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⑤④ **Anisotropic plastic bonded magnet of the Nd-Fe-B-type and method for making same.**

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⑦③ Proprietor : **MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.**
1006, Oaza Kadoma
Kadoma-shi, Osaka-fu, 571 (JP)

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⑦② Inventor : **Furuyama, Shizuo**
4-11-18 Hoshida Yamate
Katano-shi, Osaka 576 (JP)
Inventor : **Kojima, Kiyoshi**
4-5-17 Mayumi Ikoma-shi
Nara Prefecture 630-01 (JP)

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⑦④ Representative : **Wain, Christopher Paul et al**
A.A. THORNTON & CO.
Northumberland House
303-306 High Holborn
London WC1V 7LE (GB)

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Description

This invention relates to a plastic bonded magnet containing an anisotropic neodymium-iron-boron system magnetic powder. Such magnets are used in various devices including stepping motors, spindle motors, torque motors, automotive motors, various actuators, speakers, and other magnetic-field generating devices.

Extensive efforts have been made to improve the maximum energy product (hereinafter abbreviated to $(BH)_{\max}$) of resin-bonded rare-earth magnets. There are three essential requirements for a higher $(BH)_{\max}$ plastic bonded magnet, namely (1) higher magnetization $4\pi I$ and higher coercive force iH_c , (2) higher mold density, and (3) higher magnetic orientation of the magnetic powder. Various ways have been found to attain requirements (1) and (2), including those methods disclosed by in Japanese Laid-Open Patents Sho 60-207302 and Sho 60-220907. Patent Abstracts of Japan, Vol.13, Number 235 (E-766), <3583>, 30 May 1989 discloses a bonded magnet having large coercive force and energy product formed from rare earth-iron-boron particles. EP-A-155082 discloses the use of epoxy resin compositions to make bonded rare earth-iron alloy magnets. The magnets exhibit high bond strengths and are resistant to flux loss upon aging. However, virtually no satisfactory way has to date been found to improve the magnetic powder orientation.

We have now devised a new neodymium-iron-boron system plastic bonded magnet having an improved $(BH)_{\max}$ achieved by attaining a higher orientation of the magnetic powder thereof. In particular the invention provides a plastic bonded magnet which comprises an anisotropic neodymium-iron-boron system magnetic powder and a resin binder characterised in that the magnetic powder has a rounded shape, from 10 to 20% by weight of the magnetic powder has a grain size in the range 10 to 49 μm , and from 80 to 90% by weight of the magnetic powder has a grain size in the range 50 to 500 μm .

The magnetic powder grain distribution used in the magnets of the invention can be obtained by kneading and dispersing the powder (together with a binder) in a high-speed shearing machine.

By using the above-described magnetic powder constitution, a higher $(BH)_{\max}$ is obtained due to a substantially improved magnetic orientation in the compressed molded body of the plastic bonded magnet. Furthermore, an extended pot-life of molding compound and higher $(BH)_{\max}$ value can be obtained by employing an epoxy resin as the binder and curing it with a latent hardening agent which is an amine adduct of the epoxy resin.

In order that the invention may be more fully understood, reference will be made to the accompanying drawings, wherein:

Figure 1 is an enlarged cross-section of one embodiment of plastic bonded magnet of the invention;

Figure 2 shows the relationship between the grain-size of magnetic powder of the invention and the magnetic characteristics;

Figure 3 is a schematic representation of magnetic powder employed in Comparative Example 1;

Figure 4 is a schematic representation showing the magnetic powder grain configuration after kneading and dispersion of the magnetic powder shown in Figure 1; and

Figure 5 is a schematic diagram of the magnetic powder grain configuration after kneading and dispersion of magnetic powder used in Example 3.

Referring now to Figure 1, there is shown a magnet of the invention wherein 1 is anisotropic neodymium-iron-boron system magnetic powder having a grain size distribution from 50 to 500 μm (microns), reference 2 is anisotropic neodymium-iron-boron system magnetic powder having grain sizes from 10 to 49 μm (microns), and 3 is the plastic resin binder. The anisotropic neodymium-iron-boron system magnetic powder employed in this invention can be anisotropic neodymium-iron-boron system magnetic powder manufactured by upsetting its overquenched melt-spun ribbon.

The magnets may also comprise, in addition, elements such as zirconium, gallium, cobalt, praseodymium, and tin for improving temperature characteristics, anti-corrosion properties, and magnetic properties as will be understood by those skilled in the art.

The magnetic powder employed in the invention has excellent magnetic properties, (magnetization $4\pi I$, coercive force iH_c) which are substantially governed by the grain size distribution as shown in Figure 2. Thus, the magnetic characteristics of the fine powder having grain sizes of 10 to 49 μm (microns) are significantly different from those of powder of greater grain size, as Fig. 2 illustrates.

It will be understood that the invention does not use exclusively powder having grain sizes less than 49 microns: such magnets would have inferior magnetic properties. Rather, the invention introduces a limited amount only of such powder into the molding composition to improve the magnetic orientation. The magnetic orientation of the plastic bonded magnet is highly improved by introducing magnetic powder having fine grain sizes from 10 to 49 μm (microns) in an amount of 10 to 20% by weight. The reason for this cannot be attributed to higher magnetic powder density, but we believe it can be attributed to the improved flowability of the molding composition.

We prefer to exclude magnetic powder having grain sizes less than 10 μm (microns) because of its poor magnetic characteristics. The content of fine magnetic powder (10 to 49 μm (microns) should be limited to less than 20% by weight, but the orientation effect cannot be attained if its content is less than 10% by weight. Although large sized magnetic powder having grain sizes more than 500 μm (microns) has excellent magnetic characteristics, it causes a reduction in mold density and as a result no improvement in residual magnetic flux density B_r can be expected.

The molding composition will contain a suitable dispersing agent. Fatty acids, silicone-coupling agents and various surface-active agents can be employed, we prefer to use a liquid fatty acid, eg. oleic acid. This ability to function as a dispersing agent is attributed to the higher affinity for the magnetic powder. This is not shown by any of the solid fatty acids. The use of liquid fatty acids is particularly advantageous in compressive molding which is performed conventionally at room temperature since it improves productivity.

Although it is important to keep the amount of fatty acid additive as low as possible, it is preferred to introduce at least 0.8% by weight thereof (based on the weight of the magnetic powder). An amount of fatty acid less than 0.7% by weight usually results in an inadequate affinity to the magnetic powder resulting in reduced density and $(BH)_{\text{max}}$. On the other hand, if an excessive amount of fatty acid is used, eg. more than 1.3% by weight, oozing of binder tends to occur and lower strength of the molded magnet results.

Whilst various binders can be used in the present invention, for room temperature molding we prefer to use liquid epoxy resin to give good adhesion strength. The total amount of binder is determined by the attainable magnetic characteristics and the strength of the molding. The total amount of binder component used depends on the magnetic characteristics required and the strength of the molded magnet. If an amount of binder of less than 1.4% by weight is mixed with the magnetic powder, this would usually result in inadequate mold strength, but if more than 3.0% by weight is used, this would result in poor magnetic characteristics.

Any suitable curing or hardening agent can be used. Amines and acid anhydrides are generally preferred. If an amine adduct of epoxy resin is used as a hardening agent for epoxy resin used as a binder, a substantially extended pot-life of the molding composition can be obtained relative to the use of a conventional amine system hardening agent.

The molding composition is preferably prepared in a high-speed shearing machine capable of pulverizing and mixing the magnetic powder, such as a Henschell mixer, upper-mill, high-speed mixer, or a micro-mill grinder.

The following Examples illustrate the invention. The comparisons are not according to the invention.

Example 1

A plastic bonded magnetic material mixture consisting of 100 weight parts of anisotropic Nd-Fe-B system magnetic powder having grain sizes from 1 to 2 mm and 0.9 weight part of oleic acid as a dispersion agent, are ground in a micro-mill grinder for about five minutes, and 1.0 weight part of epoxy resin, eg. Epikote 828 (manufactured by Yuka-Shell Epoxy Co.), is added thereto, and ground for another ten minutes.

The dispersion process is completed by adding 0.9 weight parts of acid anhydride, eg. Kayahard MCD (manufactured by Nihon Kayaku Co.), and 0.01 weight parts of catalytic imidazone, eg. Epikure EMI-24 (manufactured by Shikoku Kasei Kogyo Co.) thereto, and the mixing continued for another ten minutes.

The molding composition so made is loaded into a die cavity, and is molded at room temperature using a pressure of 6 ton/cm² (6×10^8 Pa) under application of a magnetic field of 15 KOe. After a hardening process conducted at 100°C for one hour, a plastic bonded magnet of the invention is produced.

For purposes of analysis, a molded article before hardening is removed from a mold and immersed in acetone to remove resin components. The magnetic powder obtained is dried in a nitrogen atmosphere. The grain size distribution of this magnetic powder was measured and found to be:

<u>Grain size</u>	<u>Composition</u>
500 - 800 μm	5% by weight
50 - 500 μm	80% by weight
10 - 49 μm	14% by weight
less than 10 μm	1% by weight

Comparison 1

For comparison purpose, the procedure of Example 1 is repeated except that magnetic powder having grain sizes from 300 to 500 μm is used as starting material. The grain size distribution in this case was measured and was found to be:

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<u>Grain size</u>	<u>Composition</u>
50 - 500 μm	65% by weight
10 - 49 μm	30% by weight
less than 10 μm	5% by weight

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Comparison 2

Example 1 is repeated except that magnetic powder having grain sizes from 3 to 4 mm is used. The grain size distribution was found to be:

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<u>Grain size</u>	<u>Composition</u>
1 - 2 mm	5% by weight
0.5 - 1 mm	16% by weight
50 - 500 μm	68% by weight
10 - 49 μm	10% by weight
less than 10 μm	1% by weight

Comparison 3

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Example 1 is repeated except that this time the mixture is ground for 12 hours in a ball-mill using acetone as a solvent and then the acetone is evacuated. The grain size distribution was found to be:

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<u>Grain size</u>	<u>Composition</u>
50 - 500 μm	10% by weight
10 - 49 μm	78% by weight
less than 10 μm	12% by weight

Example 2

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The general procedure of Example 1 is followed except that an electromagnetic steel plate made of soft magnetic material is inserted in the die before the molding composition is injected. The molding is then conducted at room temperature under a pressure of $6.6 \times 10^8 \text{ Pa}$ (ton/cm²) and a magnetic field of 15 KOe. After a hardening process at 100°C for two hours, a plastic bonded magnet having an integrated electromagnetic steel plate made of soft magnetic material, is obtained. The adhesion between the soft steel plate and plastic bonded magnet was found adequately high.

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The determined densities, magnetic characteristics and magnetic orientations of the plastic magnets obtained in Example 1 and in Comparisons 1, 2 and 3 are set out in Table 1.

The degree of magnetic orientation is defined as $B_r(//) / (B_r(//) + B_r(\perp))$. This represents the ease of move-

ment of magnetic powder under application of an external magnetic field, and a higher value means a higher magnetic orientation. $B_r(//)$ and $B_r(\perp)$ represent a residual magnetic flux density along the applied magnetic field and a residual magnetic flux density perpendicular to the applied magnetic field, respectively.

Table 1

Sample	Density (g/cc)	Degree of magnetic orientation	$(BH)_{\max}$ MG Oe
Example 1	6.20	0.70	15
Comparison 1	6.19	0.67	13
Comparison 2	6.15	0.68	12
Comparison 3	6.18	0.66	11

Table 1 shows that plastic bonded magnets having higher densities, degrees of magnetic orientation and $(BH)_{\max}$ values can be obtained by the present invention.

Changes in grain configurations of magnetic powder are shown in Figs. 3, 4 and 5. Figure 3 shows a schematic configuration of raw magnetic powder material, and Figure 4 is a schematic showing the rounded-off grains of powder after pulverization and mixing conducted by a high-speed shearing machine which is employed to prepare the magnet of Example 1. Figure 5 is a schematic diagram showing grain configuration of magnetic powder after kneading and dispersion of the magnetic powder of Comparison 3, showing more squarish and smaller grain sizes compared with those shown in Figure 4.

Thus, as shown in Figure 1, higher magnetic orientation and higher density, and higher $(BH)_{\max}$, can be realized by the grain size changes in accordance with the invention, for example produced by the pulverization and dispersion process using a high-speed shearing machine, as described. Whilst we do not wish to be bound by theory, we believe that the reasons for these improvements can be explained as follows. Whereas the magnet of Example 1 contains 14% by weight magnetic powder having grain sizes from 10 to 49 μm , the amounts in Comparisons 1, 2 and 3 are 30% by weight, 10% by weight, and 78% by weight, respectively. Those improvements of magnetic orientation in Example 1 and Comparison 2 are due to the containment of 10 to 20% by weight magnetic powder having grain sizes from 10 to 49 μm . While the magnetic orientation is 0.70 and 0.68 in Example 1 and Comparison 2, respectively, it is only 0.65 in Comparison 3.

This improvement of magnetic orientation is attributed to the improved flowability of the compressive molding composition containing fine magnetic powder having grain sizes from 10 to 49 μm in amounts from 10 to 20% by weight. Hitherto, the use of this type of magnetic powder had been avoided because of its lower magnetic characteristics.

Furthermore, as shown in Example 1, the improvement of $(BH)_{\max}$ is attributed to the higher magnetic orientation and improved density due to using magnetic powder which is rounded-off during said high-speed shearing process. The density of the magnet of Comparison 2 is 6.15 which is low because it contains magnetic powder having grain sizes from 1 to 2mm, and thus, the $(BH)_{\max}$ is low also.

This means that the improvements of both orientation and density contribute to the improved $(BH)_{\max}$, achieved by the present invention. Further, in the plastic bonded magnet containing an integrated soft magnetic plate, described in Example 2, the bond strength between these parts was found satisfactory even without using any adhesives, and thus, a simplification in the conventional manufacturing process can be realized.

Example 3

An anisotropic Nd-Fe-B system magnetic powder having an average grain size of 1mm and oleic acid are mixed at a weight ratio of 100 to 0.9, and this mixture is kneaded for 10 minutes in a high-speed mixer under a nitrogen atmosphere.

Then, 1.6 weight parts of an epoxy resin, eg. Epikote-828 manufactured by Yuka-Shell Epoxy Co., is added thereto and the mixture is kneaded for another ten minutes. A latent hardening agent, eg. Amicure PN-23, manufactured by Ajinomoto Co., is then added in an amount of 0.4 weight part, and the mixture kneaded for another

10 minutes before the process is complete. The resulting molding composition is then loaded into a cavity for applying a compressive pressure of 6 ton/cm² with an applied magnetic field of 15 KOe. The anisotropic plastic bonded magnet is produced after hardening for one hour at 100°C.

5 Comparison 4

Example 3 is repeated except that the materials are as follows:

10 Anisotropic Nd-Fe-B magnetic powder ... 100 weight parts.
 Oleic acid ... 0.9 weight part.
 Epoxy resin (Epikote-828) ... 1.5 weight part.
 Aliphatic amine (LX-1N, Yuka-Shell
 15 Epoxy Co.) ... 0.5 weight part.

Comparison 5

20 Example 3 is repeated except that the materials used were:

Anisotropic Nd-Fe-B magnetic powder ... 100 weight parts.
 Oleic acid ... 0.9 weight part.
 25 Epoxy resin (Epikote-828) ... 1.25 weight part.
 Aromatic amine (Achmex H-90, Nihon
 Goseikako Co.) ... 0.5 weight part.

30 The initial magnetic characteristics, $(BH)_{\max}$, of the magnets, and the pot-life of the molding compositions are shown in Table 2. The pot-life was determined as follows. The molding composition is left still at room temperature, and samples are taken out every one hour and used to mold a sample plastic bonded magnet under the above-described conditions. The elapsed time at which a sample of the composition forms a magnet which
 35 shows a magnetic characteristic which is lower by 5 % than that of the initial magnet is defined as the pot-life of the composition.

Table 2

40	<u>Sample</u>	Initial mag. Characteristics (BH) _{max}	Pot-life (hour)
45	Example 3	15 MGOe	20
	Comparison 4	12	3
	Comparison 5	15	6

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The plastic bonded magnet shown in Example 3 wherein a latent hardening agent is used, showed a value of $(BH)_{\max}$ as high as 15 MGOe and a pot-life as long as 20 hours, which are very useful features. The use of a high-speed shearing machine minimizes the possibility of stability damage of binder system due to the local heat produced at kneading.

55 On the other hand, the magnet produced by using an aliphatic amine hardening agent, shown in Comparison 4, had a value of $(BH)_{\max}$ of 12 MGOe and a pot life of only 3 hours, and this has little practical value. Although using an aromatic amine hardening agent as shown in Comparison 5, gave a value of $(BH)_{\max}$ of 15 MGOe which is reasonably high, the pot life of 6 hours is still considered inadequate for practical purposes.

As mentioned above, the $(BH)_{\max}$ of the magnet, and the pot-life of the composition are influenced by the type of hardening agent. The difference between magnetic characteristics of the magnets can be attributed in part to the difference between the affinities of binders for the magnetic powder, which results in the difference between dispersibilities.

The difference between pot-lives can be attributed to the difference between thermal stabilities of the binder systems against local heat generation. The latent hardening agent employed, i.e. a hardening agent which does not start hardening until a certain time is elapsed after it is mixed with a primary epoxy agent, is much preferred. This hardening system has a high thermal stability, and is thus considered highly useful to extend the pot-life.

Claims

1. A plastic bonded magnet which comprises an anisotropic neodymium-iron-boron system magnetic powder and a resin binder characterised in that the magnetic powder has a rounded shape, from 10 to 20% by weight of the magnetic powder has a grain size in the range 10 to 49 μm , and from 80 to 90% by weight of the magnetic powder has a grain size in the range 50 to 500 μm .
2. A magnet according to claim 1, wherein the resin binder is an epoxy resin, and the magnet further comprises an amine adduct of epoxy resin as a latent hardening agent for the epoxy resin.
3. A method of producing a plastic bonded magnet as claimed in claim 1, which method comprises the steps of preparing an anisotropic neodymium-iron-boron system magnetic powder and a resin binder, mixing the magnetic powder and the resin binder, molding the resulting mixture in a magnetic field, and conducting a hardening treatment of the molded article, characterised in that from 10 to 20% by weight of the magnetic powder has a grain size in the range 10 to 49 μm and from 80 to 90% by weight of the magnetic powder has a grain size in the range 50 to 500 μm ; and in that the step of mixing comprises a step of simultaneously pulverizing and dispersing the magnetic powder with the resin binder in a high-speed shearing machine.
4. A method according to claim 3, wherein the resin binder is an epoxy resin which is cured using an amine adduct of epoxy resin as a latent hardening agent for the epoxy resin.

Patentansprüche

1. Kunststoffgebundener Magnet, der ein Magnetpulver eines anisotropen Neodym-Eisen-Bor-Systems und einen Kunstharzbinder aufweist, dadurch gekennzeichnet, daß das Magnetpulver eine abgerundete Form besitzt, von 10 bis 20% bezogen auf das Gewicht des Magnetpulvers eine Korngröße in dem Bereich von 10 bis 49 μm besitzt und von 80 bis 90% bezogen auf das Gewicht des Magnetpulvers eine Korngröße in dem Bereich von 50 bis 500 μm besitzt.
2. Magnet nach Anspruch 1, wobei der Kunstharzbinder ein Epoxidkunstharz ist und der Magnet weiterhin ein Aminaddukt eines Epoxidkunstharzes als ein latentes Aushärtemittel für das Epoxidkunstharz aufweist.
3. Verfahren zur Herstellung eines kunststoffgebundenen Magneten nach Anspruch 1, wobei das Verfahren die Verfahrensschritte der Herstellung eines Magnetpulvers eines anisotropen Neodym-Eisen-Bor-Systems und eines Kunstharzbinders, Mischen des Magnetpulvers und des Kunstharzbinders, Gießen der sich ergebenden Mischung in einem Magnetfeld und Durchführung einer Aushärtebehandlung des gegossenen Artikels aufweist, dadurch gekennzeichnet, daß 10 bis 20% bezogen auf das Gewicht des Magnetpulvers eine Korngröße in dem Bereich von 10 bis 49 μm besitzen und 80 bis 90% bezogen auf das Gewicht des Magnetpulvers eine Korngröße in dem Bereich von 50 bis 500 μm besitzen und daß der Verfahrensschritt des Mischens einen Verfahrensschritt einer gleichzeitigen Pulverisierung und Dispergierung des Magnetpulvers mit dem Kunstharzbinder in einer Hochgeschwindigkeits-Schermaschine aufweist.
4. Verfahren nach Anspruch 3, wobei der Kunstharzbinder ein Epoxidkunstharz ist, das ausgehärtet wird, indem ein Amin-Addukt eines Epoxidkunstharzes als latentes Aushärtemittel für das Epoxidkunstharz

verwendet wird.

Revendications

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1. Aimant à liant de matière plastique qui comprend une poudre magnétique anisotrope du système néodyme-ferbore et un liant de résine, caractérisé en ce que la poudre magnétique a une configuration arrondie, 10 à 20 % en poids de la poudre magnétique ayant une dimension granulaire comprise entre 10 et 49 μm et 80 à 90 % en poids de la poudre magnétique ayant une dimension granulaire comprise entre 50 et 500 μm .

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2. Aimant selon la revendication 1, dans lequel le liant de résine est une époxyde, et l'aimant comporte en outre un produit d'addition d'amine de résine époxyde comme agent durcisseur latent pour la résine époxyde.

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3. Procédé de fabrication d'un aimant à liant de matière plastique selon la revendication 1, comprenant les étapes suivantes : la préparation d'une poudre magnétique anisotrope du système néodyme-fer-bore et d'un liant de résine, le mélange de la poudre magnétique et du liant de résine, le moulage du mélange résultant dans un champ magnétique, et l'exécution d'un traitement de durcissement de l'article moulé, caractérisé en ce que 10 à 20 % en poids de la poudre magnétique ont une dimension granulaire comprise entre 10 et 49 μm et 80 à 90 % en poids de la poudre magnétique ont une dimension granulaire comprise entre 50 et 500 μm , et en ce que l'étape de mélange comprend une étape de pulvérisation et de dispersion simultanées de la poudre magnétique avec le liant de résine dans une machine à grande vitesse ayant un gradient élevé de vitesse.

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4. Procédé selon la revendication 3, dans lequel le liant de résine est une résine époxyde qui est polymérisée avec un produit d'addition d'une amine et d'une résine époxyde comme agent durcisseur latent de la résine époxyde.

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FIG. 1

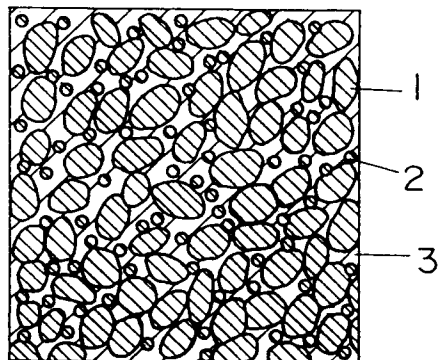


FIG. 2

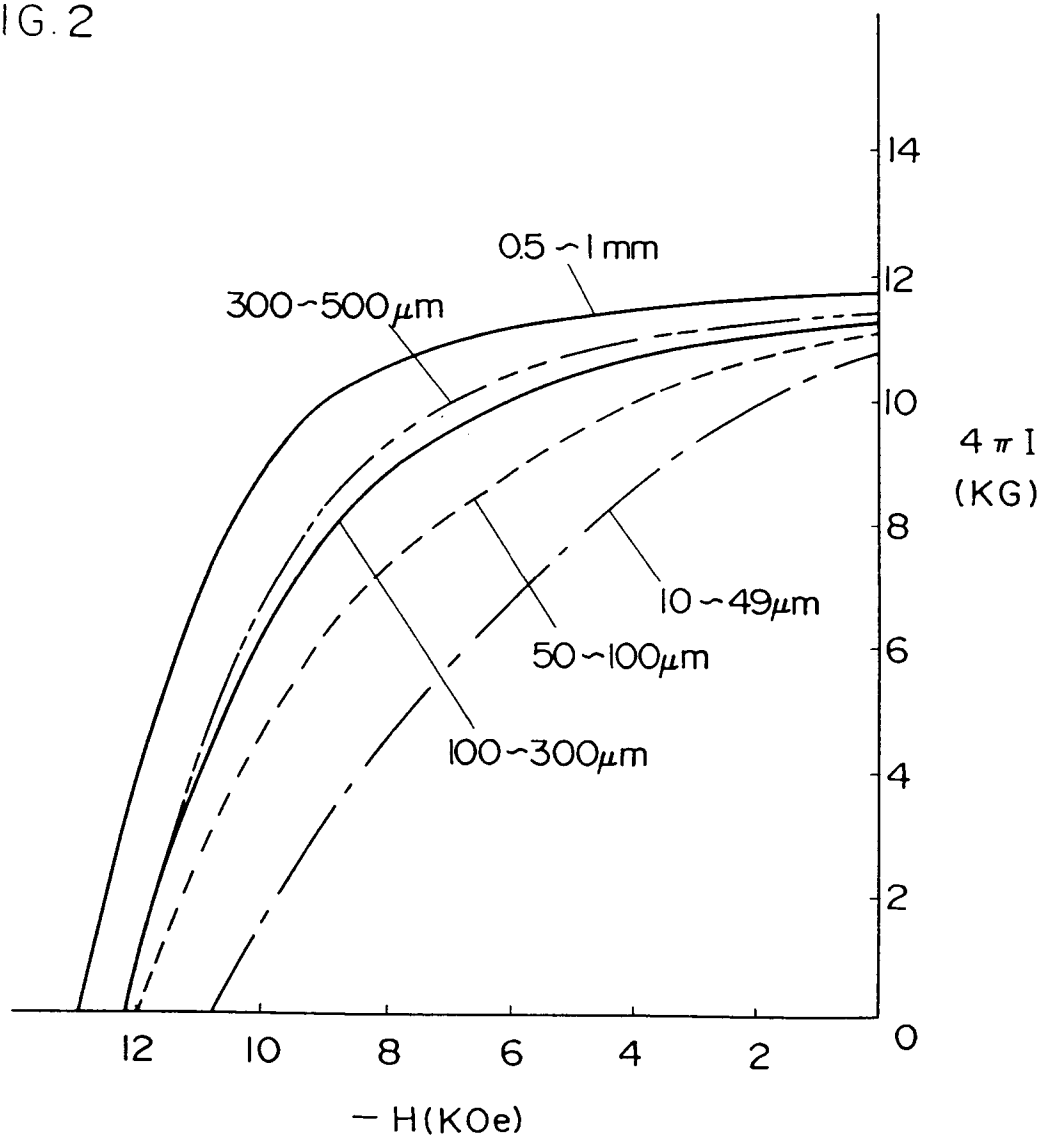


FIG. 3

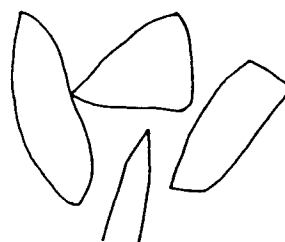


FIG. 4



FIG. 5

