infiltrated into the reinforcing filler.

[0043] In order that oxygen and nitrogen effectively exhibit the above functions, the atmosphere inside the aluminum container is preferably an oxygen-containing nitrogen gas having an oxygen partial pressure of about 10-40% and a nitrogen partial pressure of about 90-60%. From the aspect of production cost, it is preferable to use air as the oxygen-containing nitrogen gas. The air makes it unnecessary to use a facility for carrying out atmosphere control such as nitrogen substitution, etc., unlike Japanese Patent 2,905,519. If the container is filled with substantially only a nitrogen gas, for instance, without satisfying this condition, the oxidation reaction (high-temperature exothermic reaction) of the infiltration-accelerating metal and the resultant reduction reaction of an aluminum oxide film do not occur, making it unlikely that the aluminum container is melted.

[2] Pressureless infiltrating apparatus and method

(1) First embodiment

[0044] Fig. 1 schematically shows the first pressureless infiltrating apparatus for conducting the method of the present invention. The apparatus comprises a graphite crucible 1 for containing a matrix metal melt, a push member 6 made of stainless steel, etc. for keeping an aluminum container 4 containing a mixture of the reinforcing filler 2 and the infiltration-accelerating metal 3 immersed in the matrix metal melt 5, a pipe 7 communicating with the aluminum container 4 for measuring pressure variations therein, a pressure sensor 8 disposed at the other end of the pipe 7, and a thermocouple 9 disposed substantially in a center portion of the aluminum container 4.

[0045] The aluminum container 4 may be in any form as long as the inside thereof is substantially shut off from the outside atmosphere, specifically in a shape of a foil, a container with a lid or a capsule. For instance, even a commercially available aluminum foil can be used as the aluminum container 4 of the present invention, because it is shut off from the outside atmosphere when immersed in the matrix metal melt 5. In the case of the container with a lid, the lid need not be fixed to a container body unless the lid is detached during immersion. The capsule is preferable, because the inside of the capsule is unlikely to be communicating with the outside atmosphere during immersion. In any case, the thickness of the aluminum container is not particularly limited unless premature melting occurs by contact with the matrix metal melt. In practical applications, the thickness of the

aluminum container is preferably about 1-2000 μm , more preferably about 10-1000 μm .

[0046] Instead of introducing the aluminum container 4 containing a mixture of the reinforcing filler 2 and the infiltration-accelerating metal 3 into the matrix metal melt 5, the aluminum container 4 may be disposed in advance at the bottom of the graphite crucible 1 before pouring the matrix metal melt 5.

[0047] In one example of producing a composite material with this apparatus, a mixture of the reinforcing filler 2 and the infiltration-accelerating metal 3 is completely wrapped by an aluminum foil or charged into an aluminum can to provide an aluminum container 4. A stainless steel pipe 7 for measuring pressure is caused to pierce into substantially a center portion of this aluminum container 4, and the other end of the pipe 7 is connected to a pressure sensor 8. To measure temperature inside the aluminum container 4, one end of thermocouple 9 is disposed substantially in a center portion of the aluminum container 4, and the other end of thermocouple 9 is connected to a thermometer (not shown).

[0048] An aluminum melt 5 is poured into the graphite crucible 1 heated at a predetermined temperature, into which the aluminum container 4 is slowly charged. Because there is a lot of air in space in the aluminum container 4, the aluminum container 4 tends to float on a melt surface when it is simply introduced into the aluminum melt 5. This breaks an aluminum foil when reduced pressure is generated, causing the outside atmosphere to enter into the aluminum container 4, thereby not only preventing the generation of reduced pressure, but also burning pure magnesium as the infiltration-accelerating metal 3. Accordingly, the aluminum container 4 is pushed down by a stainless steel push member 6, so that the aluminum container 4 does not float to the surface.

[0049] After immersion of the aluminum container 4, the crucible 1 containing the aluminum container 4 is heated at about 620-700°C for 0.5-5 hours in an electric furnace (not shown). During immersion, pressure and temperature variations were measured in the aluminum container 4 to detect the generation of reduced pressure. After completion of turning a portion of the aluminum melt 5 in which the aluminum container 4 exists to have a composite structure, the composite body is cooled in the graphite crucible 1 to obtain a partially reinforced, aluminum-based, composite material, in which only a portion corresponding to the aluminum container 4 has a composite structure.

[0050] Though the aluminum foil is used in this embodiment, an aluminum can containing a mixture of the reinforcing filler and the infiltration-accelerating metal may be disposed in advance at the bottom of the graphite crucible 1, before pouring the aluminum melt 5.

[0051] Also, after an aluminum foil or can is melted, the aluminum melt 5 may be stirred to uniformly disperse the reinforcing filler 2 in the entire aluminum melt 5. Next, the aluminum melt having the reinforcing filler 2

dispersed is poured into an ingot case and solidified to obtain an aluminum-based, reinforced, composite material having the reinforcing filler 2 uniformly dispersed.

5 (2) Second embodiment

[0052] Fig. 2 schematically shows the second pressureless infiltrating apparatus. This apparatus comprises a die 21 for containing a matrix metal melt, and an aluminum container 24 for containing a mixture of the reinforcing filler 22 and the infiltration-accelerating metal 23. Usable as the aluminum container 24 is, for instance, a cylindrical aluminum can to increase pressure reduction effects. After the aluminum can is filled with the mixture, a lid is mounted to the aluminum container 24 to shut it from the outside atmosphere. This aluminum container 24 is placed in a die 21 preheated at a predetermined temperature at a predetermined position in such a manner that the lid is not opened. An aluminum melt 25 is poured into the die 21 in which the aluminum container 24 is disposed, and the die 21 is kept at a predetermined temperature in an electric furnace (not shown). Next, the die 21 is taken out of the electric furnace, and cooled to obtain a partially composite body of aluminum and a reinforcing filler.

[0053] Though the aluminum can with a lid is used in this embodiment, the aluminum can 24 can be sealed, for instance, by disposing the can 24 in a die without a lid, with the opening of the aluminum can abutting against an inner wall of the die 21.

[0054] Instead of using the infiltration-accelerating metal 23 in the form of small pieces as shown in Fig. 2, it may be in a shape of flat plate (Fig. 3), chip (Fig. 4), or bulk (Fig. 5). Also in this embodiment, a green sand mold, a shell mold, a gas-hardening mold such as a CO_2 mold, a self-hardening mold, etc. may be used in place of a die with a cavity.

[0055] The present invention will be explained in further detail referring to EXAMPLES below, without intention of restricting the present invention thereto.

EXAMPLE 1

[0056] 2 kg of SiC particles (GC: #240, average particle size: 58 μm) was heated at 160°C for 2 hours and sufficiently dried. The resultant dried SiC particles were uniformly mixed with 40 g of chip-shaped pure magnesium (available from Chuo Kosan K. K., purity: 99.8%, tradename: CM30) to provide a mixed powder. This mixed powder was completely wrapped with a commercially available aluminum foil (thickness: 15 μm) 4 in the form of a container. As shown in Fig. 1, to measure pressure variations inside the aluminum foil container 4 a stainless steel pipe 7 was used with one end disposed in a substantially center portion of the aluminum container and the other end connected to a pressure sensor 8. Also, to measure the temperature in the aluminum foil container 4, thermocouple 9 was used with one end dis-

posed in a substantially center portion of the aluminum foil container 4 and the other end connected to a thermometer.

[0057] 8 kg of a melt of aluminum (JIS AC4CH alloy) was poured into a graphite crucible (#60) 1 heated about 500°C. The pouring temperature was 620-700°C. As shown in Fig. 1, the aluminum foil container 4 was quietly immersed in the aluminum melt 5 in the graphite crucible 1. To prevent the aluminum foil container 4 from floating, the aluminum foil container 4 was pushed down by a push member 6 made of stainless steel. After immersion, the aluminum foil container 4 was kept at 650°C in an electric furnace (not shown) for one hour.

[0058] While heating for one hour after the immersion of the aluminum foil container 4, the variations of pressure and temperature were measured. The results are shown in Fig. 6. It was confirmed from Fig. 6 that when immersed in the aluminum melt, even the aluminum foil container 4 is shut off from the outside atmosphere, generating reduced pressure therein.

[0059] Though the aluminum melt 5 was turned to a composite material in a portion where the aluminum container 4 was immersed after one hour, the aluminum melt 5 was heated to 700°C while sufficiently stirring with a stirrer (not shown) in the electric furnace to diffuse the SiC particles 2 uniformly in the aluminum melt 5. After removing slag, it was poured into an ingot case (not shown). Thereafter, the aluminum melt 5 was cooled in the ingot case, and the solidified aluminum alloy was taken out. Thus, the entire matrix metal was turned to an SiC particle-reinforced, aluminum-based, composite material.

[0060] Though in this EXAMPLE, the aluminum foil container 4 was immersed in the aluminum melt 5 poured into the graphite crucible 1, the aluminum melt 5 may be poured into the graphite crucible 1 with the aluminum foil container 4 disposed at the bottom thereof in advance, to obtain an SiC particle-reinforced, aluminum-based, composite material.

EXAMPLE 2

[0061] An aluminum-based, composite material uniformly reinforced by SiC particles was obtained in the same manner as in EXAMPLE 1 except for charging mixed powder (SiC particles 2 kg, chip-shaped Mg: 40 g) into a commercially available, cylindrical aluminum can (thickness: 1mm, inner diameter: 100mm, height: 115mm) and closing the can with a lid, instead of being wrapped with an aluminum foil.

EXAMPLE 3

[0062] EXAMPLE 1 was repeated except for using a uniform mixed powder of 20 g of dry SiC particles (GC: #240, average particle size: 58 μm) and 0.4 g of chip-shaped pure magnesium (available from Chuo Kosan K. K., purity: 99.8%, tradename: CM30), and cooling the

aluminum melt in the graphite crucible (#60) 1 without stirring after the aluminum melt was infiltrated into SiC particles, to produce an aluminum-based, composite material partially reinforced by SiC particles, which had a composite structure only in a portion where the aluminum foil container 4 was immersed.

[0063] Though in this EXAMPLE, the aluminum foil container 4 was immersed in the aluminum melt 5 charged in the graphite crucible 1, an SiC particle-reinforced, aluminum-based, composite material was obtained by a method comprising disposing the aluminum foil container 4 at the bottom of the graphite crucible 1 in advance and pouring the aluminum melt 5 into the graphite crucible 1. Also, with a commercially available aluminum can (thickness: 0.5 mm, inner diameter: 17 mm, height: 40 mm) with a lid in place of an aluminum foil, an aluminum-based, composite material partially reinforced by SiC particles was similarly obtained.

20 EXAMPLE 4

[0064] 20 g of SiC particles (GC: #240, average particle size: 58 μm) were heated at 160°C for 2 hours and sufficiently dried. The resultant dried SiC particles were uniformly mixed with 0.4 g of chip-shaped pure magnesium (available from Chuo Kosan K. K., purity: 99.8%, tradename: CM30) to provide mixed powder. This mixed powder was charged into the same commercially available, cylindrical aluminum can 24 as in EXAMPLE 3, and the can 24 was closed with a lid. As shown in Fig. 2, this aluminum container 24 was disposed in a cavity of a die 21 preheated at about 500°C at a predetermined position. 500 g of a melt of an aluminum alloy (Al-6%Si) was poured into the die 21. The pouring temperature was 650°C. The aluminum container 24 immersed in the aluminum alloy melt 25 is substantially shut off from the outside atmosphere with the inside of the container in a sealed state.

[0065] After pouring the aluminum melt 25, the die 21 was kept at 650°C for 1.5 hours in an electric furnace (not shown). Next, the die 21 was taken out of the electric furnace and cooled to obtain an aluminum alloy partially reinforced by SiC particles.

[0066] The resultant aluminum alloy was cut at positions in a portion in which the aluminum container 24 containing SiC particles was disposed and in other portions to observe the structure of the aluminum alloy by an optical microscope. Fig. 7 is a photomicrograph (x 200) showing a cross section structure of a portion in which the aluminum container 24 containing SiC particles was disposed. It was seen in Fig. 7 that a matrix (white portion) of an aluminum alloy (Al-6%Si alloy) well infiltrated between SiC particles (black portion). On the other hand, in the cross section structure in which the aluminum container 24 containing SiC particles was not disposed, only the aluminum alloy matrix was observed. It was confirmed as a result of the above observation that an aluminum-based, partially composite material in

which only a portion corresponding to the aluminum container 24 containing SiC particles had a composite structure was obtained by the method of this EXAMPLE.

EXAMPLE 5

[0067] As shown in Fig. 3, a 0.3-mm-thick, flat plate 33 of pure magnesium (available from Osaka Fuji Corp., purity: 99%) was disposed in the same commercially available, cylindrical aluminum can 24 as in EXAMPLE 3 along its inner wall. 20 g of SiC particles (GC: #240, average particle size: 58 μ m) heated at 160°C for 2 hours and dried were then charged inside the flat plate 33 of pure magnesium. After closing the aluminum container 24 with a lid, the aluminum container 24 was disposed in a cavity of a die 21 preheated at about 500°C.

[0068] 500 g of a melt of an aluminum alloy (AC4CH alloy) was poured into a cavity of the die 21. The pouring temperature was 650°C. The aluminum container 24 immersed in the aluminum alloy melt 25 was substantially shut off from the outside atmosphere with the inside of the container in a sealed state. After pouring an aluminum melt 25, the die 21 was kept at 650°C for 1.5 hours in an electric furnace (not shown). Thereafter, die 21 was taken out of the electric furnace and cooled to obtain an aluminum-based, partially composite material in which only a portion corresponding to the aluminum container 24 containing SiC particles was reinforced.

EXAMPLE 6

[0069] As shown in Fig. 4, 1.0 g chip-shaped pure magnesium (available from Chuo Kosan K. K., purity: 99.8%, tradename: CM30) 43 was placed at the bottom of the same commercially available, cylindrical aluminum can 24 as in EXAMPLE 3. Next, 20 g of SiC particles (GC: #240, average particle size: 58 μ m) heated at 160°C for 2 hours and dried were stacked on the chip-shaped pure magnesium 43. Without mixing the chip-shaped pure magnesium 43 and the SiC particles 22, the container 24 was closed with a lid. This aluminum container was disposed in a die 21 preheated at about 500°C.

[0070] A melt 25 of an aluminum alloy (AC4CH) was poured into the die 21 in the same manner as in EXAMPLE 5, and kept at 650°C for 1.5 hours in an electric furnace (not shown). The aluminum container 24 immersed in the aluminum alloy melt 25 was substantially shut off from the outside atmosphere with the inside of the container sealed. After cooling the aluminum alloy melt 25, an aluminum-based, partially composite material, in which only a portion corresponding to the aluminum container 24 containing SiC particles was reinforced, was obtained.

EXAMPLE 7

[0071] As shown in Fig. 5, 1.0 g of a commercially

available, bulk-shaped, pure magnesium 53 was placed at the bottom of the same commercially available, cylindrical aluminum can 24 as in EXAMPLE 3. Next, 20 g of SiC particles (GC: #240, average particle size: 58 μ m) heated at 160°C for 2 hours and dried were stacked thereon. After closing the container 24 with a lid, the container 24 was disposed in a cavity of a die 21 preheated at about 500°C.

[0072] A melt of an aluminum alloy (AC4CH) was poured into the die 21 in the same manner as in EXAMPLE 5 and kept at 650°C for 1.5 hours in an electric furnace (not shown). The aluminum container 24 immersed in the aluminum alloy melt 25 was substantially shut off from the outside atmosphere with the inside of the container in a sealed state. After cooling the aluminum alloy melt 25, an aluminum-based, partially composite material, in which only a portion corresponding to the aluminum container 24 containing SiC particles was reinforced, was obtained.

COMPARATIVE EXAMPLE 1

[0073] After 20 g of SiC particles (GC: #240, average particle size: 58 μ m) were heated at 160°C for 2 hours and sufficiently dried, they were uniformly mixed with 0.4 g of chip-shaped pure magnesium (available from Chuo Kosan K. K., purity: 99.8%, tradename: CM30) to provide mixed powder.

[0074] As shown in Fig. 2, the mixed powder was charged into the same commercially available, cylindrical aluminum can 24 as in EXAMPLE 3. The cylindrical aluminum can 24 was placed without a lid in a vacuum apparatus (not shown), and the pressure inside the vacuum apparatus was reduced to about 10 kPa. A nitrogen gas of 99% in concentration was then introduced into the vacuum apparatus until the pressure of the vacuum apparatus reached atmospheric pressure, and this cycle was repeated four times to conduct nitrogen substitution in the cylindrical aluminum can 24. Thereafter, the aluminum container 24 was closed with a lid. This aluminum container 24 was disposed in the cavity of the die 21 preheated at about 500°C.

[0075] 500 g of aluminum alloy (Al-6%Si) melt was poured into the cavity of the die 21. The pouring temperature was 650°C. After pouring an aluminum melt 25, it was kept in an electric furnace (not shown) at 650°C for 1.5 hours. Next, it was taken out of the electric furnace together with the die 21 and cooled to obtain a partially composite aluminum alloy. When the partially composite aluminum alloy was cut in a portion in which the aluminum container 24 containing SiC particles was disposed, it was found that the aluminum container 24 substantially remained without melting, with SiC particles easily detached. This indicates that with an aluminum container filled with a nitrogen atmosphere, the oxidation exothermic reaction of magnesium did not take place, failing to remove an oxide film from an aluminum container surface and resulting in insufficient degree of

evacuation, and that accordingly the aluminum container was not provided with openings at as low a melt temperature as 650°C, so that the aluminum alloy melt did not sufficiently enter between SiC particles.

[0076] As described above in detail, by using the method of the present invention, a reinforcing filler can be impregnated with a matrix metal melt without pressure and at a relatively low temperature without necessitating atmosphere control. Accordingly, without necessitating a special facility that was conventionally required to produce metal-based composite materials, it is possible to achieve drastic reduction of production cost. Further, it is possible to produce metal-based materials partially composite with reinforcing fillers.

Claims

1. A method for producing a metal-based composite material comprising a matrix metal containing at least partially a reinforcing filler, comprising (1) charging at least one reinforcing filler in the form of fibers or particles and at least one infiltration-accelerating metal into an aluminum container; (2) immersing said aluminum container in a melt of a matrix metal made of an aluminum alloy or a magnesium-aluminum alloy in a state that an oxygen-containing nitrogen gas remains in said aluminum container; (3) melting said aluminum container in said matrix metal melt, so that said matrix metal melt is infiltrated into said reinforcing filler, and then (4) solidifying said matrix metal melt.
2. The method for producing a metal-based composite material according to claim 1, wherein only a predetermined portion of said matrix metal is turned composite with said reinforcing filler.
3. The method for producing a metal-based composite material according to claim 1 or 2, wherein said aluminum container is introduced into a matrix metal melt in a crucible.
4. The method for producing a metal-based composite material according to claim 1 or 2, wherein said aluminum container is disposed in a cavity of a die or a mold at a predetermined position in advance, and said matrix metal melt is charged into said cavity.
5. The method for producing a metal-based composite material according to any one of claims 1-4, wherein an aluminum foil is used as said aluminum container, and a mixture of said reinforcing filler and said infiltration-accelerating metal is completely wrapped by said aluminum foil.
6. The method for producing a metal-based composite material according to any one of claims 1-4, wherein

an aluminum can with a lid is used as said aluminum container, and after charging a mixture of said reinforcing filler and said infiltration-accelerating metal into said aluminum can, said aluminum can is closed by said lid.

7. The method for producing a metal-based composite material according to any one of claims 1-6, wherein after said matrix metal melt is infiltrated into said reinforcing filler, said matrix metal melt is stirred to uniformly disperse said reinforcing filler in said matrix metal melt.
8. The method for producing a metal-based composite material according to any one of claims 1-7, wherein said oxygen-containing nitrogen gas is air.
9. The method for producing a metal-based composite material according to any one of claims 1-8, wherein a space percentage of said container is 30-70%.
10. The method for producing a metal-based composite material according to any one of claims 1-9, wherein said infiltration-accelerating metal is at least one metal selected from the group consisting of magnesium, calcium, zirconium and alloys containing these metals.
11. The method for producing a metal-based composite material according to claim 10, wherein said infiltration-accelerating metal is pure magnesium or magnesium alloy.
12. The method for producing a metal-based composite material according to any one of claims 1-11, wherein said infiltration-accelerating metal is in the form of at least one selected from the group consisting of powder, chip, foil, plate and bulk.
13. The method for producing a metal-based composite material according to any one of claims 1-12, wherein said reinforcing filler is made of a ceramic.
14. The method for producing a metal-based composite material according to claim 13, wherein said ceramic is SiC.
15. The method for producing a metal-based composite material according to any one of claims 1-14, wherein the temperature of said matrix metal melt is between T_m and $T_m + 40^\circ\text{C}$, wherein T_m is the liquidus point of said matrix metal.

Fig. 1

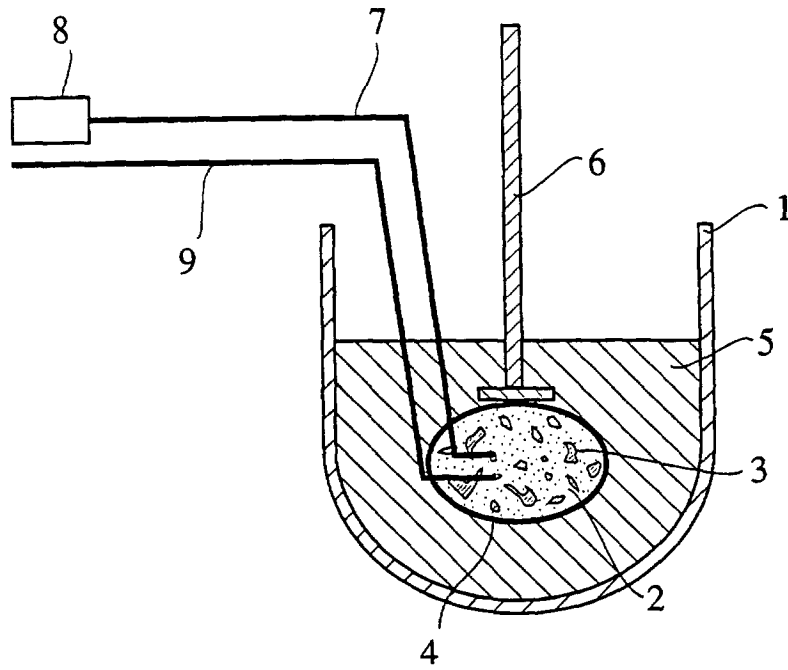


Fig. 2

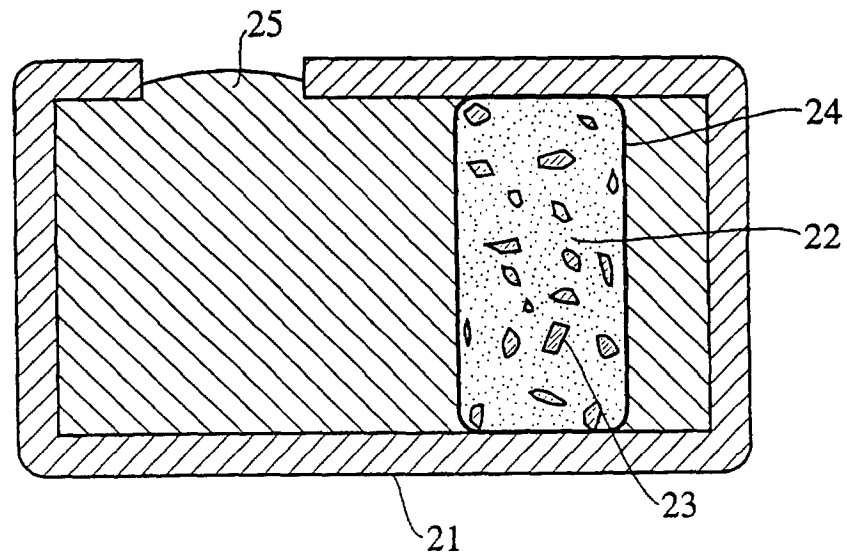


Fig. 3

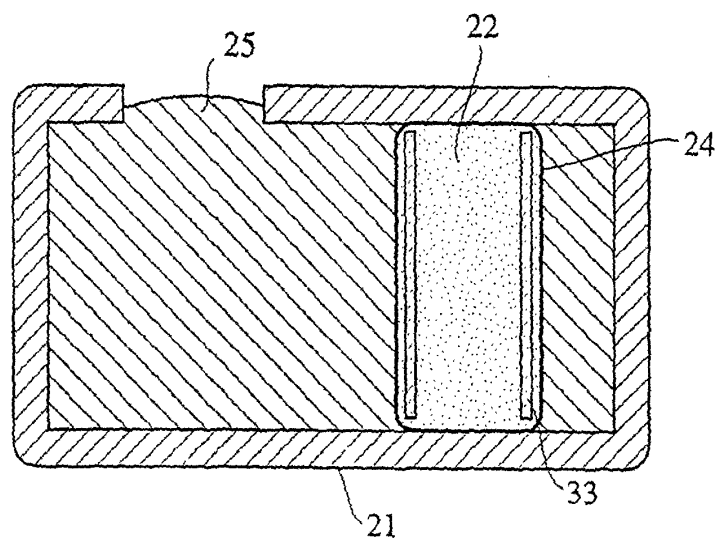


Fig. 4

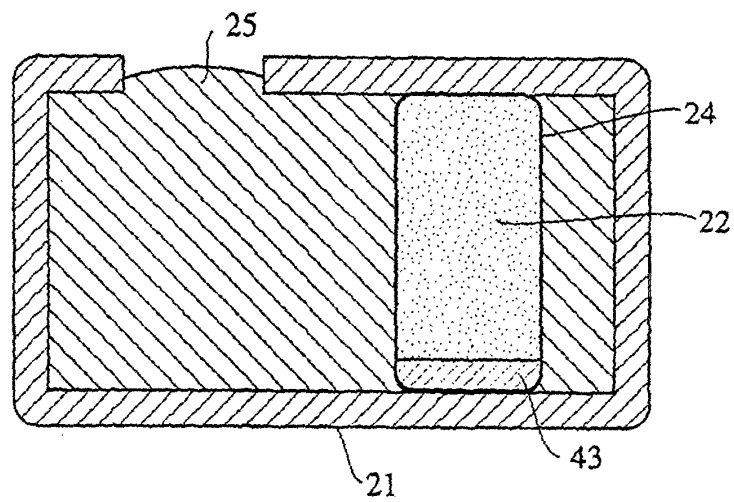


Fig. 5

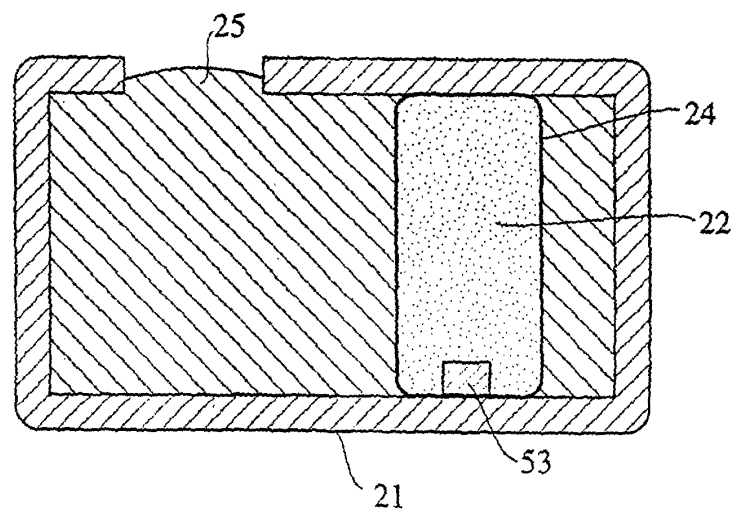


Fig. 6

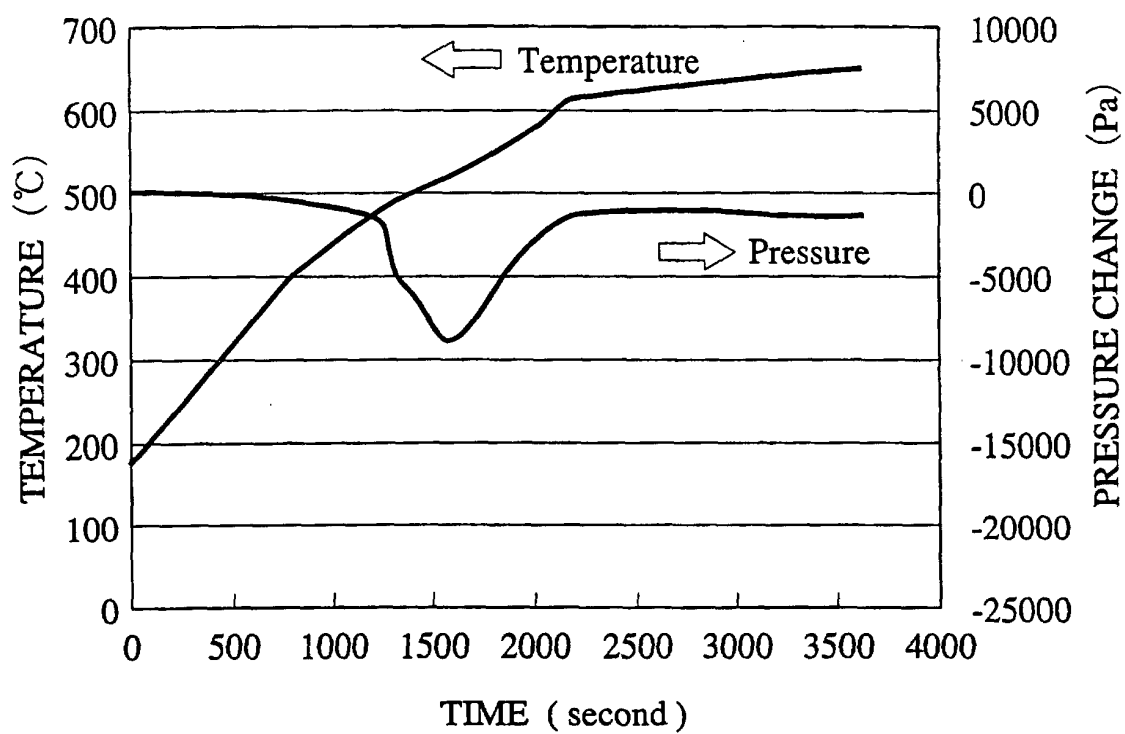
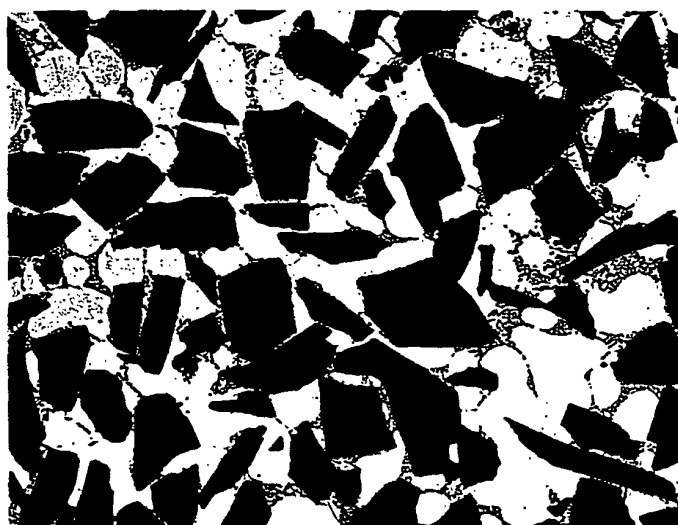


Fig. 7



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP00/07745

A. CLASSIFICATION OF SUBJECT MATTER Int.Cl. ⁷ C22C47/10		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED		
Minimum documentation searched (classification system followed by classification symbols) Int.Cl. ⁷ C22C47/00-49/14, B22D19/14		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Jitsuyo Shinan Koho 1926-1996 Toroku Jitsuyo Shinan Koho 1994-2001 Kokai Jitsuyo Shinan Koho 1971-2001 Jitsuyo Shinan Toroku Koho 1996-2001		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US, 5000249, A (Lanxide Technology Company, LP), 19 May, 1991 (19.05.91) & JP, 2905519, B2 & EP, 368787, A1	1-15
A	US, 3364976, A (The Dow Chemical Company), 23 January, 1968 (23.01.68), & DE, 1508821, A1 & FR, 1470259, A	1-15
A	US, 5669434, A (Honda Giken Kogyo Kabushiki Kaisha), 23 July, 1997 (23.07.97) & JP, 8-157982, A & DE, 19539922, A1	1-15
A	JP, 2576188, B2 (Toyota Motor Corporation), 07 November, 1996 (07.11.96) (Family: none)	1-15
A	JP, 6-93354, A (Agency of Industrial Science and Technology), 05 April, 1994 (05.04.94) (Family: none)	1-15
A	JP, 6-38978, B2 (Toyota Motor Corporation), 25 May, 1994 (25.05.94) (Family: none)	1-15
☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search 11 January, 2001 (11.01.01)		Date of mailing of the international search report 23 January, 2001 (23.01.01)
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer
Facsimile No.		Telephone No.

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP00/07745

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP, 60-2149, B2 (Toyota Motor Corporation), 19 January, 1985 (19.01.85) (Family: none)	1-15

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