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- Magnetic powder material and resin-bonded type magnet.
- There are disclosed a magnetic powder material which comprises a crystalline thermoplastic resin excellent in heat resistance which is coated or adhered to magnetic powder with 0.1 to 5 % by weight and a resin-bonded type magnet which comprises a cold compression molded material of said magnetic powder material. Also disclosed are methods for preparing these materials with high efficiency.

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MAGNETIC POWDER MATERIAL AND RESIN-BONDED TYPE MAGNET

BACKGROUND OF THE INVENTION

1. Field of the Invention

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This invention relates to a magnetic powder material and resin bonded-type magnets, more particularly, to a magnetic powder material suitable for compression molding, resin-bonded type magnet excellent in heat resistance, chemical resistance, etc., and a method for producing them.

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2. Description of the Related Arts

Heretofore, resin-bonded type magnets have mainly been prepared by the compression molding method or the injection molding method. Among these in the compression molding method, a thermosetting resin such as an epoxy resin, etc., has until now been used as a resin binder. However, since the thermosetting resin is extremely low in chemical stability with a lapse of time, the resulting resin-bonded type magnetic powder material is poor in preservability and also poor in production stability. Further, at preparation, it takes one hour or more for thermosetting processing so that productivity is low. Moreover, there are various problems in that a critical temperature for use of the resulting resin-bonded type magnet is 120° C or so, which is impractical, and it also lacks dimensional stability with a lapse of time.

Also, regarding the methods for obtaining resin-bonded type magnets by cold compression molding after coating the magnetic powder with a thermoplastic resin, various ones have been known. For example, in Japanese Patent Kokai No. 104254/1975, there is disclosed the method in which magnetic powder particles coated with Nylon precipitated from about 2% by weight of a solution is subjected to compression molding to obtain a resin-bonded type magnet. Further, in Japanese Patent Kokai No. 186908/1983, there is disclosed the method in which ferrite series magnetic powder is suspended in a radical polymerizable monomer solution to carry out polymerization, and then the magnetic powder particles coated their surfaces with resin are subjected to compression molding in a magnetic field to obtain a resin-bonded type magnet having a magnetic anisotropy. However, these are unsatisfactory in resin characteristics such as heat resistance, chemical resistance, etc., so they could not be used practically.

In recent years, a crystalline resin excellent in heat resistance, chemical resistance and dimensional stability, such as polyphenylenesulfide (PPS), polyether ether ketone (PEEK), etc. has been known. In Japanese Patent Kokai No. 113403/1985, there is disclosed the method in which the above PPS, PEEK and magnetic powder are fused and kneaded, and then a rare earth resin-bonded type magnet is obtained by injection molding.

However, the crystalline resin such as PPS or PEEK requires a high temperature for fusion molding such as 350°C or higher, so that there is a disadvantage that magnetic powder of the rare earth is likely to be oxidized at molding.

In addition to the above, various methods have been proposed and for example, in Japanese Patent Kokai No. 134517/1974 and No. 103309/1984, there are proposed the methods in which a mixture or a coated material of thermoplastic resin powder such as a polyamide, a polyolefin, a polycarbonate, etc. and magnetic powder is subjected to hot press molding. However, in this method, when removing a molded material from the mold, it takes a long time for cooling and solidification so that there is a problem of low productivity. Also, in Japanese Patent Kokai No. 186908/1983, there is described a radical polymerizable monomer such as methyl acrylate, etc. is brought into contact with ferrite series magnetic powder to carry out polymerization, and after coating the powder surface with the polymer, cold press molding is carried out. However, since heat resistance and chemical resistance of the binder resin are insufficient, it has not yet been used practically.

Further, in Japanese Patent Kokai No. 279106/1986, the technique is proposed in which by using PPS as a binder, after PPS is fused under heating and kneaded with magnetic powder, injection molding or extrusion molding is carried out. However, in order to carry out such a fusion under heating and kneading, fluidity in kneaded material is required and at least 20 to 40% by volume (6 to 15% by weight) of a resin binder content is required therefor. Thus, lowering in magnetic characteristics of the resulting magnet is inevitable and there is a serious problem that magnetic characteristics is lowered since the magnetic powder is exposed to high temperatures for a long time whereby oxidation of the magnetic powder is

promoted.

SUMMARY OF THE INVENTION

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An object of the present invention is to provide a resin-bonded type magnet having excellent heat resistance and chemical resistance as well as excellent magnetic characteristics.

Another object of the present invention is to provide a resin-bonded type magnet which is excellent in dimensional stability with a lapse of time, dimensional stability at molding, etc. in addition to the above mentioned characteristics.

A further object of the present invention is to provide a magnetic powder material suitable for molding of the above resin-bonded type magnet.

Also, a further object of the present invention is to provide methods for preparing the above resinbonded type magnet and magnetic powder material by simple procedures with high productivity and efficiency.

That is, the present invention relates to a magnetic powder material which comprises magnetic powder to which a crystalline thermoplastic resin with heat resistance is coated or adhered in amount of 0.1 to 5% by weight.

Also, the present invention relates to a resin-bonded type magnet comprising a compression molded material of the above magnetic powder material. Further, the present invention relates to a method for preparing the above magnetic powder material which comprises, to a mixture obtained by dissolving a crystalline thermoplastic resin with heat resistance in a solvent and dispersing magnetic powder, effecting (1) addition of a bad solvent of said resin, or (2) volatilization or evaporation of the solvent in said mixture, or (3) cooling said mixture.

Also, the present invention relates to a method for preparing the above magnetic powder material which comprises dissolving by heating a crystalline thermoplastic resin with heat resistance in a solvent, and then dispersing and mixing a gel obtained by cooling and magnetic powder, and crushing the resultant simultaneously with volatilizing or evaporating the solvent.

Further, the present invention relates to a method for preparing the above resin-bonded type magnet, characterized in that the above magnetic powder material is compression molded.

DESCRIPTION OF PREFERRED EMBODIMENTS

In the present invention, a crystalline thermoplastic resin excellent in heat resistance is used as a resin (a binder resin) to be coated on or adhered to magnetic powder. Here, as the thermoplastic resin, various ones can be used, but generally those having a melting point of 200° C or higher, preferably 230° C or higher. Among them, those having at least one -S- bond or -O- bond in the chemical bond skeleton.is particularly preferred. Specific examples of these resins include polyether ether ketone (PEEK), polyether ketone, polyphenylenesulfide (PPS), polysulfide ketone, etc.

Also, if the molecular weight of these resins is too large, there are problems in that pressure applied at molding mentioned thereinbelow becomes too large so that the resulting molded material sometimes reverts tack or crushing thereof becomes difficult. Further, if the molecular weight thereof is too small, coating with resin becomes difficult. Accordingly, the molecular weight of the resin should optionally be selected in view of the above matters. For example, in case of PPS, a limiting viscosity thereof in α -chloronaphthalene solvent at 206 °C is 0.1 dl/g or higher, preferably 0.15 to 0.3 dl/g and in case of PEEK, a limiting viscosity thereof in p-chlorophenol solvent at 60 °C is 0.3 dl/g or higher, preferably 0.3 to 0.85 dl/g.

On the other hand, the type of the magnetic powder is not particularly limited and various ones can be optionally selected depending on the purposes. Specific examples thereof include ferrite powder such as BaO*6Fe₂O₃, MnO*ZnO*Fe₂O₃, γ-Fe₃O₄ PbO*6Fe₂O₃, SrO*6Fe₂O₃, etc.; arnico powder such as MCA160, MCA230, MCB500, MCB580, MCB4DOH, etc. of JIS standard; rare earth cobalt powder such as SmCo₅, PrCo₅, NdCo₅, MMCo₅ (here, MM represents a Misch metal), SmPrCo₅, SmPrNdCo₅, SmMMCo₅, R₂Co₁₇ (wherein R represents a series of rare earth elements of atomic numbers from 58 to 71), Sm₂Co₁₇, Pr₂Co₁₇, Sm₂ (Co, Fe, Cu)₁₇ and Sm₂ (Co, Fe, Cu, M)₁₇ (wherein M represents Ti, Zr or Hf). Further, there can be mentioned rare earth*iron*boron powder (Nd₂Fe₁₄B, Nd₂Fe₁₂Co₂B, Pr₂Fe₁₄B, etc.). In addition, there can be mentioned Fe-Cr-Co magnetic powder, Mn-Al-C magnetic powder, Pt-Fe magnetic powder, cunife magnetic powder, etc.

In the present invention, the above magnetic powder can be used as it is by mixing with the above

thermoplastic resin, but in order to prevent oxidation of the magnetic powder and improve adhesiveness into a binder (the themoplastic resin), it is preferred to surface treat them with a coupling agent in an amount of 5% by weight or less, particularly 0.5 to 2.0% by weight based on the magnetic powder.

Here, coupling agents which can be used include various ones but titanate series and silane series ones are preferable. The titanate series coupling agents include isopropyltriisostearoyl titanate, isopropyltris(dioctylpyrophosphate)titanate, isopropyltri(N-aminoethyl-aminoethyl) titanate,

isopropyltridodecylbenzenesulfonyl titanate,

isopropylisostearoyldiacryl titanate,

isopropyltri(dioctylphosphate)titanate,

isopropyltricumylphenyl titanate,

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tetraisopropylbis(dioctylphosphite)titanate.

tetraoctylbis(ditridecylphosphite)titanate, tetra(2,2-diallyloxymethyl-1-butyl)bis(ditridecyl)phosphite titanate, bis(dioctylpyrophosphate)oxyacetate titanate,

bis(dioctylpyrophosphate)-ethylene titanate, etc. These may be used singly or in combination. Also, the silane series coupling agents include γ -mercapto-propyl-trimethoxysilane, 2-styryl-ethyltrimethoxysilane, N- β -(aminoethyl) γ -aminopropyl-trimethoxysilane, β -(3,4-epoxycyclohexyl)ethyltrimethoxysilane, γ -aminopropyl-trimethoxysilane, γ -glycidoxy-propyltrimethoxysilane, phenyltrimethoxysilane, methyldimethoxysilane, etc. and these may be used singly or in combination. Further, the titanium series coupling agents and the silane series coupling agent can be used in combination.

The surface treatment by the coupling agent can be carried out by wetting magnetic powder with 5 to 20% by volume of the coupling agent solution (using an alcohol or toluene as a solvent), and then drying it at a temperature not less than room temperature, preferably 120 to 150°C. According to the surface treatment due to the coupling agent, water repellent property and lubricity are applied to the magnetic powder so that the mechanical strength, etc. of the resin-bonded type magnet obtained after molding can be improved.

The magnetic powder material in the present invention can be obtained by coating or adhering the above crystalline thermoplastic resin with heat resistance onto the above magnetic powder with a ratio of 0.1 to 5% by weight, preferably 1 to 4% by weight. As the method for preparing the magnetic powder material by coating or adhering the thermoplastic resin on or to the magnetic powder, there may be mentioned the method in which said resin and magnetic powder are mixed in the temperature range between the crystal fusion initiating temperature and the melting point of the resin, and then the mixture is cooled to coat thereon or adhere thereto; by utilizing the crystallinity of the resin. However, this method requires high temperatures so that there is danger of causing deterioration of the magnetic powder due to oxidation, and sometimes it is difficult to uniformly disperse the magnetic powder and completely coat or adhere the resin thereon. Accordingly, as preferred methods for avoiding these problems, there can be mentioned (1) the method in which coating or adhering is effected by precipitating the resin in a solvent, (2) the method in which coating or adhering is effected by precipitating the resin by volatilizing the solvent, or (3) the method in which adhering is effected by mechanically pressing resin fine particles, smaller than the magnetic powder particles, on the surface thereof, etc.

For preparing the magnetic powder material, the crystalline thermoplastic resin with excellent heat resistance is coated on or adhered to the magnetic powder by the method as mentioned above (1) or (2), and at this time, the amount of the resin to be coated or adhered is 0.1 to 5% by weight (ratio based on the total magnetic powder material) as already mentioned. If the coated or adhered amount is less than 0.1 % by weight, the resin cannot act as the binder and the shape when molded cannot be retained. Also, if it exceeds 5% by weight, magnetic characteristics are decreased.

An amount of the coated resin can be calculated by dissolving the resin of the magnetic powder material to remove with p-chlorophenol or α -chloronaphthalene, etc., having strong dissolving power and measuring the weight decreased.

Next, as the solvent for dissolving the resin, it is preferred to use a polar solvent having high dissolving power, which can dissolve the above crystalline thermoplastic resin with heat resistance, and may include, for example, N-methylpyrrolidone, α -chloronaphthalene, dichloroacetic acid, 1,3-dimethyl-2-imidazolidinone, dimethylsulfoxide, dimethylacetamide, dimethylformamide, p-chlorophenol, etc.

The kind of the solvent to be used should be optionally selected depending on the kinds or molecular weights of the resin, and generally, it is preferred that α -chloronaphthalene is used when PPS is employed as the resin, and dichloroacetic acid or α -chloro-naphthalene is used when PEEK is used as the resin.

Also, an amount of the solvent may be varied depending on the amount of the resin supplied, or kinds,

grain size distribution, wettability, adhesiveness to the resin of the magnetic powder, etc. For example, when an anisotropic one such as ferrite magnetic powder or samarium cobalt magnetic powder is coated with a resin, in order to decrease aggregation of the magnetic powder with each other, it is preferred to make the condition wherein a mixed slurry of the magnetic powder and the resin solution is diluted to about 5 to 50% by weight. Accordingly, a ratio of the amount of the resin to the solvent, i.e. a polymer concentration (amount of charged resin (g)/amount of solvent (dl)) is preferably 0.1 to 5 (g/dl).

Also, in order to dissolve the resin in a solvent, it is carried out by charging the above solvent and the resin in a powder state in a suitable stirring tank and then heating while stirring. The heating temperature at this time is to be raised, for example, to 190° C or higher when N-methylpyrrolidone is used as the solvent, and to 250° C or higher when α -chloronaphthalene is used. Heating and stirring are preferably continued to uniformly dissolve the resin.

For effecting the resin coating to the magnetic powder by using the thus prepared resin solution, it is carried out by four kinds of methods mentioned hereinbelow, the method employed for the resin coating can be optionally selected depending on kinds of resin solution or magnetic powder to be used or conditions thereof.

(1) Method of using a bad solvent

By adding a bad solvent of the resin to a hot solution of the resin (a mixture wherein magnetic powder is dispersed in a solution dissolved the resin therein), the solubility thereof can be made lower so that the resin can be precipitated on the magnetic powder. The bad solvent means a solvent in which the resin is insoluble or little soluble.

The dropwise addition of the bad solvent is preferably started at temperatures wherein the hot solution of the resin is uniformly dissolved. That is, the temperature whereat almost all the resin in the solution does not start to precipitate upon cooling, and is preferably the temperature not more than the boiling point of the bad solvent.

As the bad solvent, an organic solvent other than α -chloronaphthalene (in the range of 205 to 250 °C) and water for PPS, an organic solvent other than dichloroacetic acid (150 °C \sim), α -chloronaphthalene (205 to 250 °C) and p-chlorophenol (50 °C \sim), and water for PEEK can be employed; so that it may be optionally selected depending on the molecular weight of the resin to be used, concentration of the resin and solubility of the resin depending on the dissolution temperature. Specific examples of the bad solvents include water, methanol, isopropyl alcohol, acetone, toluene, etc. Also, solvent with high boiling point such as N-methylpyrrolidone, α -chloronaphthalene, 1,3-dimethyl-2-imidazolidinone, dimethylsulfoxide, dimethylacetamide, dimethylformamide, etc. may be used as the bad solvent depending on the temperature.

The added amount of the bad solvent depends on the concentraion of the resin solution, but generally an amount equivalent to the amount of the basic solvent or more is preferred. As to the method of addition, it is preferred to supply with liquid drops as minute as possible (for example, a liquid in the atomizing state) under vigorous dispersion and stirring.

(2) Method due to volatilizaiton and evaporation

The method in which the solvent is volatilized and evaporated to precipitate the resin on the magnetic powder may be carried out, for example, by supplying with a liquid transferring pump, a mixed slurry (mixture) of the magnetic powder, the resin solution and a low boiling point solvent having high solubility to the resin to a heating tube overheated and explosively blowing out in a room with high temperature and vacuum, to evaporate (volatilize) the solvent in a moment.

However, in this case, when selection of a mixed solvent with a low boiling point solvent having high solubility to the resin is difficult, evaporation (volatilization) of the solvent in a moment can be carried out with a good solvent alone if temperatures of the heating tube and the room with high temperature and vacuum are not less than the boiling point of the solvent.

As an apparatus for carrying out such procedures, there may be applied the Cracks System (an instantaneous, vacuum drying apparatus) produced by Orient Chemical Industry Co., Ltd.

(3) The method by cooling

By cooling a hot solution of the resin (a mixed solution of magnetic powder dispersed in a solution of dissolved resin) and decreasing the solubility of the resin, the resin can be precipitated on the magnetic powder. Further, the low molecular weight component, which is a part of the solution and cannot be precipitated, can be precipitated by adding a bad solvent.

More specifically, it can be carried out by mixing the hot solution of the resin uniformly dissolved with magnetic powder in a stirring tank having a jacket, and cooling the mixture to room temperature while effecting dispersion and stirring with a wet dispersing and stirring apparatus and flowing cold water through the jacket.

Further, by adding a bad solvent with an amount equivalent to or more than the volume of the resin solution, almost all of the resin can be precipitated.

As the above wet dispersing and stirring apparatus, there may be applied, for example, an ultrahomomixer produced by Nippon Seiki Seisakusho K.K. or an ultradisperser produced by IKA K.K.

The cooling rate and precipitating time may optionally be selected since the time of forming precipitates is different depending on the solubility of the resin or dissolution conditions, but generally it is preferred to provide a precipitating time of one hour or more after cooling to room temperature for 1 to 2 hours. If no precipitation time is provided, it may cause delay in precipitation of the resin due to its supercooled state.

Also, bad solvents to be used include water, methanol, isopropyl alcohol, acetone, toluene, etc.

Also, the case where cooling is effected by using particularly concentrated solution of the resin is as follows:

When a hot concentrated solution is cooled, it does not stop at a jelly-like state but rather as solid material such as wax or soap. By using this physical property, it can be effected to coat the resin on the magnetic powder simultaneously with grinding thereof. That is, by adding dropwise a hot solution of the resin to magnetic powder preheated to a temperature which is the same as the hot solution of the resin under high speed dispersion and stirring, and cooling, the resin can be coated on the magnetic powder and grinding can be carried out simultaneously by precipitating and solidifying the resin.

As the stirring disperser, a dry type disperser such as Henshel mixer, high speed mixer and super mixer, etc can be used. At the time of disperse by these mixers, grinding ability can be increased by adding a ball made of ceramics such as an alumina, etc.

Also, the concentration of the resin solution is preferably 2 (g/dl) or more. If it is less than 2 (g/dl), it becomes a jelly-like state and aggregation is inevitable so that it is impossible to carry out coating and grinding simultaneously. In this method, when the amount of the solvent is decreased as little as possible, productivity can be increased but as the amount of the solvent is decreased, the resin does not dissolve completely in the solvent whereby a uniform solution cannot be obtained. That is, the solution concentration may optionally be selected at which coating and grinding can be carried out simultaneously and easily in the concentration range wherein the resin is dissolved in the solvent uniformly, and generally it is preferred to 5 to 25 (g/dl). Also, by drying under reduced pressure at a temperature of 100° C or higher while carrying out dispersion and grinding simultaneously in a mixer, the solvent can be removed.

The resin-coated magnetic powder obtained by this method is subjected to compression molding without removing the solvent to carry out desolvation (solvent removal) simultaneously with compression molding. Also, when desolvation is effected before molding, after adding a bad solvent so as to become a slurry containing 30 to 50% by weight of the magnetic powder, the solvent can be removed with the bad solvent by using an instantaneous vacuum drying device.

(4) Method by volatilization or evaporation of the solvent in a gel

A gel (solid component) previously prepared from a resin solution and magnetic powder are dispersed and mixed, and then removal of the solvent was carried out while effecting dispersion and grinding whereby adhering to or coating on the magnetic powder can be carried out.

As the mixer, a Henshel mixer in method (3) can be used. Also, in order to improve grinding ability, balls may be used in combination. The size, hardness or number of balls may be optionally determined depending on grinding ability.

The concentration of the resin solution is preferably 5 to 25 g/dl. If it is less than 5 g/dl, an amount of the solvent to be used increases so that productivity decreases, while if it exceeds 25 g/dl, dispersion of a gel is likely to become ununiform.

Regarding the magnetic powder coated or adhered with the thermoplastic resin, obtained by the above methods (1) to (4), grinding treatment is carried out, if necessary. For this grinding treatment, it is preferred

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to use an impact type mill and more specifically, a hammer mill (for example, a sample mill produced by Fuji Powdal K.K., or an atomizer) may be applied. This is to supply a resin-coated magnetic powder from a hopper via a feeder to a hammer-shaped rotary wing rotating with high speed (6000 to 12000 rpm), to collide with the hammer, whereby the grinding treatment is carried out.

This grinding is generally carried out at a normal temperature and normal pressure, but it may be carried out at a low temperature by using a coolant such as dry ice, etc. or a liquid nitrogen atomsphere. Also, in case of using a rare earth magnetic powder, in order to avoid oxidation due to collision, it is preferred to effect the procedure at a low temperature under an inert gas atmosphere such as liquid nitrogen, etc. The shearing force to be applied may be optionally selected depending on the number of rotation and numbers of grinding treatments.

Thus, the magnetic powder material of the present invention can be prepared. Also, for preparing a resin-bonded type magnet of the present invention, the resin-coated (or adhered) magnetic powder material obtained by the above methods is, if necessary, ground, and then molded. For molding, various means such as the hot compression molding, the cold compression molding, etc can be used. Among these, the cold compression molding is the particularly optimum one since productivity is high and there is no fear in lowering magnetic characteristics.

For conducting the cold compression molding, the molding pressure may be set at or more than a pressure that causes plastic deformation to the binder resin, but generally it is optionally selected within the range of not less than 1 t/cm². Also, a temperature is sufficient at a room temperature. By the cold compression, the binder resin causes plastic deformation to adhere therewith so that strength of the molded material obtained is increased, whereby a resin-bonded type magnet having excellent physical properties can be obtained. In the present invention, since the crystalline thermoplastic resin is used as the binder resin, particularly, compression molding is possible even at a room temperature. Here if the amorphous thermoplastic resin is used, it is hard or impossible to effect cold compression molding at not more than the glass transition temperature since elongation at breakage is small.

Also, for effecting this cold compression, an anisotropic resin-bonded type magnet can be obtained while the procedure is carried out by applying a magnetic field. In this case, it is effective to apply a magnetic field of 15 kOe or more. Also, if the cold compression molding is effected without applying a magnetic field, an isotropic resin-bonded type magnet which is capable of magnetizing to all directions can be obtained.

After compression molding, if necessary, heat treatment may be carried out. This heat treatment may only be carried out by allowing the magnet to stand at a temperature not less than the softening (pour) point or the melting point of the resin for several minutes. By this heat treatment, the resin is fused and crystallized whereby recombination is progressed and the strength of the resin-bonded type magnet can further be improved.

Further, magnetization after molding is carried out by the conventional method such as applying a magnetic field of 20 kOe or more.

In the method for preparing the resin-bonded type magnet according to the present invention, a magnetic powder material to which a resin is coated or adhered by the other methods may be used, and the magnetic powder material obtained by the method for preparing the magnetic powder material of the present invention may be molded to a magnet obtained by the other methods, but by combining both methods of the present invention, production stability and mass productivity can be improved, whereby magnets having excellent characteristics can easily be obtained.

As explained above, the magnetic powder material and the resin-bonded type magnet of the present invention are good in preservability as compared with the case where a thermosetting resin is used as a binder since its chemical stability with a lapse of time is excellent.

On the other hand, according to the method for preparing the resin-bonded type magnet of the present invention, a practically endurable magnet can be prepared easily, which is excellent in heat resistance, chemical resistance, water absorption resistance, dimensional stability with a lapse of time, dimensional stability at molding, linear expansion coefficient, etc. And yet, a magnet can be molded by the cold compression molding, without heating, so that production step is simple and inexpensive in costs such as equipment and working, and also excellent in production stability and mass productivity.

Accordingly, the resin-bonded type magnet in the present invention can be widely and effectively utilized for various electric and electronic devices including motors, etc. used in places with high temperature circumstances or requiring chemical resistance.

Also, according to the method for preparing the magnetic powder material of the present invention, a magnetic powder material suitable with the above method for preparing the resin-bonded type magnet can be effectively prepared.

Next, the present invention will be described in more detail by referring to Examples. Starting materials and devices used in the following Examples are as follows:

5 (1) Magnet powder

Ferrite powder

Strontium ferrite; SrO 6Fe₂O₃

OP-71 produced by Nippon Bengara Industry K.K. (a product surface treated with a silane coupling agent)
Rare earth cobalt powder

Samarium cobalt 2-17 series; Sm₂Co₁₇

R-30 produced by Shinetsu Chemical Industries, Ltd. (32 mesh under):

Into a bortex pulverizer, 3 kg of magnetic powder and 5 liters of isopropanol were introduced, and after replacing the atmosphere with N_2 gas therein sufficiently, pulverization was carried out for 7 minutes and classified to obtain a powder having an average particle size of 37 μ m. The resulting magnetic powder (3 kg) was introduced into a supermixer and the temperature was raised to 100° C under a N_2 gas atmosphere, and 300 g of an isopropanol solution containing 10 % of a silane coupling agent (A-1120 produced by Nippon Unicar K.K. (N- β -aminoethyl- γ -amino-propyltrimethoxy-silane)) was added dropwise over 5 minutes. Thereafter, stirring was continued for 10 minutes, subsequently desolvation (solvent removal) was carried out by blowing nitrogen gas. Then, the mixture was maintained in an oven at 100° C for one hour.

Neodium series magnetic powder

(Rare earth-iron-boron powder): Nd₂Fe₁₄B

MQ-II powder produced by General Motors Co., Ltd.

Into a ball mill was introduced 3 kg of magnetic powder, and while stirring, 500 g of a toluene solution containing 3 % by weight of titanate coupling agent (KRTTS: isopropyltriisostearoyltitanate produced by Ajinomoto K.K.) was added dropwise and the mixture was treated for 6 hours. Under a 80° C bath, the mixture was dried by using an aspirator, and further vacuum dried at 60° C and classified to obtain a powder having an average particle size of $37 \,\mu m$.

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(2) Resin binder

Polyphenylene sulfide (PPS): Examples 1 to 5,7,8 and 11 are produced by Philips Co., Ltd. and others are produced by Idemitsu Petrochemical Co., Ltd. (limiting viscosity 0.2 dl/g, 206° C, α -chloronaphthalene).

Polyether ether ketone (PEEK): Produced by Mitsui Toatsu K.K., limiting viscosity 0.85 dl/g.

Epoxy resin: Mixture of Epirets SU-8 (produced by Celaneese Co., Ltd.) (R) and 1-(2-hydroxy-propyl)-2-methylimidazole (C) (C/R ratio = 0.04).

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(3) Stirring disperser

Ultradisperser produced by IKA Co., Ltd. (rotary number: 10000 rotations per minute).

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(4) Instantaneous vacuum drying device

Cracks system produced by Orient Chemical Industry Co., Ltd.

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(5) Dipserser (High speed mixer)

LFG-GS-1 Type (agitator rotary number: 2000 rotations per minute) produced by Shinko Industry Co., Ltd.

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Examples 1 to 12

Here, by using starting materials and conditions shown in Table 1, magnets were prepared by cold compression molding.

Magnet powder, a binder resin and α -chloronaphthalene or p-chlorophenol as a solvent were mixed in a flask and the binder was dissolved by heating to 240°C under an argon stream, and then the mixture was gradually cooled to 50°C over 4 hours while stirring to coat the surface of the magnetic powder with the binder resin by precipitation. After washing the residual solvent, it was dried. In a flask was charged 10 g (the value precisely measured is W₁) of the magnetic powder after resin coating, and by using 150 ml of α -chloronaphthalene in case of PPS and 150 ml of p-chlorophenol in case of PEEK, dissolution and extraction under heating was carried out at a temperature of 250°C in case of PPS and at 60°C in case of PEEK. After repeating the above procedure by causing no nebula to form when water was added to the extracted filtrate, p-chlorophenol remained in the magnetic powder was removed by methanol and then the powder was vacuum dried at 50°C. A weight (W₂) of the ççresulting magnetic powder removing the coated resin was measured and a decreased weight was made as the amount of the coated resin. The amount of the coated resin (% by weight) was calculated from the following equation (hereinafter the same).

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Coated polymer amount =
$$\frac{W_1 - W_2}{W_1} \times 100$$
(% by weight)

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Provided that when the strontium ferrite powder was used, the powder was ground by a grinding machine after coating with the resin.

Then, by using a magnetic field compression molding device (produced by High Tech Co., Ltd.), molding was effected at a magnetic field of 15 kOe and at room temperature to obtain a square pillar sample having an approximate dimension of 8 x 14 x 7 mm and a cylinder sample having a diameter of 20 mm and 10 g/each.

The resulting samples were subjected to heat treatment in an oven of an argon atmosphere for 3 minutes, and then magnetization was carried out in a magnetic field of 20 kOe to obtain permanent magnets.

With respect to these permanent magnets, the following experiments were carried out and the results are shown in Table 1.

Thermal deformation temperature: according to ASTM-D648.

Load 18.6 kg/cm²

Compression strength: according to JIS-K7208. Water absorbing ratio: according to ASTM-D570.

Comparative Examples 1 and 2

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An epoxy resin, magnetic powder and acetone were mixed at room temperature, and after impregnation, desolvation (solvent removal) was carried out under reduced pressure. The resulting magnetic powder was compression molded in a magnetic field in the same manner as in Examples described above, and then curing treatment was carried out under an argon atmosphere at 150 °C for one hour. This was magnetized in the same manner as in Examples to obtain permanent magnets.

With respect to the resulting magnets, the same experiments were carried out and the results are shown in Table 1.

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5	conditions	Heat treatment temperature (°C)	310	305	300	305	305	370	305	300	370	370	305	370	150	150
10	Molding	Molding pressure (t/cm)	4	Ditto	Ditto	S.	9	4	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto
		Heating (°C)	250	Ditto	Ditto '	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Ditto	Room temperature	Room temperature
20		r i														
25	Coating conditions	Amount of solvent (ml)	30	40	50	40	40	40	30	40	30	40	40	40	20	50
Table 1	Coating	Aamount magnetic powder (9)	98.6	98.1	97.0	98.1	98.1	98.2	0.66	98.5	0.66	98.8	98.5	98.5	0.86	98.0
30		n i l							•	•			•			
		Amount of resin (g)	1.40	1.90	3.00	1.90	1.90	1.80	1.00	1.50	1.00	1.20	1.50	1.50	2.0	2.0
35	1															
40	Resin	Coated amount (% by weight)	1.32	1.84	2.96	1.84	1.84	1,76	0.93	1.44	96.0	1.12	1.27	1.25	2.0	2.0
		Kinds	PPS	Ditto	Ditto	Ditto	Ditto	PEEK	Sdd	Ditto	PEEK	Ditto	PPS	PEEK	Epoxy resin	Ditto
45	I	·			Q	۵	Ω			a		Ω			шн	1
		Magnetic powder	SrO 6Fe ₂ 0 ₃	Ditto	Ditto	Ditto	Ditto	Ditto	$\mathrm{Sm_2Co_{17}}$	Ditto	Ditto	Ditto	Nd ₂ Fe ₁₄ B	Ditto	$\mathrm{Sm_2^{Co}_{17}}$	Nd ₂ Fe ₁₄ B
50		NO.	Example 1	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7	Example 8	Example 9	Example 10	Example 11	Example 12	Comparative Example 1	Comparative Example 2
55	!		Ä	Ä	EX	Ä	ΞX	EX	EX	ΕX	EX	Ex	EX	EX	S A	SÄ

5	1															
10		Maximum energy product (BH) max (MGOe)	1.70	1.62	1.38	1.65	1.72	1.64	14.1	12.8	14.3	13.2	7.3	7.2	14.7	8.0
15		Magnetic characteristics Coersive ener ty force bhc (BH)	2.30	2.37	2.12	2.29	2.31	2.39	6.2	6.3	6.2	6.4	4.8	4.8	8.0.	5.3
20		lagnet Y	1													
25		Residual magnetic flux density	2.65	2.56	2.37	2.61	2.68	2.57	8.0	7.5	8.0	7.6	5.4	5.4	9.9	6.1
30	Table 1 (continued)	of molded material Water Sorbing Compression strength 10-3 %) (kg/cm)	810	780	750	800	820	1120	160	740	1010	1000	740	. 086	460	380
35		ng (%														
40		<pre>istics of mol Water absorbing ratio (10-3 %)</pre>	3.7	4.8	5.1	4.6	4.5	4.9	3.8	4.9	4.3	4.9	4.6	4.7	8.2	8.0
45		Character Thermal deformation temperature (*C)	280	279	277	280	280	328	280	279	330	328	279	328	200	. 205
50		. ov	Example 1	Example 2	Example 3	Example 4	Examplt 5	Example 6	Example 7	Example 8	Example 9	Example 10	Example 11	Example 12	Comparative Example 1	Comparative Example 2
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a. Coating step

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Example 13

In to a glass apparatus supplied therein 975 g of ferrite powder preheated to 200° C and equipped with a 5 liter jacket was poured at 250° C 2 liters of α -chloronaphthalene resin solution dissolved therein 25 g of PPS resin, and then the mixture was cooled to room temperature by flowing cold water in the jacket while vigorously stirring and dispersing with a stirring disperser.

Thereafter, stirring and dispersing were carried out while adding dropwise 6 liters of isopropyl alcohol from a dropping funnel over 30 minutes. The resulting slurry was subjected to solvent replacement due to decantation using 2 liters of isopropyl alcohol and repeated three times, and then the solvent was removed by an instantaneous vacuum drying device. Working conditions of the instantaneous vacuum drying device were a heating pipe jacket temperature of 130°C, a temperature in the powder collecting room of 120°C, vacuum degree of 35 torr, a liquid transferring pump of 250 cc/min and a discharging pressure of 0.1 to 2.5 kg/cm².

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Example 14

In the same manner as in Example 13 except for replacing the ferrite powder with neodium series magnetic powder, resin coating was carried out under an inert atmosphere of argon stream.

Example 15

In the same manner as in Example 13 except for replacing the ferrite powder with samarium cobalt series magnetic powder, resin coating was carried out under an inert atmosphere of argon stream.

Example 16

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In to the glass apparatus supplied therein 975 g of ferrite powder preheated to 150°C and equipped with a 5 liter jacket was poured at 180°C 2 liters of dichloroacetic acid resin solution dissolved therein 25 g of PEEK resin, and then the mixture was cooled to room temperature by flowing cold water in the jacket while vigorously stirring and dispersing with a stirring disperser.

Thereafter, stirring and dispersing were carried out while adding dropwise 6 liters of isopropyl alcohol from a dropping funnel over 30 minutes, The resulting slurry was subjected to solvent replacement due to decantation using 2 liters of isopropyl alcohol and repeated three times, and then the solvent was removed by an instantaneous vacuum drying device. Working conditions of the instantaneous vacuum drying device were a heating pipe jacket temperature of 130°C, a temperature in the powder collecting room of 120°C, vacuum degree of 30 torr, a liquid transferring pump of 250 cc/min and a discharging pressure of 0.1 to 2.0 kg/cm².

Example 17

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In the same manner as in Example 16 except for replacing the ferrite powder with neodium series magnetic powder, resin coating was carried out in the same manner as in Example 16 under an inert atmosphere of argon stream.

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Example 18

In the same manner as in Example 16 except for replacing the ferrite powder with samarium cobalt

series magnetic powder, resin coating was carried out in the same manner as in Example 16 under an inert atmosphere of argon stream.

5 Example 19

In 3 liters of α-chloronaphthalene was dissolved 18 g of PPS resin at 205°C, and while strongly stirring and dispersing with a stirring disperser, 982 g of ferrite magnetic powder was supplied to an instantaneous vacuum drying apparatus through a transfer pump to effect direct solvent removal. Working conditions of the instantaneous vacuum drying apparatus were a heating pipe jacket temperature of 170°C, a temperature in a powder collecting room of 160°C, vacuum degree of 30 torr, a liquid transferring pump of 250 cc/min and a discharging pressure of 0.1 to 0.2 kg/cm².

15 Example 20

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In the same manner as in Example 19 except for replacing the ferrite powder with neodium series magnetic powder, resin coating was carried out as in Example 19 under an inert atmosphere of argon stream.

Example 21

In the same manner as in Example 19 except for replacing the ferrite powder with samarium cobalt series magnetic powder, resin coating was carried out as in Example 19 under an inert atmosphere of argon stream.

Example 22.

In to 2 liters of dichloroacetic acid was dissolved 18 g of PEEK resin at 150 °C, and while strongly stirring and dispersing with a stirring disperser, 982 g of ferrite magnetic powder was supplied to an instantaneous vacuum drying apparatus with a stirring disperser through a transfer pump to effect direct solvent removal. Working conditions of the instantaneous vacuum drying apparatus were a heating pipe jacket temperature of 160 °C, a temperature in the powder collecting room of 160 °C, vacuum degree of 30 torr, a liquid transferring pump of 250 cc/min and a discharging pressure of 0.1 to 2.5 kg/cm².

Example 23

In the same manner as in Example 22 except for replacing the ferrite powder with neodium series magnetic powder, resin coating was carried out as in Example 22 under an inert atmosphere of argon stream.

Example 24

In the same manner as in Example 22 except for replacing the ferrite powder with samarium cobalt series magnetic powder, resin coating was carried out as in Example 22 under an inert atmosphere of argon stream.

Example 25

In 200 ml of α-chloronaphthalene was dissolved under heating 18 g of PPS at 250°C, and this was added dropwise in an apparatus of a disperser supplied therein 982 g of ferrite powder preheated to 240°C under high speed dispersing and stirring over 5 minutes. Then, while the mixture was cooling to room temperature by flowing water in the jacket, it was dispersed, mixed and stirred by setting an agitator to

1500 rpm. At the same time, solvent removal was also carried out at 130°C and by coexisting balls to effect grinding, whereby resin coated magnetic powder was prepared. A hundred balls made of alumina and having a diameter of 10 mm were used.

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Example 26

In the same manner as in Example 25 except for replacing the ferrite powder with neodium series magnetic powder, resin coating was carried out in the same manner as in Example 25 under an inert atmosphere of argon stream.

Example 27

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In the same manner as in Example 25 except for replacing the ferrite powder with samarium cobalt series magnetic powder, resin coating was carried out in the same manner as in Example 25 under an inert atmosphere of argon stream.

20 Example 28

In the same manner as in Example 25 except for replacing PPS with PEEK, resin coating was carried out in the same manner as in Example 25 under an inert atmosphere of argon stream.

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Example 29

In the same manner as in Example 26 except for replacing PPS with PEEK, resin coating was carried out in the same manner as in Example 26 under an inert atmosphere of argon stream.

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Example 30

In the same manner as in Example 27 except for replacing PPS with PEEK, resin coating was carried out in the same manner as in Example 27 under an inert atmosphere of argon stream.

Examples 31 to 36

40 In to 120 ml o

In to 120 ml of α -choronaphthalene as a solvent was dissolved 18 g of the following resins by heating at 240 $^{\circ}$ C under stirring to obtain a hot solution. 15 g/dl of this hot solution was cooled to room temperature to prepare a solid material (gel).

Next, 982 g of the following magnetic powder and the previously prepared gel (15 g/dl) containing a solvent were supplied in a disperser and dispersing and mixing were carried out with an agitator at 1000 rpm. Thereafter, while carrying out vacuum drying at 130 °C, grinding with an agitator at 1500 rpm (in the copresence of 100 balls having a diameter of 10 mm and made of alumina) and simultaneously with solvent removal were carried out to prepare resin coated magnetic powder. Provided that when rare earth series magnetic powder was used, supplying was effected under inert atmosphere.

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Kinds of magnetic powder and resin

Example 31

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Ferrite magnetic powder and PPS gel

Example 32

Neodium series magnetic powder and PPS gel

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Example 33

Samarium cobalt series magnetic powder and PPS gel

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Example 34

Ferrite magnetic powder and PEEK gel

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Example 35

Neodium series magnetic powder and PEEK gel

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Example 36

Samarium cobalt series magnetic powder and PEEK gel

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b. Grinding step

The ferrite magnetic resin-coated powder obtained in the above Examples 13, 16, 19 and 22 were effected grinding twice by using a grinding machine (produced by Fuji Powdal K.K., Sample mill) at 10000 rpm, respectively, and the rare earth series magnetic resin-coated powder obtained in Examples 14, 15, 17, 18, 20, 21, 23 and 24 were effected grinding once by useing the same machine and flowing a liquid nitrogen with copresence thereof at 6000 rpm, respectively.

s c. Compression molding step

The anisotropic ferrite magnetic resin-coated powder of the above Examples 13, 16, 19 and 22 after grinding, and the anisotropic ferrite magnetic resin-coated powder obtained in Examples 25, 28, 31 and 34 were subjected to compression molding at room temperature and an applied pressure of 3 ton/cm² in a magnetic field of 10 kOe.

The anisotropic samarium cobalt magnetic resin-coated powder of the above Examples 14, 17, 20 and 23 after grinding, and the anisotropic samarium cobalt magnet resin-coated powder obtained in Examples 26, 29, 33 and 36 were subjected to compression molding at room temperature and an applied pressure of 6 ton/cm² in a magnetic field of 10 kOe.

The isotropic needium magnetic resin-coated powder of the above Examples 14, 18, 21 and 24 after grinding, and the isotropic needium magnetic resin-coated powder obtained in Examples 27, 30, 32 and 35 were subjected to compression molding at room temperature and an applied pressure of 7 ton/cm² in a non-magnetic field.

A test piece for measuring thermal deformation temperature mentioned hereinafter was molded to a square pillar shape having 8 X 14 x 7 mm and a test plece for measuring bending strength was molded to a shape of 40 x 4 x 3 mm, respectively.

d. Thermal treatment

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With respect to each magnet after the above compression molding, it was exposed at 350°C for 3 minutes and then at 260°C for 5 minutes. Provided that rare earth ones were subjected under an inert atmosphere of argon stream.

e. Magnetization

To the anisotropic ferrite magnet, 15 kOe of the magnetic field was applied and to the rare earth series magnet, 20 kOe was applied.

Regarding each magnet thus prepared, various experiments were carried out and the results are shown in Table 2. The experimental conditions are as follows:

Bending strength: According to JIS-R1601

Thermal deformation temperature: According to ASTM-D648 Load 18.6 kg/cm²

Compression strength: According to JIS-K7208

Table

	,				,			
	Thermal deformation temporature	Water absorbing ratio	Linear expansion coefficient	Bending strength	Residual magnetic flux	Specific coersive force	Maximum energy product	Coated resin amount
	(20)	(10 ⁻³ %)	(10^{-3}) $(\text{cm/cm}^{\circ}\text{C})\text{x}10^{-6}$	(kg/cm^2)	Dr(KG)	iHc(KOe)	(BH) _{max} (MODe)	(Wt 8)
Example 13	246	4.9	3.0	650	2.55	2.28	1.46	2.1
Example 14	248	4.3	3.2	009	6.10	8.25	8.30	1.3
Example 15	249	4.6	3.1	700	8.50	10.50	16.30	1.8
Example 16	290	6.3	3.6	550	2.57	2.18	1.49	2.3
Example 17	285	5.4	3.8	200	00.9	8.20	8.00	1.5
Example 18	287	5.7	3.7	009	8.45	10.55	16.20	1.9
Example 19	250	4.7	3.0	029	2.65	2.36	1,55	1.8
Example 20	248	0.9	3.2	620	6.30	8.30	8.50	1.8
Example 21	246	5.5	3.1	730	8.60	10.60	16,35	1.8
Example 22	290	0.9	3.8	570	2.60	2.30	1.50	1.8

Table 2 (Contd)

	Thermal deforma- tion tem- perature	Water absorbing ratio	Linear expansion coefficient	Bending strength	Residual magnetic flux density	Specific coersive force	Maximum energy product (BH) (M30e)	Coated resin amount
Example 23	287	-	3.7	520	6.20	8.20	8.26	1.8
Example 24	288	5.8	3.6	620	8.50	10.50	16.15	1.8
Example 25	244	4.3	3.1	089	2.60	2.36	1.56	1.7
Example 26	246	4.0	3.3	630	6.30	8.45	8.60	1.7
Example 27	247	4.3	3.2	730	8.70	11.00	17.50	1.7
Example 28	288	5.5	3.7	580	2.62	2.32	1.59	1.7
Example 29	283	5.1	3.9	530	6.50	8.50	8.80	1.8
Example 30	285	5.4	3.8	630	8.65	10.70	16.80	1.8
Example 31	277	4.5	3.1	630	2.54	2.25	1.45	1.8
Example 32	276	4.3	3.3	009	6.12	8.34	8.32	1.7
Example 33	280	4.5	3.0	069	8.55	10.55	16.40	1.8
Example 34	325	5.5	3.5	220	2.57	2.20	1.52	1.8
Example 35	322	5.3	3.7	510	00.9	8.25	8.20	1.7
Example 36	326	5.6	3.6	680	8.48	10.56	16.22	1.8

Also, regarding the magnet of PPS-coated obtained in Example 14 and the magnet of PEEK-coated obtained in Example 23, chemical resistance, dimensional stability with a lapse of time and dimensional stability at molding were evaluated. The results are shown in Table 3 (chemical resistance of the magnet PPS-coated), Table 4 (chemical resistance of the magnet PEEK-coated), Table 5, (dimensional stability with

a lapse of time) and Table 6 (dimensional stability at molding).

The chemical resistance was according to JIS-K7114 and visual inspection and measurements of changes in weight and dimension were carried out.

Table 3

Chemicals	Chemical resistance	Chemicals	Chemical resistance
Hydrochloric acid (10 %) Sulfuric acid (10 %) dil. Nitric acid (5 %) conc. aqueous ammonia Saline (saturated) Potassium carbonate (saturated) Formic acid (10 %) Acetic acid Pyridine Benzene	0000000000	Toluene Xylene Methyl alcohol Butyl alcohol Formalin Acetone Methyl alcohol Ethyl acetate Diethyl ether Trichloroethylene Carbon tetrachloride	00000000000
No change in weight and dimer			

O: Usable depending on the conditions.

 Δ : Dissolved or swelled.

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Table 4

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Chemicals	Chemical resistance	Chemicals .	Chemical resistance
Hydrochloric acid (10 %) Sulfuric acid (10 %) dil. Nitric acid (5 %) conc. aqueous ammonia Saline (saturated) Potassium carbonate (saturated) Formic acid (10 %) Acetic acid Pyridine Benzene	00000000	Toluene Xylene Methyl alcohol Butyl alcohol Formalin Acetone Methyl alcohol Ethyl acetate Diethyl ether Trichloroethylene Carbon tetrachloride	00000000000

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(a): No change in weight and dimension.

O: Usable depending on the conditions.

 Δ : Dissolved or swelled.

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Table 5

Changed ratio in dimension (%)

PPS 0.001

PEEK 0.004

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Maximum value of dimension changed in a month.

$$\begin{array}{c} \text{Dimension} & \text{Standard} \\ \text{Changed ratio} & \text{(%)} &= \frac{\text{at present} - \text{dimension}}{\text{Standard dimension}} \times 100 \\ \end{array}$$

Standard dimension: Dimension immediately after thermal treatment (room temperature) Test piece: Approximate dimension 8 x 14 x 7 mm

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	Changed ratio in dimension after heat treatment (%)
PPS	0.012
PEEK	0.010

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Claims

- 1. A magnetic powder material which comprises magnetic powder to which a crystalline thermoplastic resin with heat resistance is coated or adhered in an amount of 0.1 to 5% by weight.
 - 2. A resin-bonded type magnet comprising a cold compression molded material of the magnetic powder material according to Claim 1.
- 3. A method for preparing a magnetic powder material according to Claim 1, which comprises adding a bad solvent of the thermoplastic resin to a mixture of the crystalline thermoplastic resin with heat resistance dissolved therein and magnetic powder dispersed therein.
- 4. A method for preparing a magnetic powder material according to Claim 1, which comprises volatilizing and evaporating a solvent in a mixture of the crystalline thermoplastic resin with heat resistance dissolved therein and magnetic powder dispersed therein.
- 5. A method for preparing a magnetic powder material according to Claim 1, which comprises cooling a mixture of the thermoplastic resin with heat resistance dissolved therein and magnetic powder dispersed therein.
- 6. A method for preparing a magnetic powder material according to Claim 1, which comprises effecting dispersion, mixing and grinding of a gel prepared by dissolving the crystalline thermoplastic resin having with heat resistance in a solvent by heating and then cooling, with magnetic powder, and simultaneously effecting volatilization and evaporation of the solvent.
- 7. A method according to any of Claims 3 to 6, wherein said magnetic powder is surface treated with 5% by weight or less of a coupling agent.
 - 8. A method for preparing a resin-bonded type magnet which comprises effecting compression molding

of the magnetic powder material according to Claim 1.

- 9. A method for preparing a resin-bonded type magnet which comprises effecting compression molding of the magnetic powder material according to Claim 1 under the condition of applying a magnetic field.
- 10. A method according to Claim 8 or 9, wherein the compression molding is the cold compression molding.
- 11. A method according to Claim 8 or 9, wherein the magnetic powder material is subjected to grinding treatment.
- 12. A method according to Claim 8 or 9, wherein the magnetic powder material is surface treated with 5% by weight or less of a coupling agent.
- 13. A method according to Claim 8 or 9, wherein the magnetic powder material is obtained by any of the methods as claimed in Claims 3 to 6.

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