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(71) Applicant: **TOUDAI TLO, Ltd.**
Tokyo 113-0033 (JP)

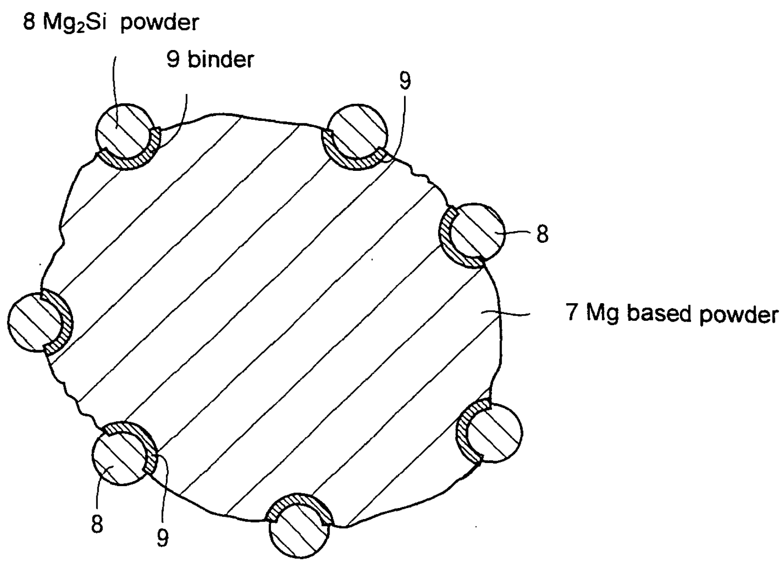
(72) Inventor: **KONDOH, Katsuyoshi,**
The University of Tokyo
Meguro-ku, Tokyo 153-0041 (JP)

(74) Representative: **Kirschner, Klaus Dieter**
Kirschner & Kollegen
Südliche Münchnerstrasse 53
82031 Grünwald (DE)

(54) MAGNESIUM-BASE COMPOSITE POWDER, MAGNESIUM-BASE ALLOY MATERIAL AND METHOD FOR PRODUCTION THEREOF

(57) Magnesium based composite powder that is a starting raw material to manufacture a Mg_2Si dispersion type of magnesium based composite material comprises Mg based powder (7) that is a main component constituting the matrix of a magnesium alloy, and Mg_2Si powder attached to the surface of the Mg based powder (7) through a binder (9).

FIG. 3



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Description

TECHNICAL FIELD

[0001] The present invention relates to a magnesium based composite powder containing Mg_2Si having high rigidity, a magnesium based alloy base material, and manufacturing methods of them.

BACKGROUND ART

[0002] A magnesium alloy is characterized by light in weight because of low specific gravity and commercialized and practically used as a package of a mobile phone and portable audio device mainly. In designing a product or a member, the rigidity of a base material is an important factor as well as strength and hardness.

[0003] For example, in a case where a magnesium alloy is applied to a housing case for an automatic transmission (AT), even when it has the same tensile strength and creep strength as that of a presently used aluminum alloy (ADC12, for example), since the rigidity (Youngs modulus) of an existing magnesium alloy is about 60% of the aluminum alloy, it is bend and deformed when a load is applied in case of the same dimension and thickness. Therefore, when the magnesium alloy is practically used, since it is necessary to increase the thickness in accordance with a product and member to apply, the effect of its light in weight cannot be provided.

[0004] To improve the rigidity of a metal material including the magnesium alloy, it is effective in general to use a method of diffusing compound grains having rigidity higher than that metal material, that is, to use a composite material. For example, since magnesium silicide (Mg_2Si) has Young's modulus of 120GPa and it is considerably higher than a general magnesium alloy having Young's modulus of 43 to 44GPa, rigidity can be improved in a composite material in which its grains are dispersed in an alloy.

[0005] However, when a Mg-Si group alloy is manufactured by melting and casting methods, an eutectic point exists in the vicinity where a Si content is about 1% by weight. Thus, when Si is added far beyond 1% by weight, Mg_2Si formed by the reaction with Mg grows to be coarse. When the magnesium alloy contains such coarse Mg_2Si particles, strength and toughness are lowered due to stress concentration of the coarse particles, and explosion could be generated in the course of melting because heat is generated due to the reaction between Mg and Si. In addition, since the coarse Mg_2Si particles exist, performance in casting to a mold (casting property) is lowered and many defects and holes exist in the casting alloy base material. Thus, AS21 alloy (Mg-2%Al-1%Si) or AS41 alloy (Mg-4%Al-1%Si) and the like is used as Mg_2Si containing magnesium alloy that can be manufactured by the melting and casting method. However, when 1% by weight of Si is added, a volume ratio of the generated Mg_2Si particles is less than 1% of the total volume, it is difficult to improve the rigidity of the magnesium alloy satisfactorily.

[0006] Meanwhile, S. K. THAKUR and the like (adapted from Metallurgical and Materials Transactions A, Vol. 35A, March 2004, p.1167-1176) proposes a manufacturing method of magnesium alloy containing Mg_2Si particles using an infiltration method in which a preform provided by solidifying mixed powder of three kinds including Si powder is prepared and it is impregnated with a melted magnesium alloy while a pressure is applied. However, according to this method, Mg_2Si particle formed by the reaction between a molten Mg alloy and Si grows in the course of reaction and becomes as coarse as 70 to 100 μm finally in the magnesium alloy. As a result, the above various problems arise in performance.

[0007] The inventor of the present invention disclosed a technique for manufacturing a magnesium based composite material in which Mg_2Si particles are diffused using a powder metallurgy method in Japanese Patent Application No. 2003-2602 (filed on January 8, 2003). Here, the inventors proposes a magnesium composite powder in which fine Si powder or SiO_2 powder is attached on the surface of magnesium based alloy powder by a mechanical bonding method or a bonding method using a binder, and its manufacturing method. Furthermore, a warm plastic process is performed to the composite powder to generate Mg_2Si using the solid-phase reaction of Mg with Si or SiO_2, and to finally provide the magnesium based composite material in which Mg_2Si particles are uniformly dispersed.

[0008] According to the magnesium based composite material provided by the technique disclosed in Japanese Patent Application No. 2003-2602, high tensile strength is provided but it is necessary to heat to a high temperature (400 to 550°C, for example) for the reaction of Mg with Si or SiO_2. In this process, the magnesium crystal grain grows to be coarse. In other words, in order to implement further high strength, the heating temperature is preferably low, however, it is difficult to lower the temperature to about 300°C in view of the above solid-phase reaction.

DISCLOSURE OF THE INVENTION

[0009] It is an object of the present invention to provide high-rigidity and high-strength magnesium alloy containing a lot of fine Mg_2Si particles without high-temperature heating.

[0010] The inventor of the present invention has found that it is effective to use magnesium based composite powder in which Mg_2Si particles exist on the surface and/or inside of the Mg based powder when a magnesium based composite material in which Mg_2Si particles are dispersed is manufactured. The inventor has found that the following advantages can be provided by using Mg_2Si particles, instead of the method in which Mg_2Si particle is synthesized by the solid-phase reaction of Mg powder with Si grain as disclosed in Japanese Patent Application No. 2003-2602.

(1) Since heating to high temperature of 400 to 550°C required for promoting the above Si-Mg reaction is not needed, the Mg crystal grain of a matrix is prevented from growing to become coarse and the strength of Mg based alloy is prevented from being lowered.
(2) Since heat generation in the course of the Si-Mg reaction is avoided, Mg_2Si particle and Mg crystal grain are prevented from becoming coarse.

[0011] That is, a magnesium based alloy can be manufactured through a warm extruding process at 200 to 400°C without a high-temperature heat treatment for the solid-phase reaction between Mg and Si. As a result, there can be provided a high-rigidity and high-strength magnesium based alloy containing a lot of fine Mg_2Si particles.
[0012] A magnesium based composite powder according to the present invention comprises magnesium based powder, and magnesium silicide (Mg_2Si) that is dispersed on the surface and/or inside of the Mg based powder.
[0013] According to the magnesium based composite powder, the maximum particle diameter of the Mg_2Si particle is not more than 50μ m, preferably not more than 20 μm, and more preferably not more than 5 μ m. In addition, it is preferable that the Mg based composite powder contains 5 to 60% by volume of Mg_2Si.
[0014] A magnesium based alloy base material according to the present invention is provided by compacting and sintering the magnesium based composite powder, and Mg_2Si particles are dispersed in the matrix.
[0015] According to one embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder.
b) A step of applying a binder to the surface of the Mg based powder.
c) A step of bonding the Mg_2Si powder to the surface of the Mg based powder by mixing and stirring the Mg based powder to which the binder has been applied and the Mg_2Si powder.

[0016] According to another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder.
b) A step of blending the Mg based powder and the Mg_2Si powder and mechanically mixing them to bond the Mg_2Si powder to the surface of the Mg based powder mechanically.

[0017] According to the above methods, the method of bonding the Mg_2Si powder to the surface of the Mg based powder mechanically is such that the blended powder is mechanically mixed using a ball mill, a mixing and grinding mill, a roller compactor, or a rolling machine, for example.
[0018] According to still another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder.
b) A step of mixing the Mg based powder and the Mg_2Si particles.
c) A step of manufacturing a Mg based powder compact in which the Mg_2Si particles are dispersed by compacting the mixed powder in a mold.
d) A step of manufacturing a Mg based sintered alloy in which the Mg_2Si particles are dispersed by sintering the Mg based powder compact.
e) A step of powdering the Mg based sintered alloy by mechanical grinding or cutting.

[0019] According to still another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and silicon (Si) powder.
b) A step of mixing the Mg based powder and the Si powder.
c) A step of manufacturing a Mg based powder compact in which the Si particles are dispersed by compacting the mixed powder in a mold.
d) A step of synthesizing Mg_2Si by heating the Mg based powder compact to react Mg with Si, and at the same time manufacturing a Mg based sintered alloy in which the Mg_2Si particles are dispersed.

e) A step of powdering the Mg based sintered alloy by mechanical grinding or cutting.

[0020] According to still another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of inputting magnesium silicide (Mg_2Si) powder to molten magnesium based alloy and stirring it.
b) A step of manufacturing a casting base material by casting the molten material in a mold.
c) A step of powdering the casting base material by mechanical grinding or cutting.

[0021] According to still another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder.
b) A step of mixing the Mg based powder and the Mg_2Si powder.
c) A step of manufacturing a Mg based powder compact in which the Mg_2Si particles are dispersed by compacting the mixed powder in a mold.
d) A step of manufacturing a Mg_2Si particle dispersed Mg based casting base material by dissolving and casting the Mg based powder compact.
e) A step of powdering the casting base material by mechanical grinding or cutting.

[0022] According to still another embodiment, a manufacturing method of magnesium based composite powder in the present invention comprises the following steps.

a) A step of preparing magnesium (Mg) based powder and silicon (Si) powder.
b) A step of mixing the Mg based powder and the Si powder.
c) A step of manufacturing a Mg based powder compact in which the Si particles are dispersed by compacting the mixed powder in a mold.
d) A step of synthesizing Mg_2Si by heating the Mg based powder compact to react Mg with Si, and at the same time manufacturing a Mg based sintered alloy in which the Mg_2Si particles are dispersed.
e) A step of manufacturing a Mg_2Si particle dispersed Mg based casting base material by dissolving and casting the Mg based sintered alloy.
f) A step of powdering the casting base material by mechanical grinding or cutting.

[0023] A manufacturing method of a magnesium based alloy base material according to the present invention comprises a step of compacting the above described magnesium based composite powder, a step of heating the powder compact to 200 to 400°C in an inert gas atmosphere or a non-oxygenated gas atmosphere, and a step of extruding the powder compact to a dense state immediately after the heating.
[0024] The characteristics and the working effect of the present invention will be described in the following.

BRIEF DESCRIPTION OF DRAWINGS

[0025]

Fig. 1 is a schematic view showing one example of a method of bonding Mg_2Si powder onto the surface of Mg based powder with a binder;
Fig. 2 is a schematic view showing another example of a method of bonding Mg_2Si powder onto the surface of Mg based powder with a binder;
Fig. 3 is a schematic view showing one example of Mg based composite powder in which the Mg_2Si powder are attached on the Mg based powder;
Fig. 4 is a schematic view showing another example of Mg based composite powder in which the Mg_2Si powder are attached on the Mg based powder;
Fig. 5 is a view showing one example of a method of manufacturing Mg based composite powder;
Fig. 6 is a schematic view showing one example of Mg based composite powder in which the Mg_2Si particles are dispersed in the matrix of the Mg based powder;
Fig. 7 is a view showing another example of the method of manufacturing the Mg based composite powder;
Fig. 8 is a view showing still another example of the method of manufacturing the Mg based composite powder;
Fig. 9 is a view showing still another example of the method of manufacturing the Mg based composite powder;
Fig. 10 is a view showing still another example of the method of manufacturing the Mg based composite powder;

Fig. 11 is a microscope photograph showing one example of a sectional structure of the Mg based composite powder in which the Mg_2Si particles are dispersed in the matrix of the Mg based alloy;
Fig. 12 is a view showing one example of X-ray diffraction of the Mg based composite powder; and
Fig. 13 is a view showing another example of X-ray diffraction of the Mg based composite powder.

BEST MODE FOR CARRYING OUT THE INVENTION

[0026]

(1) Mg based composite powder

(A) Mg_2Si content
When it is assumed that the whole volume of magnesium based composite powder is 100%, 5 to 60% by volume of Mg_2Si is contained. In addition, in view of machine processability (machinability) of magnesium based alloy provided by solidifying the composite powder, a more preferable Mg_2Si content is 20 to 40% by volume. When the Mg_2Si content is less than 5%, the magnesium alloy does not have sufficient rigidity. On the other hand, when the Mg_2Si content is more than 60%, the Mg_2Si particles are segregated and aggregated in the magnesium based composite powder containing the Mg_2Si particles, so that the magnesium based alloy provided by solidifying that powder is low in strength and toughness. Thus, to provide the same level of rigidity as an aluminum alloy and to secure superior strength and machinability, a more preferable Mg_2Si content is 20 to 40% by volume.
(B) Maximum particle diameter of Mg_2Si
The maximum particle diameter of Mg_2Si contained in the magnesium based composite powder is to be not more than 50 μ m, preferably not more than 20μm, and more preferably not more than 5μm. When the maximum particle diameter of the Mg_2Si particle is more than 50μm, the mechanical property and machinability of the provided magnesium based alloy is lowered. When the value is not more than 20 μ m, even if 40% by volume of Mg_2Si particles is contained, preferable machinability can be maintained. Furthermore, when the maximum particle diameter of the Mg_2Si particle is not more than 5μ m, the machinability of the magnesium based alloy is improved and at the same time tensile strength of that alloy is improved because of dispersion of fine Mg_2Si particles.
(C) Rigidity (Young's modulus)
The Young's modulus of the magnesium based alloy is 48 to 90GPa. When the Young's modulus is less than 48GPa, an increase rate of the existing magnesium alloy with respect to the Young's modulus becomes 10% or less, it is difficult to apply the alloy to a cover case associated part for a car or a package part of a personal computer or a portable device. On the other hand, when the Young's modulus is more than 90GPa, since the Mg_2Si content is 60% or more by volume, the toughness and the machinability of the alloy base material are lowered as described above.

(2) Manufacturing method of magnesium based composite powder

(A) Magnesium based composite powder using bonding with binder
Figs. 1 and 2 show manufacturing methods of the Mg based composite powder using a binder solution, and Fig. 3 schematically shows the sectional structure of the Mg based composite powder provided by the methods. According to the above methods, the composite powder is manufactured by a wet granulating machine or a spray dryer. According to the method shown in Fig. 1, a mixture 2 of Mg based powder and Mg_2Si powder is inputted into a container 1, and warm air 3 is supplied from the lower part of the container to float the mixture 2. In this state, a binder solution 4 is sprayed from the above to the mixture 2 to apply the binder to the surface of each powder particle and at the same time to dry it at high temperature. As a result, as shown in Fig. 3, Mg_2Si powder 8 is attached and bonded to the surface of a Mg based powder 7 through a binder 9.
According to the method shown in Fig 2, in a state where the mixture of Mg based powder and Mg_2Si powder is floated by a relatively low air amount in the container 1, the binder solution 4 is perpendicularly sprayed with respect to the air flow direction from beneath.
In addition, although it is not shown, Mg_2Si powder can be attached and bonded to the surface of Mg based powder through a binder similarly by mixing and stirring the Mg_2Si powder in the binder solution and spray and applying the binder solution to the Mg based powder floated by the warm air.
Furthermore, according to another method, a predetermined amount of Mg based powder is inputted into the container and 0.2 to 0.5% by weight of olein acid serving as a binder is added to the Mg based powder and then, the olein acid is applied to the surface of the Mg based powder in the container by vibrating or rotating the whole container. Then, Mg_2Si powder is added into the container and the Mg_2Si powder is attached to the Mg based powder to which the olein acid has been applied by vibrating or rotating the container again. Thus,

there is provided Mg based composite powder as shown in Fig. 3

(B) Magnesium based composite powder provided by mechanical bonding

Meanwhile, according to a method of mechanical bonding, Mg based powder and Mg_2Si powder are mixed and inputted to a ball mill, a mixing grinding mill, a roller compactor, a rolling machine and the like, and compressing and shearing processes and the like are applied to the mixture. As a result, there is provided a Mg based granulated substance in which the Mg_2Si powder is mechanically attached and bonded to the surface of the Mg based powder surface. According to need, Mg based composite powder having a predetermined dimension and configuration having a sectional structure as shown in Fig. 4 can be provided from this granulated substance using a grinding and sieving machine. According to Mg based composite powder 15 shown in Fig. 4, Mg_2Si powder 8 is mechanically bonded and attached to the surface of Mg based powder 7.

(C) Mg based composite powder using Mg_2Si particle dispersion type of Mg based sintered alloy

(a) Method shown in Fig. 5

Mg based powder and Mg_2Si powder are prepared as a starting material and both are mixed at a predetermined blend ratio and stirred and inputted into a mold. Then, it is solidified by pressure, whereby a Mg based powder compact in which Mg_2Si particles are dispersed is manufactured.

When the above powder compact is heated to a temperature lower than a melting point of the Mg based powder in an inert gas or non-oxygenated gas or vacuum, a Mg_2Si particle dispersion type of Mg based sintered alloy is provided by solid-phase diffusion among Mg based powder.

When the Mg_2Si particle dispersion type of Mg based sintered alloy is powdered by mechanical processing such as grinding using a ball mill or a crusher mill or cutting, a Mg based composite powder 16 having a predetermined dimension and configuration in a sectional structure is provided as shown in Fig 6. According to the Mg based composite powder 16 shown in Fig. 6, Mg_2Si particles are mainly dispersed in the matrix of Mg based powder 7.

In addition, in the above Mg_2Si particle dispersion type of Mg based sintered alloy, when the Mg_2Si content is more than 60% by volume, the Mg_2Si particles are segregated and aggregated and a problem in machinability in which a tool is damaged in the cutting process is generated. In view of the above, it is preferable that the Mg_2Si content is not more than 60%.

(b) Method shown in Fig. 7

Mg based powder and Si powder are prepared as a starting material and both are mixed at a predetermined blend ratio and stirred and inputted into a mold. Then, it is solidified by pressure, whereby a Mg based powder compact in which the Si particles are dispersed is manufactured.

When the above powder compact is heated to a temperature lower than a melting point of the Mg based powder in an inert gas or non-oxygenated gas or vacuum, Mg_2Si is synthesized by a solid-phase reaction between Si-Mg and at the same time, a Mg_2Si particle dispersion type of Mg based sintered alloy is provided by solid-phase diffusion among Mg based powder.

When the Mg_2Si particle dispersion type of Mg based sintered alloy is powdered by mechanical processing such as grinding using a ball mill or a crusher mill or cutting, Mg based composite powder containing Mg_2Si particles and having a predetermined dimension and configuration in a sectional structure is provided as shown in Fig 6.

(D) Magnesium based composite powder using Mg_2Si particle dispersion type of Mg based casting alloy

(a) Method shown in Fig. 8

Mg_2Si powder prepared as a starting material are inputted to a molten Mg based alloy and stirred and cast into a mold. In the Mg based casting alloy taken out of the mold, added Mg_2Si powder is uniformly dispersed. When the above casting alloy is powdered by mechanical processing such as grinding using a ball mill or a crusher mill or cutting, Mg based composite powder containing Mg_2Si particles and having a predetermined dimension and configuration in a sectional structure is provided as shown in Fig 6. In addition, the dissolution temperature of the molten Mg based alloy after the Mg_2Si powder has been inputted is set to be less than the solidus line between Mg and Mg_2Si in a Mg-Si phase equilibrium diagram. Meanwhile, when it is heated to the solidus line temperature or more, Mg_2Si becomes a solid solution in the molten Mg based alloy and Mg_2Si grows to be coarse in the course of solidification after casting.

(b) Method shown in Fig. 9

Mg based powder and Mg_2Si powder are prepared as a starting material and both are mixed at a predetermined blend ratio and stirred and inputted into a mold. Then, it is solidified by pressure, whereby a Mg based powder compact in which Mg_2Si particles are dispersed is manufactured. This powder compact is inputted into a crucible and heated to manufacture molten Mg based alloy in which Mg_2Si particles are dispersed. Then, it is sufficiently stirred and cast in a mold.

In a Mg based casting alloy taken out of the mold, the added Mg_2Si powder is uniformly dispersed and when this alloy is powdered by mechanical processing such as grinding using a ball mill or a crusher mill or cutting, Mg based composite powder containing Mg_2Si particles and having a predetermined dimension and configuration in a sectional structure is provided as shown in Fig 6.

(c) Method shown in Fig. 10

Mg based powder and Si powder are prepared as a starting material and both are mixed at a predetermined blend ratio and stirred and inputted into a mold. Then, it is solidified by pressure, whereby a Mg based powder compact in which Si particles are dispersed is manufactured.

When the above powder compact is heated to a temperature lower than a melting point of the Mg based powder in an inert gas or non-oxygenated gas or vacuum, Mg_2Si is synthesized by a solid-phase reaction between Si-Mg and at the same time, a Mg_2Si particle dispersion type of Mg based sintered alloy is provided by solid-phase diffusion among Mg based powder.

This powder compact is inputted into a crucible and heated to manufacture molten Mg based alloy in which Mg_2Si particles are dispersed. Then, it is sufficiently stirred and cast in a mold. In a Mg based casting alloy taken out of the mold, the added Mg_2Si powder is uniformly dispersed and when this alloy is powdered by mechanical processing such as grinding using a ball mill or a crusher mill or cutting, Mg based composite powder containing Mg_2Si particles and having a predetermined dimension and configuration in a sectional structure is provided as shown in Fig 6.

In addition, since cutting oil used in the cutting process is attached to the Mg based composite powder, the above is to be used as a raw material after the cutting oil component has been removed by a cleaning process.

(3) Magnesium based alloy base material

The above Mg based composite powder containing the Mg_2Si particles as a starting raw material is solidified to provide a Mg based alloy in which the Mg_2Si particles are dispersed.

[0027] After a powder compact is provided from the Mg based composite powder, it is heated up and processed by extruding, forging or rolling. At this time, it is preferable that the heating temperature of the compact is about 200 to 400°C. When the heating temperature is lower than 200°C, the extruding process is hard to perform in some cases. On the other hand, when it is higher than 400°C, as the extruding process is speeded up, the temperature of the material after the extruding process is raised, which could lower the strength because of the course crystal grain.

[0028] In the Mg based alloy provided as described above, fine Mg_2Si particles are uniformly dispersed. Since the particle diameter of the Mg_2Si particle dispersed in the alloy is the same as the particle diameter in the Mg based composite powder, the maximum particle diameter of the Mg_2Si particle in the Mg based alloy is not more than 50 μ m, preferably not more than 20μ m, and more preferably not more than 5 μ m. In addition, the Mg based alloy contains 5 to 60% by volume of Mg_2Si. As a result, the Mg based alloy has high rigidity and high strength.

EXAMPLE 1

[0029] Pure Mg powder (purity is 99.9% and average particle diameter is 350 μ m) and Si powder (purity is 99.9% and average particle diameter is 22μ m) were prepared and both powder were mixed at the ratio such that Mg : Si = 2 : 1 (molar ratio) and a mixing process was performed with a ball mill for 30 minutes. The mixed powder was inputted into a carbon mold (inner diameter is 35mm ϕ) and set in a spark plasma sintering machine and sintered for 15 minutes at a sample temperature of 600°C under a pressure of 100MPa in vacuum. As a result, a disk-shaped sample comprising Mg_2Si and having an outer diameter of 35mm ϕ and a thickness of 12mm was provided.

[0030] The disk-shaped sample was ground by a jet mill machine and finely ground and sieved to become a Mg_2Si powder having a maximum particle diameter of 15 μ m or less, so that the Mg_2Si powder as a starting raw material was manufactured. On the other hand, as Mg based powder, AZ31 (nominal composition is Mg-3Al-1Zn/mass%) alloy powder having a diameter of approximately 2mm was prepared.

[0031] Based on the method shown in Fig. 5, the AZ31 powder and the Mg_2Si powder were mixed at a predetermined ratio and inputted into a mold having a diameter of 60mm ϕ . A pressure of 400MPa was applied to it to manufacture a powder compact. This powder compact was sintered at 550°C for 1 hour in a nitrogen gas atmosphere, whereby a AZ31 sintered alloy in which Mg_2Si particles are dispersed was provided. Then, Mg based composite powder having a diameter of 0.5 to 3mm was manufactured from the sintered alloy by a cutting process.

[0032] Fig. 11 shows an observation result of the sectional structure of the composite powder when the Mg based composite powder contains 16% by volume of Mg_2Si particles. The Mg_2Si particles having a particle diameter of 15 μ m or less are uniformly dispersed without segregating and aggregating, so that the Mg based composite powder according to the present invention was provided.

EXAMPLE 2

[0033] Based on the method shown in Fig. 7, pure Mg powder (purity is 99.9% and average particle diameter is 350μ m) and Si powder (purity is 99.9% and average particle diameter is 22 μ m) were prepared and both powder were mixed at a predetermined ratio and a mixing process was performed with a ball mill for 30 minutes. The mixed powder was inputted into a mold having a diameter of 60mm ϕand a pressure of 400MPa was applied to it to manufacture a powder compact.

[0034] When the powder compact is heat treated at 590°C for 1 hour in vacuum, Mg_2Si particles are synthesized by the solid-phase reaction between Si and Mg, and at the same time, a Mg_2Si dispersed Mg sintered material was provided by promoting the sintering in Mg powder. Then, this sintered material was ground by a ball mill to manufacture Mg based composite powder having a diameter of 0.3 to 1mm.

[0035] Fig. 12 shows a result of X-ray diffraction of the Mg based composite powder when the Mg based composite powder contains 7% by volume of Mg_2Si particles. Since only peaks of Mg and Mg_2Si are shown and there is no peak of Si used as the starting material, it is found that Si has completely reacted with Mg and consumed to synthesize Mg_2Si. In addition, since there is no peak of MgO, oxidation is not generated in the sintering process.

[0036] In addition, as a result of a structure observation by an optical microscope, it has been found that the average grain diameter of Mg_2Si dispersed in the powder material is about 24μ m and since it is the same as the particle diameter of Si powder of the starting raw material, no conspicuous coarse grain is not generated in the process of reaction with Mg. As a result, the Mg based composite powder in which the Mg_2Si particles are uniformly dispersed defined by the present invention was provided.

EXAMPLE 3

[0037] Pure Mg powder (purity is 99.9% and average particle diameter is 350 μ m) and Si powder (purity is 99.9% and average particle diameter is 22μ m) were prepared and both powder were mixed at the ratio such that Mg : Si = 2 : 1 (molar ratio) and a mixing process was performed with a ball mill for 30 minutes. The mixed powder was inputted into a carbon mold (inner diameter is 35mm ϕ) and set in a spark plasma sintering machine and sintered for 30 minutes at a sample temperature of 600°C under a pressure of 100MPa in vacuum. As a result, a disk-shaped sample comprising Mg_2Si and having an outer diameter of 35mm ϕ and a thickness of 18mm was provided.

[0038] The disk-shaped sample was ground by a jet mill machine and finely ground and sieved to become a Mg_2Si powder having a maximum particle diameter of 10 μ m or less, so that the Mg_2Si powder as a starting raw material was manufactured.

[0039] Based on the method shown in Fig. 8, AZ61 (nominal composition is Mg-6Al-1Zn/mass%) molten alloy was prepared in a carbon crucible. The above Mg_2Si powder is added at a predetermine ratio, keeping the molten temperature at 720 to 740°C and sufficiently stirred. Then, it is cast in a mold to manufacture a AZ61 casting alloy base material in which the Mg_2Si particles are dispersed.

[0040] Thus, Mg based composite powder comprising AZ61 alloy and having a diameter of 0.5 to 3mm was manufactured from the above casting alloy by a cutting process. Table 1 shows the Mg_2Si content (by volume) in the whole casting alloy material. In addition, the observation result of the sectional structure of the provided composite powder and damaged state of a super-hard tool after the cutting process to manufacture the powder are also shown in Table 1.

[0041] According to samples No.1 to 5 that are examples of the present invention, since appropriate amount of Mg_2Si is contained, the Mg_2Si particles are uniformly dispersed in the matrix of the Mg based composite powder without segregating and aggregating. In addition, regarding tool abrasion (damaged state) generated when the Mg based composite powder is manufactured by the cutting process, although it is a little scraped, there is no problem.

[0042] Meanwhile, according to a sample No.6 that is a comparative example, since the Mg_2Si content is as high as 65%, the Mg_2Si particles are aggregated in the powder matrix and since the casting alloy contains a lot of hard Mg_2Si, a tool is deeply damaged when the powder is manufactured by the cutting process and at the same time Mg is adhered at that damaged part.

[Table 1]

Sample No.	Mg_2Si content (by volume)	Dispersion state of Mg_2Si particles	Damaged state of a tool
1	7	Uniformly dispersed in the matrix	Good surface
2	12	Uniformly dispersed in the matrix	Good surface

(continued)

Sample No.	Mg_2Si content (by volume)	Dispersion state of Mg_2Si particles	Damaged state of a tool
3	28	Uniformly dispersed in the matrix	Good surface
4	38	Uniformly dispersed in the matrix	Very little scraped
5	57	Uniformly dispersed in the matrix	Little scraped
6	65	Aggregation of Mg_2Si particles	Severe damage and adherence of Mg

EXAMPLE 4

[0043] Pure Mg powder (purity is 99.9% and average particle diameter is 350μ m) and Si powder (purity is 99.9% and average particle diameter is 22 μm) were prepared and both powder were mixed at the ratio such that Mg : Si = 2 : 1 (molar ratio) and a mixing process was performed with a ball mill for 30 minutes.

[0044] The mixed powder was inputted into a carbon mold (inner diameter is 35mm ϕ) and set in a spark plasma sintering machine and sintered for 15 minutes at a sample temperature of 600°C under a pressure of 100MPa in vacuum. As a result, a disk-shaped sample comprising Mg_2Si and having an outer diameter of 35mm ϕ and a thickness of 12mm was provided.

[0045] The disk-shaped sample was ground by a jet mill machine. At that time, Mg_2Si powder having different diameters were manufactured by changing the grinding condition.

[0046] Based on the method shown in Fig. 9, as Mg based powder, AM60 (nominal composition is Mg-6A1-0.5Mn/mass%) alloy powder having a diameter of 3mm was prepared as a starting material, and this and Mg_2Si powder was mixed at a predetermined ratio and inputted to a mold having a diameter of 60mm ϕ and a pressure of 400MPa was applied to manufacture a powder compact.

[0047] Then, the powder compact is inputted in AM60 molten alloy in a carbon crucible (molten alloy temperature is 720 to 740°C) and sufficiently stirred and cast in a mold to manufacture a AM60 casting alloy base material in which Mg_2Si particles are dispersed. Then, Mg based composite powder (diameter is 0.5 to 3mm) in which the matrix was AM60 alloy was manufactured from the casting alloy by a cutting process. In addition, the provided Mg based casting alloy contains 22% by volume of Mg_2Si.

[0048] In order to calculate the maximum particle diameter of the Mg_2Si particle dispersed in the Mg based composite powder matrix, the sectional structure of the composite powder was observed by an optical microscope and the maximum particle diameter of the Mg_2Si particle was found from the result by image analysis. The result is shown in Table 2. In addition, a damaged state of a super-hard tool generated in the cutting process when the Mg based composite powder is manufactured from the casting alloy is shown in Table 2.

[0049] According to samples No. 7 to 10 that are examples of the present invention, since the casting alloy contains the Mg_2Si particles having appropriate particle diameter, tool abrasion or damage is not generated when the powder is manufactured by the cutting process and a preferable surface property is provided.

[0050] Meanwhile, according to samples No. 11 and 12 that are comparative examples, since the maximum particle diameter of the Mg_2Si particle contained in the casting alloy is as large as 50 μ m or more, a deep damage is generated in a tool at the time of cutting process and Mg is adhered to that damaged part.

[Table 2]

Sample No.	maximum particle diameter of the Mg_2Si (μm)	Damaged state of a tool
7	3	Good surface
8	12	Good surface
9	26	Good surface
10	42	Good surface
11	75	Severe damage and adherence of Mg
12	92	Severe damage and adherence of Mg

EXAMPLE 5

[0051] The Mg based composite powder described in the above EXAMPLES 3 and 4 were prepared as starting materials and a powder compact was manufactured from each powder by molding. Each powder compact was heated and maintained at 350 °C for 5 minutes in a nitrogen gas atmosphere and immediately an extrusion process was performed to it (extrusion ratio 37) to manufacture an extruded base material. A tensile test piece was made from each extruded base material and its tensile characteristics (tensile strength and toughness) were evaluated at room temperature and its Young's modulus was measured. That result is shown in Table 3.

[0052] According to samples No. 1 to 5 and 7 to 10 that are examples of the present invention, there are provided magnesium based alloy having excellent strength and toughness and according to the samples No. 4 and 5 especially, they have rigidity equal to that of an aluminum alloy. In addition, as shown in the samples No. 7 and 8, when the maximum particle diameter of the Mg_2Si particle dispersed in the alloy is as fine as 5μ m or 20 μm or less, extension is considerably increased in addition to strength.

[0053] Meanwhile, according to a sample No. 6 that is a comparative example, since the Mg_2Si content is high, the Mg based alloy is fragile and it is difficult to manufacture a tensile test piece by machining. In addition, according to samples No. 11 and 12 that are comparative examples, since the maximum particle diameter of Mg_2Si particle is as large as 50μ m or more, the toughness of the Mg based alloy is lowered and its tensile strength is also lowered.

[Table 3]

Sample No.	Tensile strength (MPa)	Toughness (%)	Youngs modulus (GPa)	Remarks
1	278	18	49	
2	293	16	53	
3	314	13	63	
4	322	9	70	
5	326	5	82	
6	-	-	109	Fragile and impossible to make a test piece
7	310	19	59	
8	307	17	58	
9	303	15	59	
10	294	12	58	
11	221	7	57	
12	206	4	57	

EXAMPLE 6

[0054] Pure Mg powder (purity is 99.9% and average particle diameter is 350 μ m) and Si powder (purity is 99.9% and average particle diameter is 22μ m) were prepared and both powder were mixed at the ratio such that Mg : Si = 2 : 1 (molar ratio) and a mixing process was performed with a ball mill for 30 minutes. The mixed powder was inputted into a carbon mold (inner diameter is 35mm ϕ) and set in a spark plasma sintering machine and sintered for 30 minutes at a sample temperature of 600°C under a pressure of 100MPa in vacuum. As a result, a disk-shaped sample comprising Mg_2Si and having an outer diameter of 35mm ϕ) and a thickness of 18mm was provided.

[0055] The disk-shaped sample was ground by a jet mill machine, and Mg_2Si powder having a maximum particle diameter of 10μ m or less and serving as a starting raw material was manufactured through grinding and sieving processes.

[0056] Then, 200 grams of pure Mg powder (purity 99%) having a particle diameter of 0.5 to 2mm was inputted to a vinyl container having capacity of 350ml and 0.6 gram of olein acid was added to the container and vibrated for 15 minutes by a vibration mill to apply the olein acid to the surface of the pure Mg powder in the container uniformly. Then, the Mg_2Si powder was added (13% by volume in the whole mixed powder) into the container and vibrated for 15 minutes to attach the Mg_2Si powder on the surface of the pure Mg powder, whereby Mg based composite powder defined by the present invention was provided.

[0057] Fig. 13 shows the result of X-ray diffraction of the provided Mg based composite powder. Only peaks of Mg and Mg_2Si that were inputted as raw materials are shown and it is found that fine Mg_2Si powder is uniformly attached onto the surface of the coarse pure Mg powder from the result of observation by a scanning electron microscope. As described above, it is confirmed that the Mg based composite powder can be manufactured using the olein acid as a binder.

[0058] Although the embodiments of the present invention have been described with reference to the drawings in the above, the present invention is not limited to the above-illustrated embodiments. Various kinds of modifications and variations may be added to the illustrated embodiments within the same or equal scope of the present invention.

INDUSTRIAL APPLICABILITY

[0059] According to the magnesium based alloy provided by the present invention, low rigidity that is a problem in performance of the conventional magnesium alloy can be considerably improved and the magnesium based alloy can be advantageously applied to a car component such as an engine part or a mission part or a structural member that requires high rigidity.

Claims

1. A magnesium based composite powder comprising magnesium based powder, and magnesium silicide (Mg_2Si) that is dispersed on the surface and/or inside of the matrix of said magnesium based powder.

2. The magnesium based composite powder according to claim 1, wherein the maximum particle diameter of said Mg_2Si is not more than 50μ m.

3. The magnesium based composite powder according to claim 1, wherein the maximum particle diameter of said Mg_2Si is not more than 20μ m.

4. The magnesium based composite powder according to claim 1, wherein the maximum particle diameter of said Mg_2Si is not more than 5 μ m.

5. The magnesium based composite powder according to claim 1, wherein the magnesium based composite powder contains 5 to 60% by volume of said Mg_2Si.

6. A magnesium based alloy base material provided by compacting and sintering the magnesium based composite powder according to claim 1, wherein Mg_2Si particles are dispersed in the matrix.

7. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder;
a step of applying a binder to the surface of said Mg based powder; and
a step of bonding the Mg_2Si powder to the surface of the Mg based powder by mixing and stirring the Mg based powder to which said binder has been applied and said Mg_2Si powder.

8. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder; and
a step of blending said Mg based powder and said Mg_2Si powder and mechanically mixing them to bond the Mg_2Si powder to the surface of the Mg based powder mechanically.

9. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder;
a step of mixing said Mg based powder and said Mg_2Si powder;
a step of manufacturing a Mg based powder compact in which said Mg_2Si particles are dispersed by compacting said mixed powder in a mold;
a step of manufacturing a Mg based sintered alloy in which said Mg_2Si particles are dispersed by sintering said Mg based powder compact; and
a step of powdering said Mg based sintered alloy by mechanical grinding or cutting.

10. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and silicon (Si) powder;
a step of mixing said Mg based powder and said Si powder;
a step of manufacturing a Mg based powder compact in which said Si particles are dispersed by compacting said mixed powder in a mold;
a step of synthesizing Mg_2Si by heating said Mg based powder compact to react Mg with Si, and at the same

time manufacturing a Mg based sintered alloy in which said Mg_2Si particles are dispersed; and
a step of powdering said Mg based sintered alloy by mechanical grinding or cutting.

11. A manufacturing method of magnesium based composite powder comprising:

a step of inputting magnesium silicide (Mg_2Si) powder to molten magnesium based alloy and stirring it;
a step of manufacturing a casting base material by casting said molten material in a mold; and
a step of powdering said casting base material by mechanical grinding or cutting.

12. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and magnesium silicide (Mg_2Si) powder;
a step of mixing said Mg based powder and said Mg_2Si powder;
a step of manufacturing a Mg based powder compact in which said Mg_2Si particles are dispersed by compacting said mixed powder in a mold;
a step of manufacturing a Mg_2Si particle dispersed Mg based casting base material by dissolving and casting said Mg based powder compact; and
a step of powdering said casting base material by mechanical grinding or cutting.

13. A manufacturing method of magnesium based composite powder comprising:

a step of preparing magnesium (Mg) based powder and silicon (Si) powder;
a step of mixing said Mg based powder and said Si powder;
a step of manufacturing a Mg based powder compact in which said Si particles are dispersed by compacting said mixed powder in a mold;
a step of synthesizing Mg_2Si by heating said Mg based powder compact to react Mg with Si, and at the same time manufacturing a Mg based sintered alloy in which said Mg_2Si particles are dispersed;
a step of manufacturing a Mg_2Si particle dispersed Mg based casting base material by dissolving and casting said Mg based sintered alloy; and
a step of powdering said casting base material by mechanical grinding or cutting.

14. A manufacturing method of a magnesium based alloy base material comprising:

a step of compacting the magnesium based composite powder according to claim 1;
a step of heating said powder compact to 200 to 400°C in an inert gas atmosphere or a non-oxygenated gas atmosphere; and
a step of extruding said powder compact to a dense state immediately after said heating.

FIG. 1

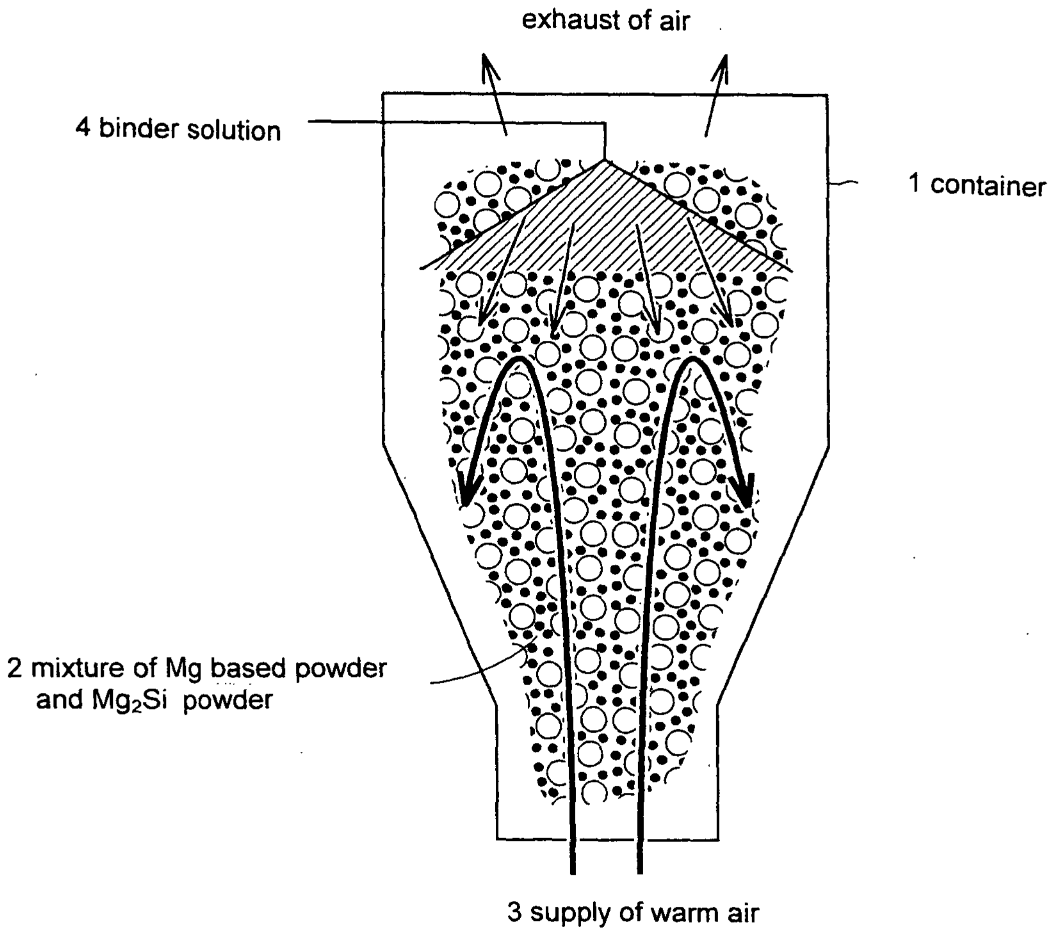

exhaust of air
4 binder solution
1 container
2 mixture of Mg based powder
and Mg_2Si powder
3 supply of warm air

FIG. 2

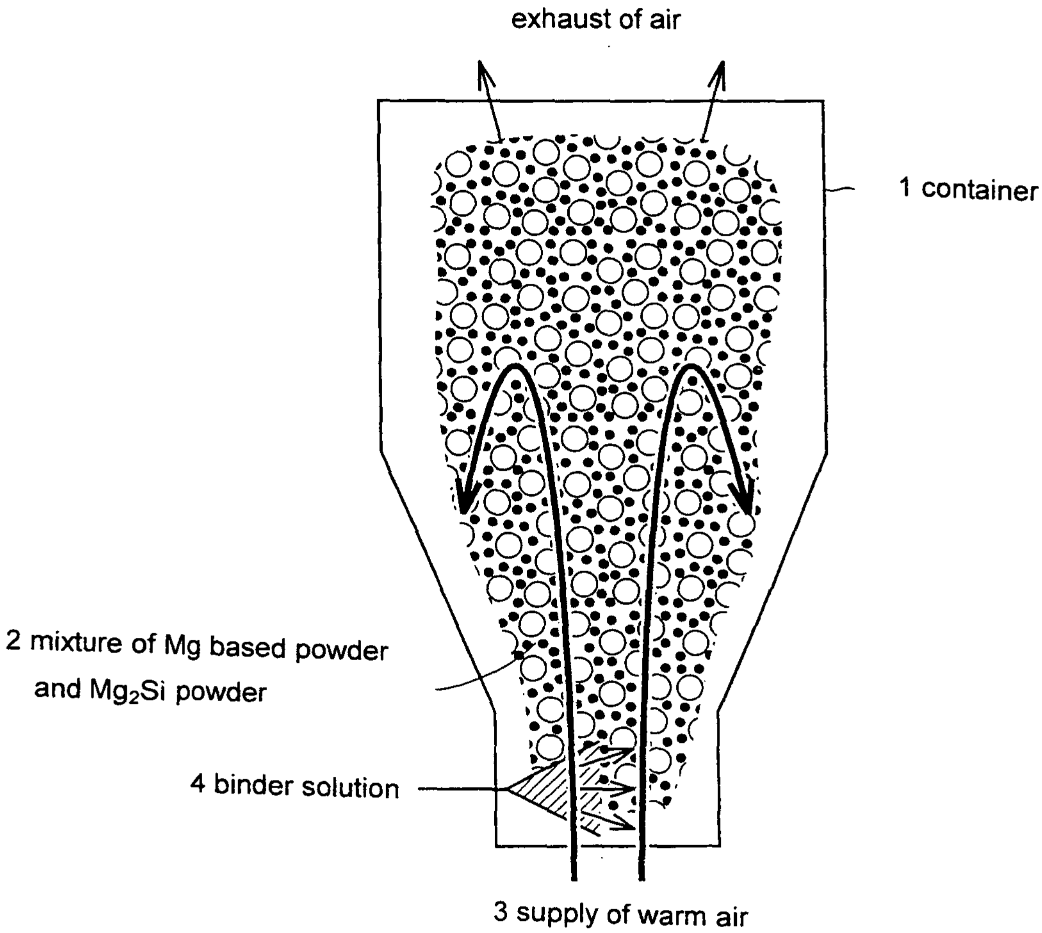

exhaust of air
1 container
2 mixture of Mg based powder and Mg_2Si powder
4 binder solution
3 supply of warm air

FIG. 3

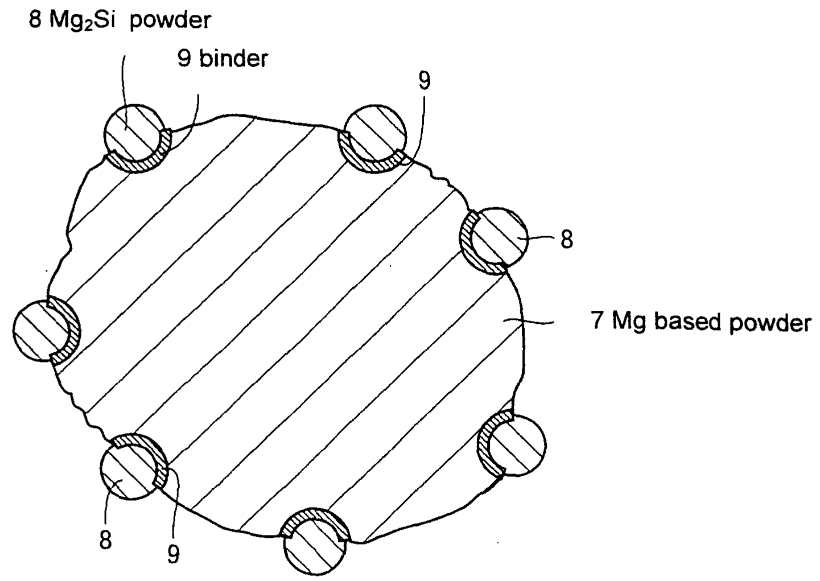

8 Mg2Si powder
9 binder
9
8
7 Mg based powder
8
9

FIG. 4

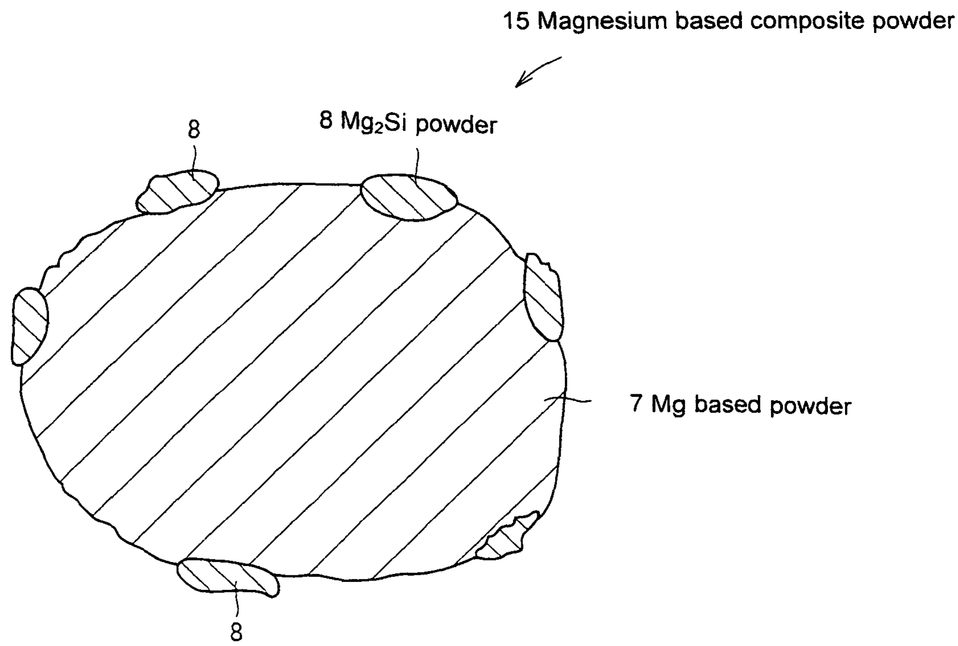

15 Magnesium based composite powder
8
8 Mg2Si powder
7 Mg based powder
8

FIG. 5

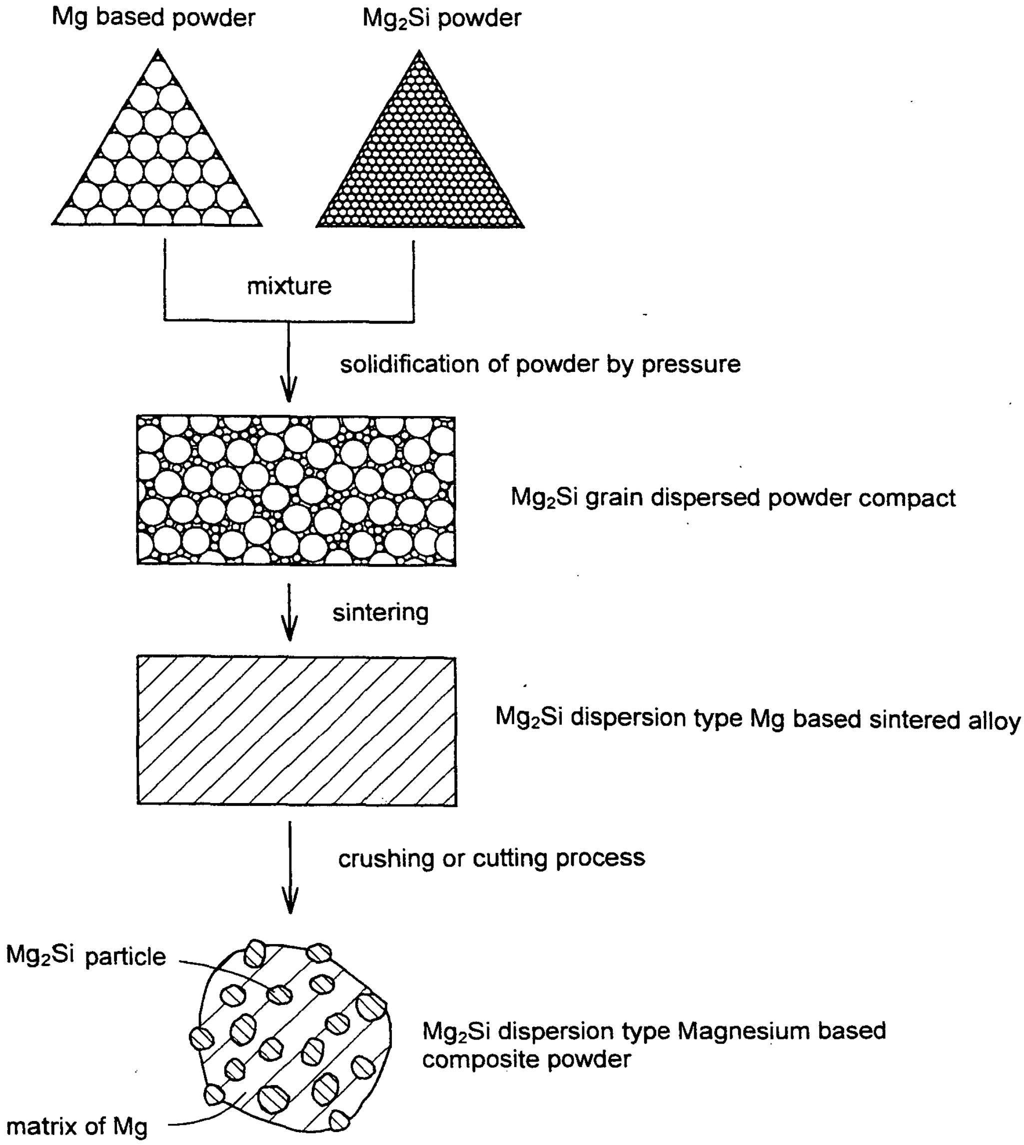

Mg based powder
Mg_2Si powder
mixture
solidification of powder by pressure
Mg_2Si grain dispersed powder compact
sintering
Mg_2Si dispersion type Mg based sintered alloy
crushing or cutting process
Mg_2Si particle
matrix of Mg
Mg_2Si dispersion type Magnesium based composite powder

FIG. 6

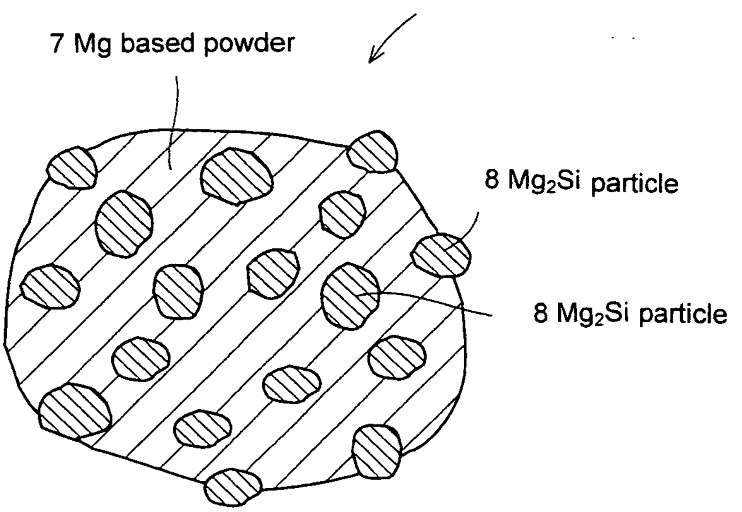

16 Magnesium based composite powder
7 Mg based powder
8 Mg_2Si particle
8 Mg_2Si particle

FIG. 7

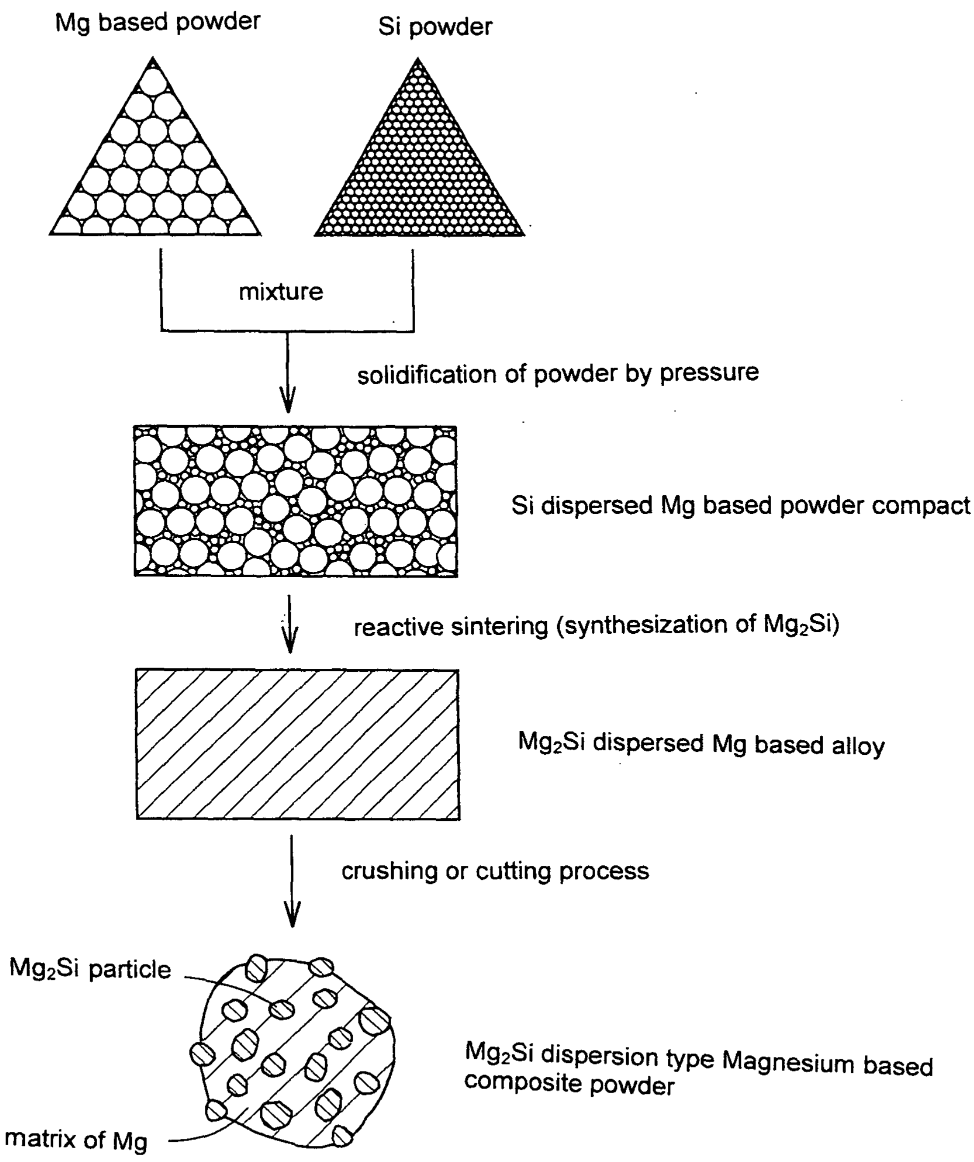

Mg based powder
Si powder
mixture
solidification of powder by pressure
Si dispersed Mg based powder compact
reactive sintering (synthesization of Mg_2Si)
Mg_2Si dispersed Mg based alloy
crushing or cutting process
Mg_2Si particle
matrix of Mg
Mg_2Si dispersion type Magnesium based composite powder

FIG. 8

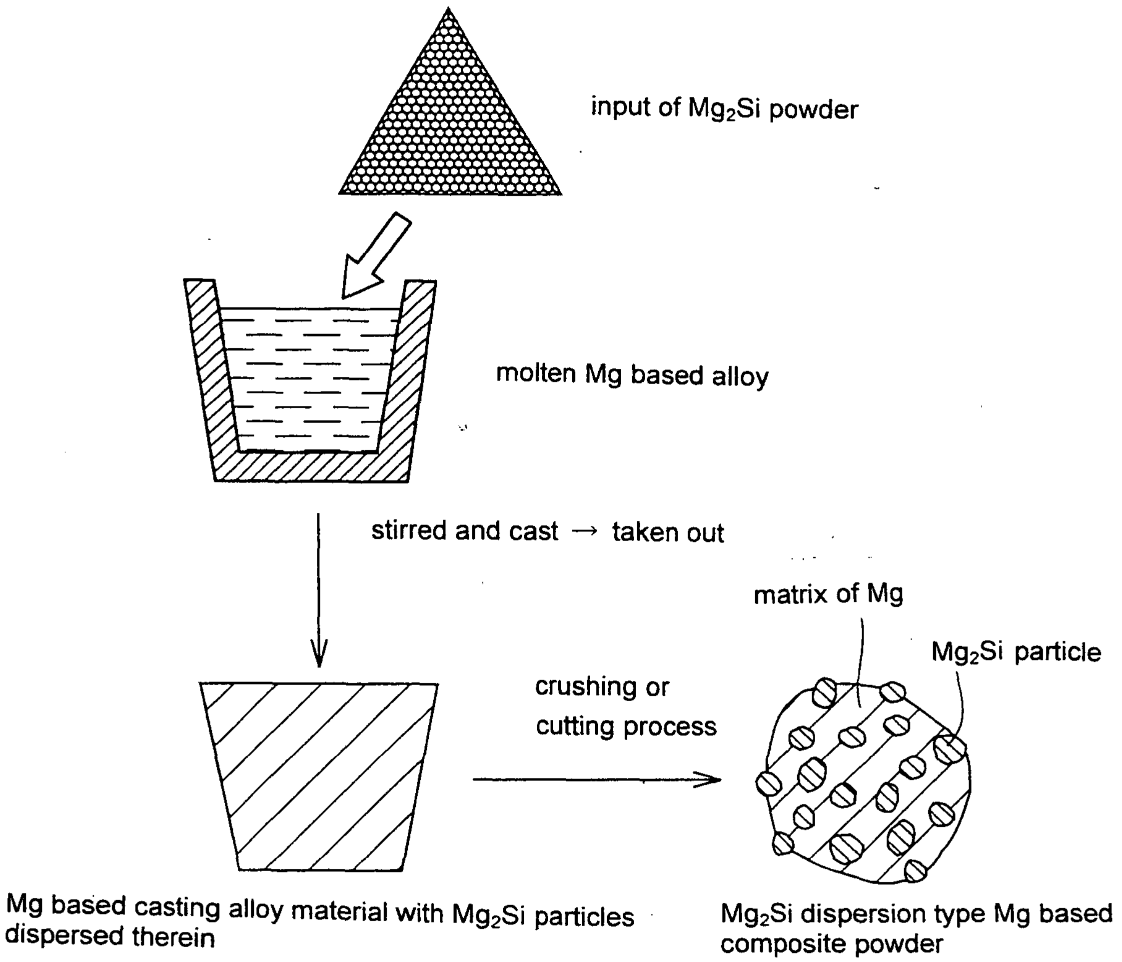

input of Mg_2Si powder
molten Mg based alloy
stirred and cast → taken out
matrix of Mg
Mg_2Si particle
crushing or
cutting process
Mg based casting alloy material with Mg_2Si particles dispersed therein
Mg_2Si dispersion type Mg based composite powder

FIG. 9

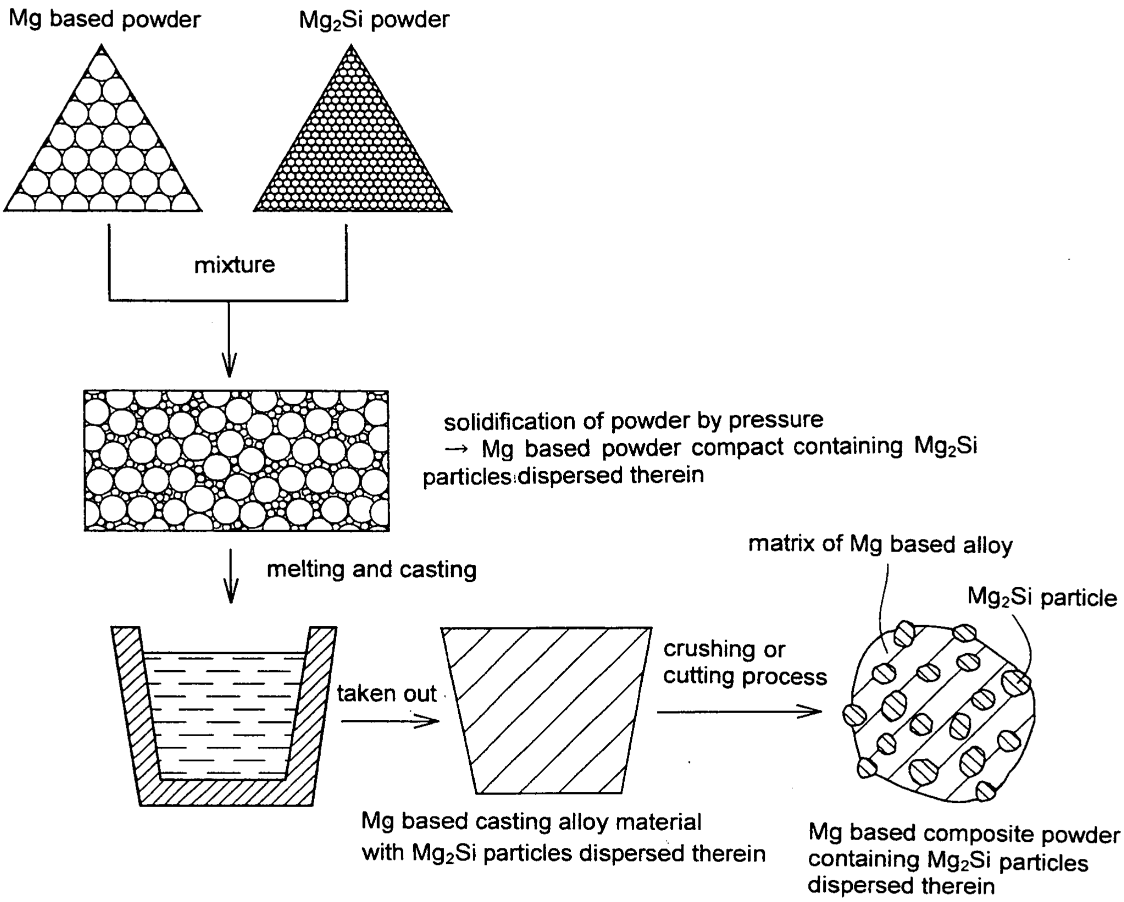

Mg based powder
Mg_2Si powder
mixture
solidification of powder by pressure
→ Mg based powder compact containing Mg_2Si particles dispersed therein
melting and casting
matrix of Mg based alloy
Mg_2Si particle
taken out
crushing or cutting process
Mg based casting alloy material with Mg_2Si particles dispersed therein
Mg based composite powder containing Mg_2Si particles dispersed therein

FIG. 10

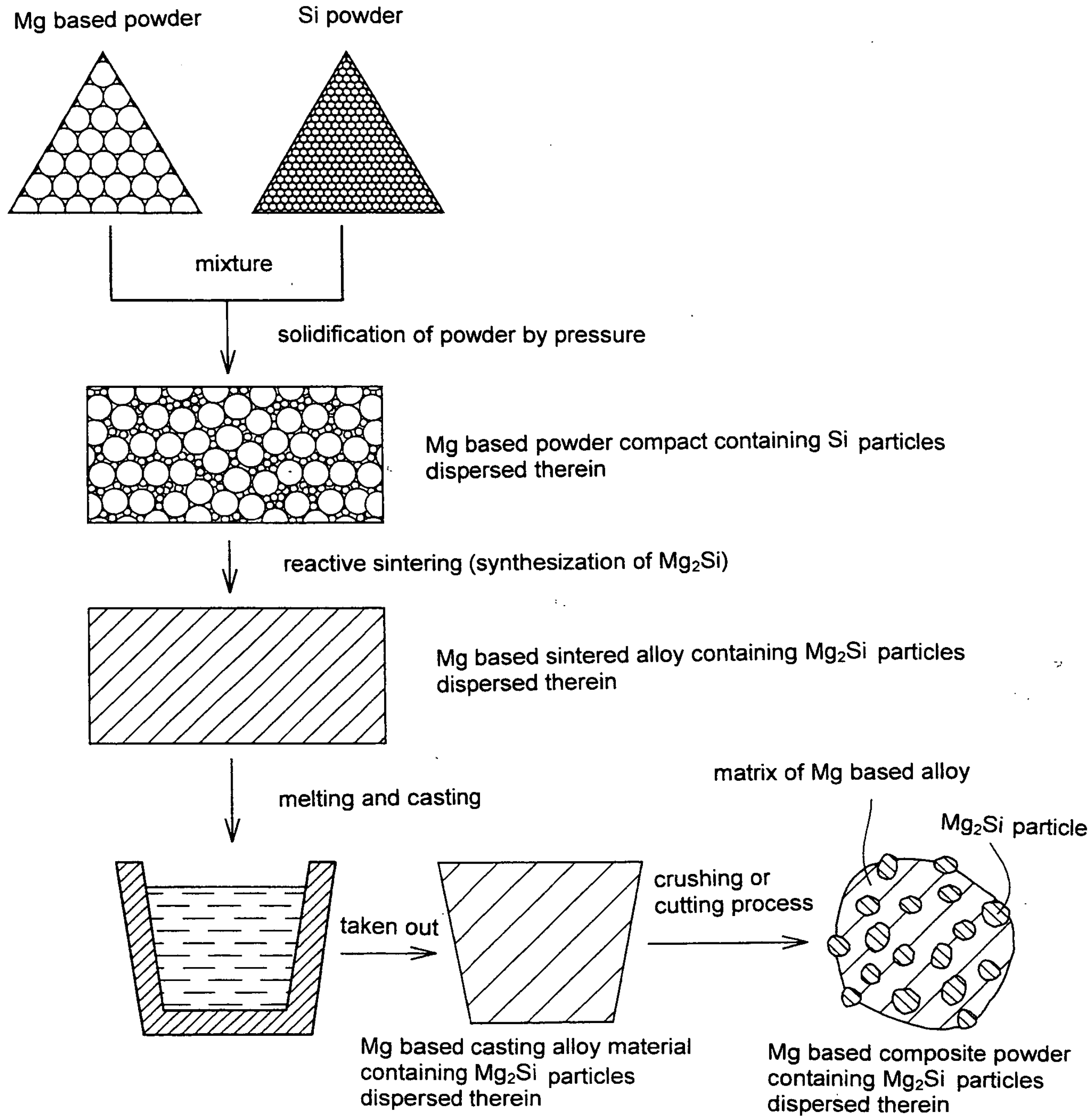

Mg based powder
Si powder
mixture
solidification of powder by pressure
Mg based powder compact containing Si particles dispersed therein
reactive sintering (synthesization of Mg_2Si)
Mg based sintered alloy containing Mg_2Si particles dispersed therein
melting and casting
matrix of Mg based alloy
Mg_2Si particle
taken out
crushing or cutting process
Mg based casting alloy material containing Mg_2Si particles dispersed therein
Mg based composite powder containing Mg_2Si particles dispersed therein

FIG. 11

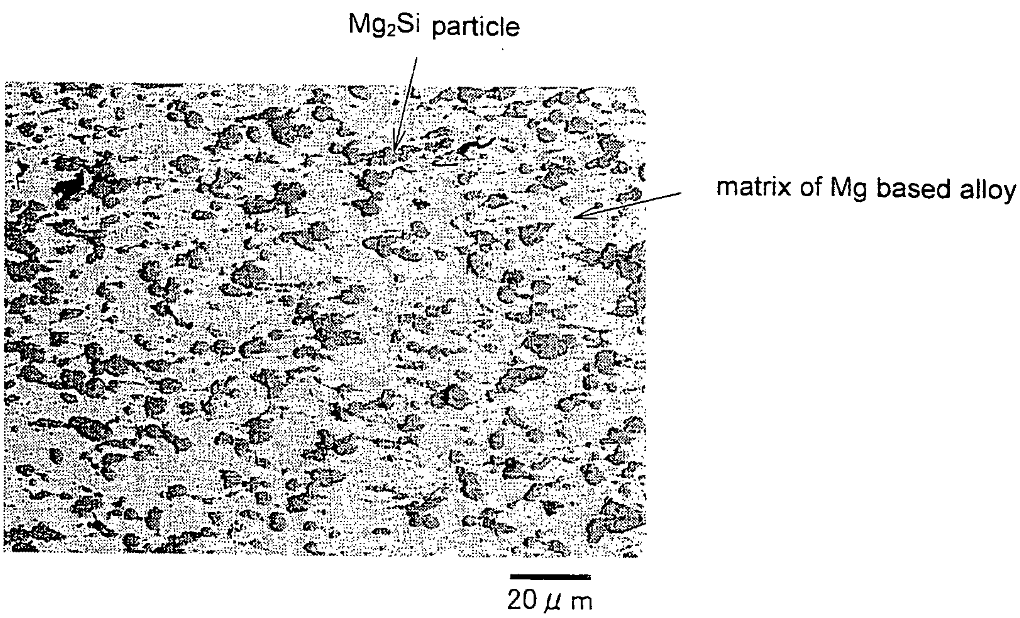


FIG. 12

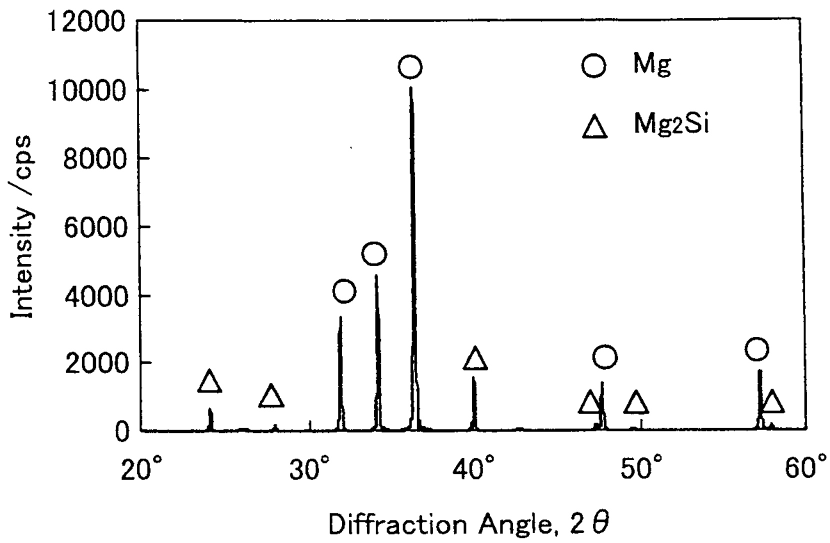
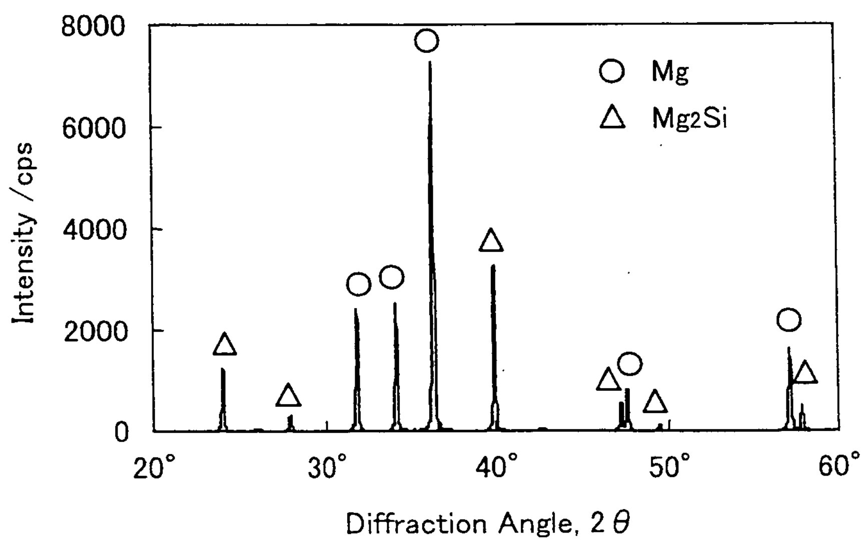

12000
10000
8000
6000
4000
2000
0
Intensity /cps
Mg
Mg2Si
20°
30°
40°
50°
60°
Diffraction Angle, 2θ

FIG. 13


8000
6000
4000
2000
0
Intensity /cps
Mg
Mg2Si
20°
30°
40°
50°
60°
Diffraction Angle, 2θ

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2005/011744

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl[7] B22F1/00, 1/02, 3/20, 3/24, 9/04, C22C1/04, 1/05, 1/10, 23/00

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] B22F1/00, 1/02, 3/20, 3/24, 9/04, C22C1/04, 1/05, 1/10, 23/00

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Jitsuyo Shinan Toroku Koho	1996-2005
Kokai Jitsuyo Shinan Koho	1971-2005	Toroku Jitsuyo Shinan Koho	1994-2005

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.
X	JP 2002-249801 A (National Institute of Advanced Industrial Science and Technology (AIST)), 06 September, 2002 (06.09.02), Claims; Par. No. [0014] & EP 1433862 A1	1
A	WO 2003-027342 A (TOUDAI TLO, Ltd.), 03 April, 2003 (03.04.03), Claims (Family: none)	1
A	JP 06-316751 A (Mitsui Mining & Smelting Co., Ltd.), 15 November, 1994 (15.11.94), Claims (Family: none)	1

☒ 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 application or patent 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	Date of mailing of the international search report
09 September, 2005 (09.09.05)	27 September, 2005 (27.09.05)
Name and mailing address of the ISA/ Japanese Patent Office	Authorized officer
Facsimile No.	Telephone No.

Form PCT/ISA/210 (second sheet) (January 2004)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2005/011744

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 04-198408 A (Sumitomo Metal Industries, Ltd.), 17 July, 1992 (17.07.92), Claims (Family: none)	1
A	JP 08-092603 A (Suzuki Motor Corp.), 09 April, 1996 (09.04.96), Claims (Family: none)	1

Form PCT/ISA/210 (continuation of second sheet) (January 2004)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2005/011744

Box No. II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This international search report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:

2. ☐ Claims Nos.:
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:

3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box No. III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:

The matter common to claims 1-6, 9-14 is a magnesium-base composite powder comprising a magnesium-base powder and magnesium silicide (Mg_2Si) dispersed within the base material of the magnesium-base powder.

The international search, however, has revealed that this magnesium-base composite powder is not novel since it is disclosed in JP 2002-249801 A (National Institute of Advanced Industrial Science and Technology), 6 September, 2002 (06.09.02).

Consequently, the common matter is not a special technical feature within the meaning of PCT Rule 13.2, second sentence, since the above-mentioned powder makes no contribution over the prior art. (Continued to extra sheet)

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.

2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.

3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:

4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.: 1

Remark on Protest

☐ The additional search fees were accompanied by the applicant's protest.

☐ No protest accompanied the payment of additional search fees.

Form PCT/ISA/210 (continuation of first sheet (2)) (January 2004)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2005/011744

Continuation of Box No.III of continuation of first sheet(2)

Since there is no common matter which can be considered as a special technical feature within the meaning of PCT Rule 13.2, second sentence, no technical relationship within the meaning of PCT Rule 13 can be seen between those different inventions.

Therefore, it is obvious that claims 1-6, 9-14 do not satisfy the requirement of unity of invention.

Form PCT/ISA/210 (extra sheet) (January 2004)

REFERENCES CITED IN THE DESCRIPTION

This list of references cited by the applicant is for the reader's convenience only. It does not form part of the European patent document. Even though great care has been taken in compiling the references, errors or omissions cannot be excluded and the EPO disclaims all liability in this regard.

Patent documents cited in the description

- JP 2003002602 A **[0007] [0008] [0010]**

Non-patent literature cited in the description

- **S. K. THAKUR.** *Metallurgical and Materials Transactions,* March 2004, vol. 35A, 1167-1176 **[0006]**