



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

(43) Date of publication:
28.11.2007 Bulletin 2007/48

(51) Int Cl.:
B22D 7/00 (2006.01) B22D 21/04 (2006.01)

(21) Application number: **06729182.3**

(86) International application number:
PCT/JP2006/305161

(22) Date of filing: **15.03.2006**

(87) International publication number:
WO 2006/098381 (21.09.2006 Gazette 2006/38)

(84) Designated Contracting States:
DE FR GB

(30) Priority: **15.03.2005 JP 2005072308**

(71) Applicants:
• **THE JAPAN STEEL WORKS, LTD.**
Tokyo 100-8456 (JP)
• **HONDA MOTOR CO., LTD.**
Tokyo 107-8556 (JP)

(72) Inventors:
• **AOKI, Yasuhiro**
uroran-shi, Hokkaido, 0518506 (JP)

• **YAMADA, Hitohisa**
uroran-shi, Hokkaido, 0518505 (JP)
• **MURO, Masahiko**
uroran-shi, Hokkaido, 0518505 (JP)
• **IENAGA, Yuuichi**
itama, 3510193 (JP)

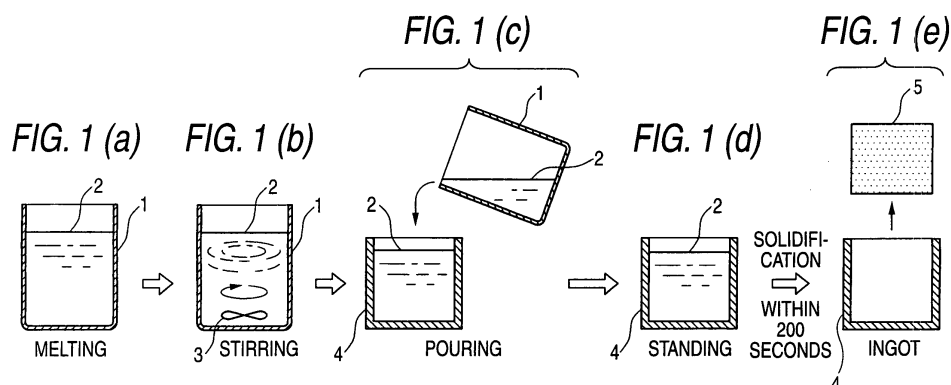
(74) Representative: **Grünecker, Kinkeldey,**
Stockmair & Schwanhäusser
Anwaltssozietät
Maximilianstrasse 58
80538 München (DE)

(54) **PROCESS FOR PRODUCING MAGNESIUM ALLOY**

(57) An object of the present invention is to obtain an ingot containing homogeneous ingredients with suppressing ingredient segregation of Y in the melt-production of an Mg alloy containing Y.

An ingot is formed in a solidification time of not more than 200 seconds after an Mg alloy containing Y is melted. The above solidification is desirably performed after the melt liquid of the Mg alloy is stirred and left standing. The above Mg alloy, for example, contains Y in an amount of 0.5 to 20% by weight. There is an advantage that an Mg alloy having low ingredient segregation and contain-

ing homogeneous ingredients is obtained. Therefore, in the production of alloys whose performance considerably varies depending on the ingredient concentration, such as functional materials, high-quality products can be produced in good yield rates. In order to achieve the above object, specifically, the raw material is melted in a melting furnace and, after melt drop, the melt liquid is sufficiently stirred and left standing. Thereafter, it is cast into a mold designed so that solidification is completed within the above-defined solidification time. Moreover, it is also possible to shorten the solidification time by casting the melt liquid into a sufficiently water-cooled mold.



Description

TECHNICAL FIELD

5 **[0001]** The present invention relates to a process for producing an Mg alloy wherein the Mg alloy containing Y in an amount of 0.5 to 20% by weight is produced with suppressing ingredient segregation.

BACKGROUND ART

10 **[0002]** Since Mg alloys are lightweight and have appropriate strength, they have been increasingly widely used in applications such as automobile parts. The Mg alloys have been produced by batch methods or continuous casting methods wherein melting raw materials are melted in crucibles for melting and cast into molds. For example, Patent Document 1 discloses a method of casting into a sand mold, and Patent Document 2 discloses a casting method using die-casting.

15 Moreover, with regard to the Mg alloys, it is proposed to add various elements in order to improve properties as alloys. For example, Patent Documents 3 and 4 disclose Mg alloys to which rare earth metals such as Y are added.

Patent Document 1: JP-A-H06-279890

Patent Document 2: JP-A-2003-305554

20 Patent Document 3: JP-A-H05-070880

Patent Document 4: JP-A-2004-099941

DISCLOSURE OF THE INVENTION

25 PROBLEMS THAT THE INVENTION IS TO SOLVE

[0003] However, when an Mg alloy containing an element having a large atomic weight, such as Y, is cast in a mold, the Y ingredient precipitates toward a lower part of the mold. Accordingly, there occurs a phenomenon that a difference in concentration of Y between an upper part and a lower part of the resulting ingot becomes large. Recently, in applications using Mg alloys, the requirements in quality are increasingly high, so that an Mg alloy having a deteriorated quality owing to the ingredient segregation of Y cannot satisfy the high requirements in product quality.

[0004] The present invention was developed in consideration of the aforementioned circumstances. An object of the present invention is to provide a process for producing an Mg alloy containing Y, which is capable of obtaining a high-quality Mg product by producing an ingot wherein the ingredients are as homogeneous as possible with suppressing ingredient segregation.

MEANS FOR SOLVING THE PROBLEMS

[0005] That is, the process for producing an Mg alloy according to the invention comprises melting an Mg alloy containing Y and solidifying the molten Mg alloy in a solidification time of not more than 200 seconds so as to obtain an ingot.

[0006] Further, the process for producing an Mg alloy according to the invention comprises stirring an Mg alloy melt liquid containing Y, leaving the Mg alloy melt liquid standing, and subsequently solidifying the Mg alloy melt liquid in a solidification time of not more than 200 seconds so as to obtain the ingot.

45 **[0007]** Still further, the process for producing an Mg alloy according to the invention is the process wherein the Mg alloy contains Y in an amount of 0.5 to 20% by weight.

[0008] Still further, the process for producing an Mg alloy according to the invention is the process wherein the solidification time is 10 seconds or more.

[0009] That is, according to the invention, in the Mg alloy containing Y, solidification is effected before remarkable precipitation of Y to suppress the sedimentation of Y in the ingot production. Thereby, ingredient segregation of Y is prevented to obtain an ingot containing homogeneous ingredient. When the solidification time exceeds 200 seconds on this occasion, a sufficient effect for suppressing the ingredient segregation of Y is not attained and hence quality deterioration owing to the segregation becomes remarkable. When the solidification time is not more than 200 seconds, solidification can be achieved depending on an allowable segregation degree and a segregation ratio can be controlled to 10% or less by adopting a solidification time of not more than 200 seconds. The segregation ratio is represented by the following equation. Incidentally, when comparing the segregation ratios, absolute values thereof are compared.

$$\text{Segregation ratio (\%)} = ((\text{Produced amount}) - (\text{Target content})) / (\text{Target content}) * 100 \cdots (\text{equation})$$

Moreover, when a segregation ratio of 5% or less is intended to obtain, the solidification time is desirably not more than 100 seconds. The solidification time means a period of time from the start of the solidification in a mold until the completion of the solidification. Moreover, before the start of cooling, it is desirable to homogenize the ingredients by stirring a molten Mg alloy melt liquid. The method of stirring the melt liquid is not particularly limited, and a known method such as blade stirring or electromagnetic stirring can be adopted. After stirring of the melt liquid, it is left standing to start the above solidification. In this connection, the completion of the solidification means a time point when a ratio of solid phases reaches 0.67.

[0010] As a means for achieving the above object, for example, a raw material is melted in a melting furnace and the melt liquid is sufficiently stirred after melt drop and left standing, and subsequently is cast into a mold designed so that solidification is completed within the above-defined solidification time. Moreover, it is also possible to shorten the solidification time by casting the liquid into a sufficiently water-cooled mold.

[0011] The content of Y in the above Mg alloy is not limited to a particular amount, but it is desirable that a lower limit of Y is 0.5% by weight and an upper limit is 20% by weight. The incorporation of Y in that range affords improvement in mechanical strength. In contrast, when the content is less than 0.5%, the mechanical strength is not improved. When the content exceeds 20% by weight, embrittlement of the material occurs and also the segregation at casting becomes remarkable. For the same reasons, it is more desirable to control the lower limit to 1% and the upper limit to 15%.

[0012] Moreover, with regard to the above solidification time, the upper limit is defined as above but the lower limit is not restricted to a particular value in the invention. However, for the reason of suppressing production costs when producing large ingots, the solidification time of 10 seconds is desirably defined as the lower limit. When the solidification time is lower than the above lower limit, there occurs a problem of rising production costs.

ADVANTAGE OF THE INVENTION

[0013] As mentioned above, according to the present invention, since an ingot is formed in a solidification time of not more than 200 seconds after an Mg alloy containing Y is melted, there is an advantage that an Mg alloy having low Y segregation and containing homogeneous ingredients is obtained. Therefore, in the production of alloys wherein performance considerably varies depending on ingredient concentration, such as functional materials, high-quality products can be produced in good yield rates.

BRIEF DESCRIPTION OF THE DRAWINGS

[0014]

[FIG. 1] A flow sheet in an embodiment of the invention.

[FIG. 2] A drawing illustrating a melting test apparatus used in Example.

[FIG. 3] A photograph in substitution for drawing of a test material obtained in the solidification test in Example.

[FIG. 4] A graph illustrating a relationship between a solidification rate and deviation of Y content of a test material obtained in the solidification test in Example.

[FIG. 5] A photograph in substitution for drawing of a test material obtained in the solidification test in Example.

[FIG. 6] A graph illustrating a relationship between a solidification rate and deviation of Y content of a test material obtained in the solidification test in Example. [FIG. 7] A drawing illustrating a relationship between mold inner diameter and solidification time in a mold obtained by solidification calculation.

[FIG. 8] A drawing illustrating a relationship between a Y segregation and solidification time in Example of the invention.

DESCRIPTION OF REFERENCE NUMERALS AND SIGNS

[0015]

- 1 melting furnace
- 2 Mg alloy melt liquid
- 3 stirring apparatus

- 4 mold
5 ingot

BEST MODE FOR CARRYING OUT THE INVENTION

[0016] The following will explain an embodiment of the present invention.

The Mg alloy for use in the invention contains at least Y. The suitable content of Y can be represented as 0.5 to 20% by weight. When no other additional element is incorporated, the remainder is Mg and unavoidable impurities. Moreover, the present invention may be one containing the other additional element(s). Examples of the additional elements include, as % by weight, Zn: 0.1 to 10%, Zr: 0.1 to 2%, Al: 0.1 to 10%, Ca: 0.1 to 10%, Mn: 0.1 to 2%, Mm (misch metal): 0.1 to 10%, Sr: 0.001 to 0.1%, Si: 0.1 to 2%, Sn: 0.1 to 10%, Ge: 0.1 to 10%, Ce: 0.1 to 10%, La: 0.1 to 10%, Nd: 0.1 to 10%, Gd: 0.1 to 10%, and the like. The invention is particularly suitable for suppressing ingredient segregation in Mg alloys containing Zn, rare-earth elements, and the like as ingredients.

[0017] An Mg metal whose ingredient is regulated is heated and melted in, for example, a melting furnace 1 as shown in FIG. 1(a) to form an Mg alloy melt liquid 2. The method for melting the Mg alloy is not particularly limited in the invention. For example, it can be performed by a usual method using a known melting furnace. The Mg alloy melt liquid is suitably stirred and mixed. The method and means for stirring and mixing are not particularly limited and suitable ones can be adopted. For example, as shown in FIG. 1(b), stirring of the Mg alloy melt liquid 2 is performed with a stirring blade 3.

[0018] The Mg alloy melt liquid 2 is poured into a mold 4 as shown in FIG. 1(c) and then is usually solidified in the mold 4 in a standing state. On this occasion, depending on the ingredients of the Mg alloy (solidifying point and the like vary depending on the ingredients), temperature of the Mg alloy melt liquid, cooling ability of the mold, a mass effect of the Mg alloy, and the like, the solidification rate of the melt liquid and the time until solidification, i.e., the solidification time are determined. Since the ingredient segregation ratio of Y depends on the solidification time to a large extent, the longest solidification time may be determined according to the desired ingredient segregation ratio. When the segregation ratio is controlled to not more than 10%, the solidification time is determined to be not more than 200 seconds. Moreover, when the segregation ratio is controlled to not more than 5%, the solidification time is determined to be not more than 100 seconds. It is necessary to determine the material and size of the mold, temperature of the melt liquid, presence or absence of forced cooling, method, and the like so that the solidification time falls within such a period of time.

[0019] After these conditions are determined, the Mg alloy melt liquid 2 is solidified within the determined solidification time to obtain an ingot 5. In the ingot, the segregation ratio of Y is not more than the predetermined desired segregation ratio and thus an ingot wherein ingredients are to be homogenized can be obtained. By producing an Mg product using the ingot as a starting material, a product low in ingredient segregation and excellent in quality is obtained. In this connection, the process for obtaining the Mg product using the ingot as a starting material is not particularly limited, and known processing methods and the like can be adopted.

EXAMPLE 1

[0020] The following will describe Examples of the invention.

In order to reproduce the segregation of Y ingredient, a small melting test apparatus 10 as shown in FIG. 2 was provided. In the melting test apparatus 10, a furnace casing 11 being composed of a heat-resistant material and having a cylindrical inner space inside thereof is arranged vertically with the axial center being perpendicular, a tubular heating element 12 is arranged in the furnace casing, and a furnace central tube 13 was concentrically arranged inside the heating element 12. A crucible 14 is arranged as a melting furnace in the furnace central tube 13, and the crucible 14 is placed on a supporting table 13a provided in the furnace tube 13. The temperature of the crucible 14 is elevated by operating the heating element 12 to heat the furnace central tube 13.

[0021] Moreover, upper and lower ends of the furnace central tube 13 are protruded out of the furnace casing 11 and are closed with water-cooled caps 15 and 16. Cooling water-inlet tube 15a and cooling water-outlet tube 15b, and cooling water-inlet tube 16a and cooling water-outlet tube 16b, are provided to the water-cooled caps 15 and 16, respectively. The cooling water introduced from the cooling water-inlet tubes 15a and 16a is passed through the water-cooled caps 15 and 16 and is discharged from the cooling water-outlet tubes 15b and 16b. The cooling water is passed through during heating in order to prevent damage of members in the above water-cooled caps 15 and 16.

[0022] Moreover, an Ar gas-inlet tube 17a is provided on the water-cooled cap 16. The Ar gas-inlet tube 17a communicates with the inside of the furnace central tube 13. Furthermore, an Ar gas-outlet tube 17b is provided on the water-cooled cap 15. The Ar gas-outlet tube 17b also communicates with the inside of the furnace central tube 13.

By introducing Ar gas from the above Ar gas-inlet tube 17a to the inside of the furnace central tube 13, the inside of the furnace central tube 13 can be made an Ar gas atmosphere. Accordingly, oxidation of the Mg alloy in the melting of the alloy in the crucible 14 can be prevented.

Incidentally, the furnace casing 11 includes a furnace-controlling temperature 18 which measures temperature of the

space in the furnace casing, and the water-cooled cap 15 includes a thermocouple 19 which measures temperature of the melt liquid in the crucible 14.

[0023] In the above small melting test apparatus 10, 90 g of an Mg alloy containing Y in an amount of 6.7% was inserted as a host metal into the crucible 14, heated to 750°C to melt, and solidified in the solidification time of 1000 seconds. Then, when the sample was cut in a vertical direction and structural observation (EPMA method) of the cross-sectional surface was performed, a phenomenon that the Y ingredient was thickened at the lower part of the ingot was observed as shown in FIG. 3.

[0024] Further, using the above small melting test apparatus 10, the Y ingredient of ingots of samples solidified in various solidification times was analyzed to investigate correlation between the solidification time and Y segregation. The results are shown in FIG. 4. As shown in FIG. 4, when the solidification time was long, the Y ingredient precipitated toward the lower part by just that much. It had been apparent that the Y ingredient value at the central part of the ingot became lower than the ladle value (melt liquid ingredient value) and the degree of Y segregation tended to be increased. In contrast, when the solidification time was shortened to 10 seconds, it was found that segregation of the Y ingredient was suppressed as shown in FIG. 5.

[0025] Then, using a large melting furnace, a raw material of 167 kg of Mg containing 6.7% of Y (Y: 6.7 wt%, Zr: 4.9 wt%, La: 1.0 wt%, the remainder: Mg) was inserted into a crucible and the melt liquid was cast into a square mold (320x490x440 mm) after melt drop, whereby the relationship between the ingredient segregation of Y in the mold-cast material and the solidification time. FIG. 6 shows the relationship between the ingredient segregation of Y in each part of the inside of the ingot and the solidification time. The solidification time is calculated based on the distance from the bottom of the ingot. Therefore, the solidification time and the distance from the bottom of the ingot correspond to each other one-to-one. Also in the mold-cast material, since the Y ingredient precipitated toward the lower part of the ingot as the solidification time increases, there was a tendency that the Y concentration increased in the position of 20 mm from the bottom of the ingot, whereas the Y ingredient value in the sampling position of the upper part of the ingot decreased. The decreasing ratio was found to be 5% at 100 seconds, 10% at 200 seconds, and 15% at 400 seconds.

[0026] Then, in order to produce an ingot wherein the ingredient segregation of Y was suppressed to not more than 10%, the solidification time was assessed and a mold was designed so that the solidification time fell within 200 seconds. The solidification time of a cylindrical mold of a plate having a thickness of 20 mm correlated to the inner diameter based on the solidification calculation. From FIG. 7, it was needed that the diameter should be 230 mm or less as an air-cooled mold. Based on the result, a mold having a diameter of 200 mm was prepared.

Using the large melting furnace, a raw material of the Mg alloy containing 6.7% of Y was inserted into the crucible and about 60 kg of the melt liquid was cast into the cylindrical mold (ϕ 200 mm×H 650 mm) after melt drop. FIG. 8 shows the relationship between the ingredient segregation of Y in each part of the inside of the ingot and the solidification time. The ingredient segregation of Y was suppressed to not more than 10% and a homogeneous ingot could be produced.

[0027] In the production of an ingot wherein the ingredient segregation of Y is suppressed to not more than 10%, in order to use a mold having a larger inner diameter than in the case of the above Example, there is provided a water-cooled mold wherein a water-cooling pipe is welded to the side wall of the mold and water is passed therethrough to achieve sufficient cooling of the mold. From the calculation result shown in FIG. 7, it was found that the inner diameter of the cylindrical mold capable of realizing the solidification time of 200 seconds was extended to 350 mm or less. Using the large melting furnace, a raw material of the Mg alloy containing 6.7% of Y was inserted into the crucible and about 180 kg of the melt liquid was cast into the cylindrical mold having a size of ϕ 350 mm×H 650 mm after melt drop. The ingredient segregation of Y was suppressed to not more than 10% and a homogeneous ingot could be produced.

While the invention has been described in detail and with reference to specific embodiments thereof, it will be apparent to one skilled in the art that various changes and modifications can be made therein without departing from the spirit and scope thereof.

The present application is based on Japanese Patent Application No. 2005-072308 filed on March 15, 2005, and the contents are incorporated herein by reference.

INDUSTRIAL APPLICABILITY

[0028] In the present invention, since an ingot is formed in a solidification time of not more than 200 seconds after an Mg alloy containing Y is melted, an Mg alloy having low Y segregation and containing homogeneous ingredients is obtained. Thereby, in the production of alloys whose performance considerably varies depending on the ingredient concentration, such as functional materials, high-quality products can be produced in good yield rates.

Claims

1. A process for producing an Mg alloy comprising:

EP 1 859 878 A1

melting an Mg alloy containing Y; and

solidifying the molten Mg alloy in a solidification time of not more than 200 seconds so as to obtain an ingot.

2. The process for producing an Mg alloy according to claim 1, further comprising:

5 stirring an Mg alloy melt liquid containing Y; leaving the Mg alloy melt liquid standing; and
 solidifying the Mg alloy melt liquid in a solidification time of not more than 200 seconds so as to obtain the ingot.

- 10 3. The process for producing an Mg alloy according to claim 1 or 2,
 wherein the Mg alloy contains Y in an amount of 0.5 to 20% by weight.

4. The process for producing an Mg alloy according to any one of claims 1 to 3,
 wherein the solidification time is 10 seconds or more.

15

20

25

30

35

40

45

50

55

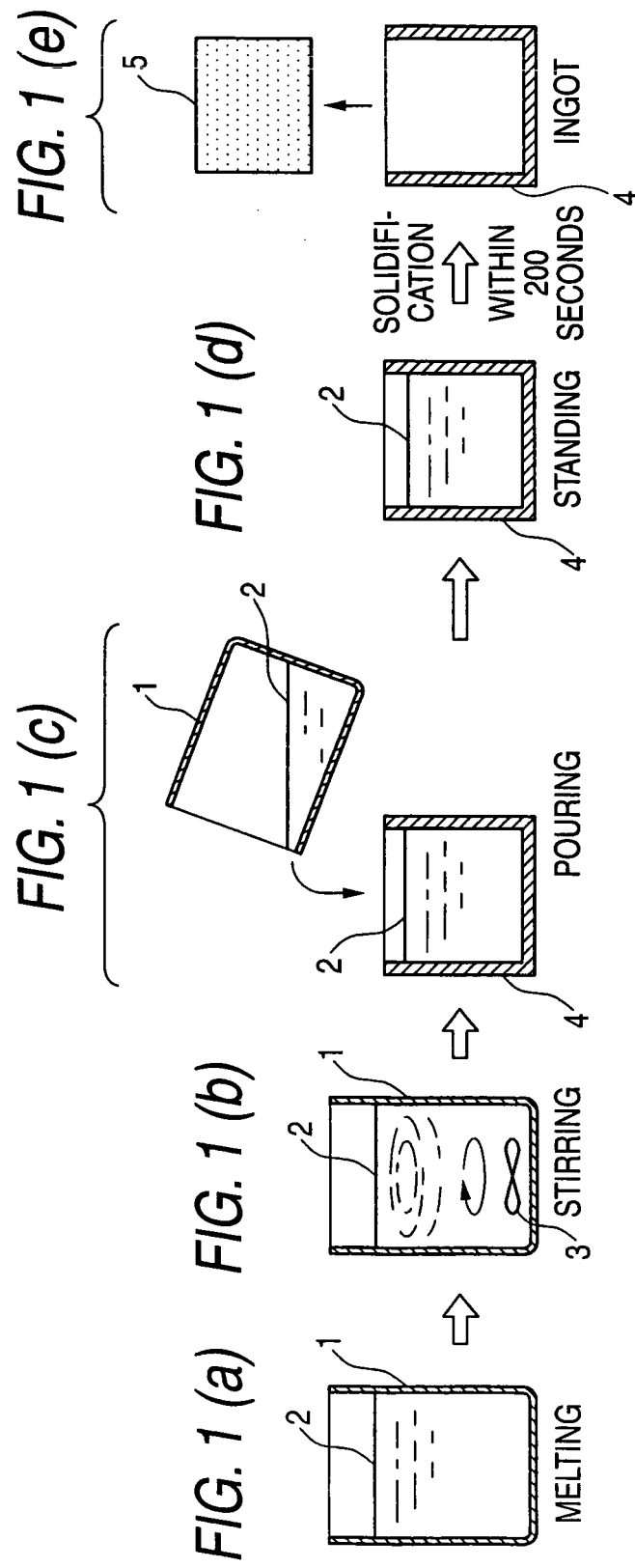


FIG. 2

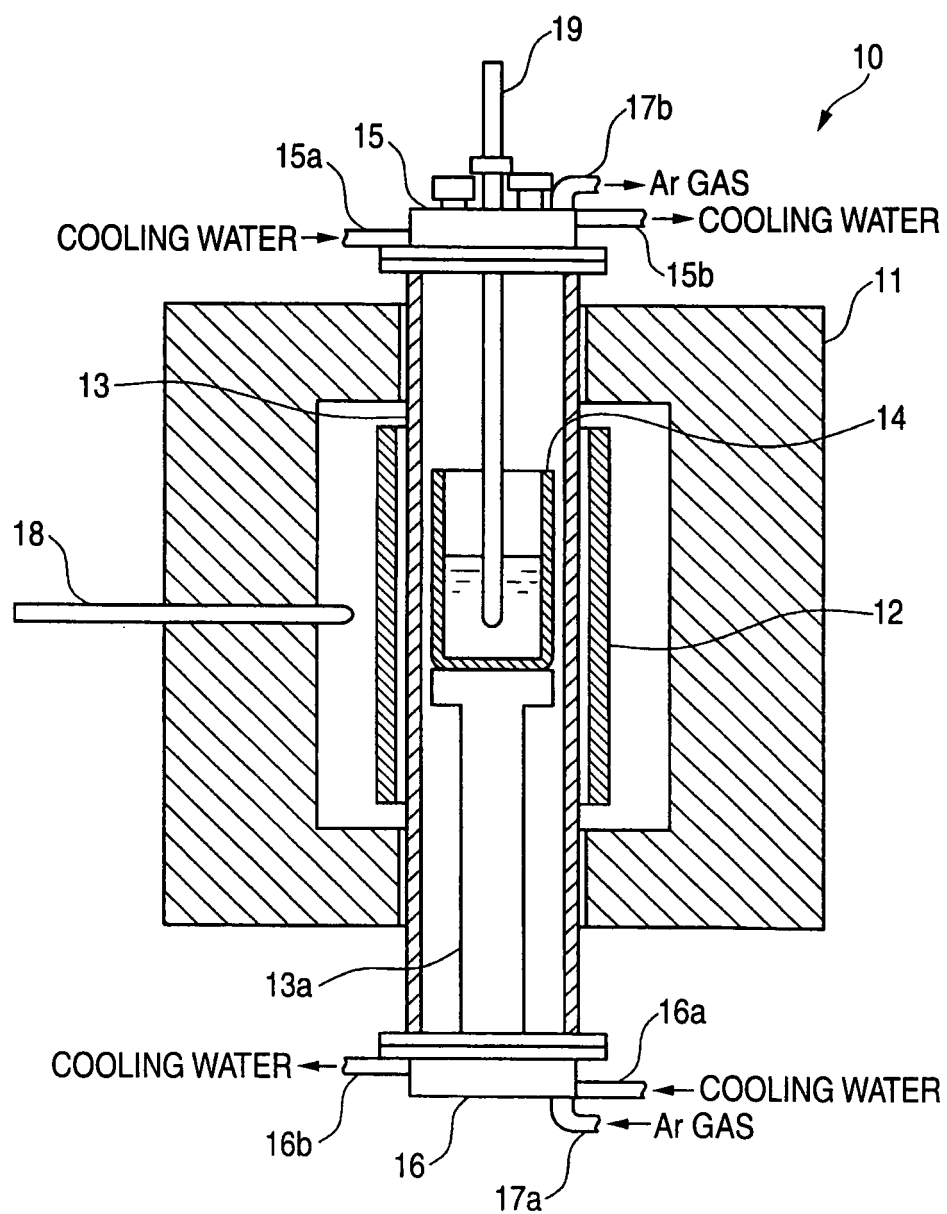
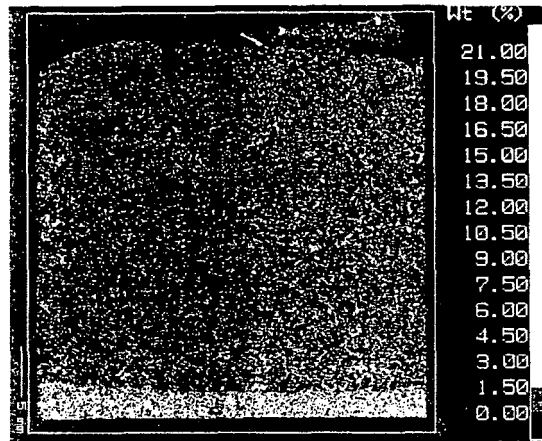


FIG. 3



PHOTOGRAPH IN SUBSTITUTION
FOR DRAWING

FIG. 4

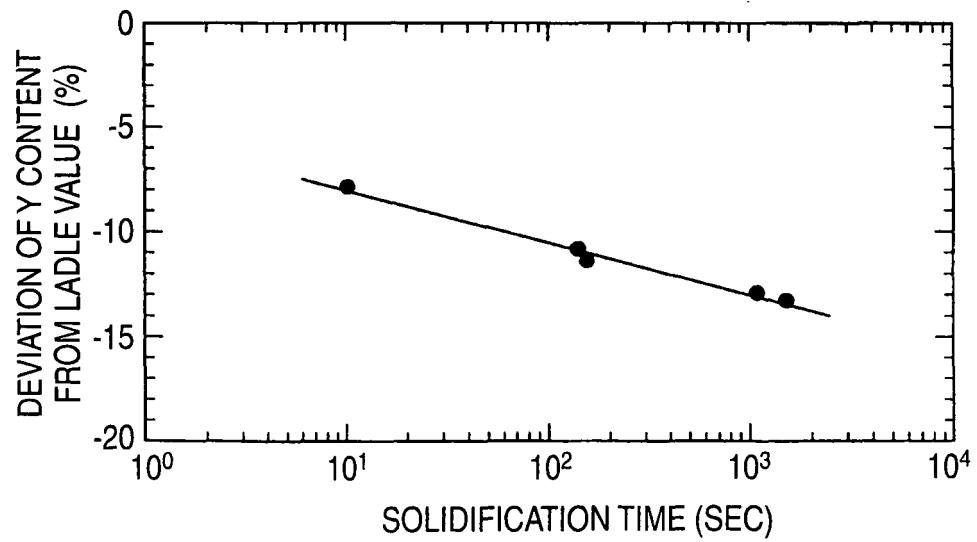


FIG. 5

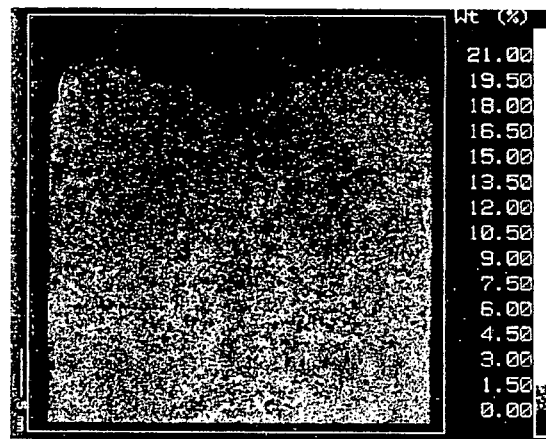
PHOTOGRAPH IN SUBSTITUTION
FOR DRAWING

FIG. 6

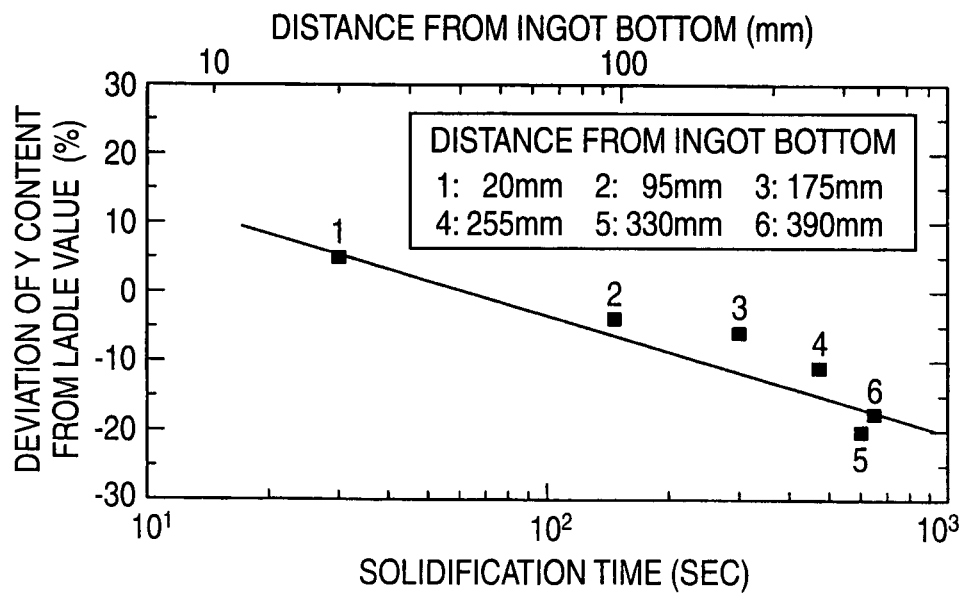


FIG. 7

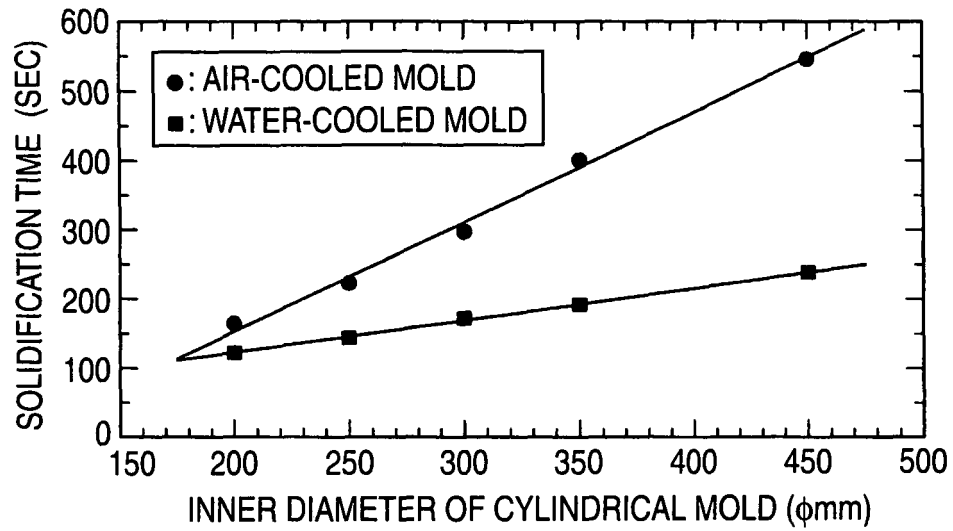
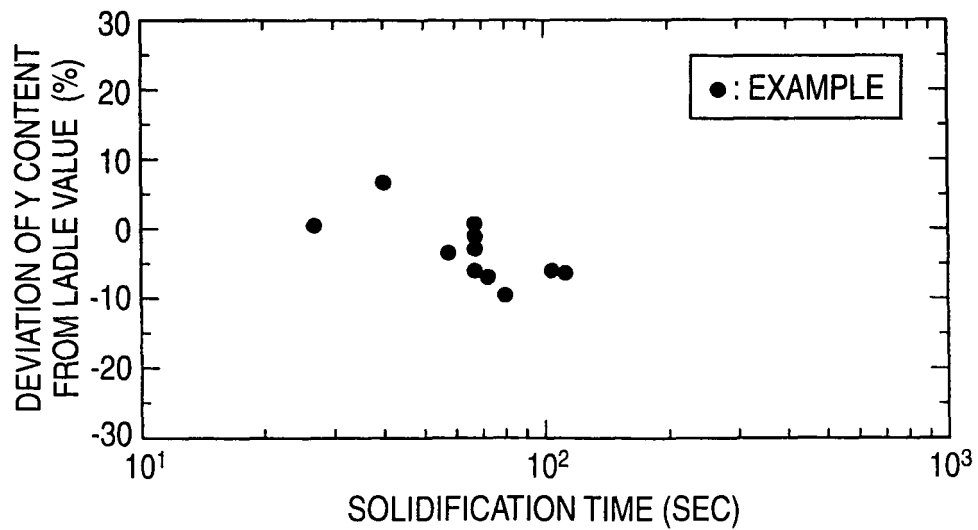


FIG. 8



INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2006/305161

A. CLASSIFICATION OF SUBJECT MATTER B22D7/00 (2006.01) , B22D21/04 (2006.01)		
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) B22D7/00 (2006.01) , B22D21/04 (2006.01)		
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 1996-2006 Kokai Jitsuyo Shinan Koho 1971-2006 Toroku Jitsuyo Shinan Koho 1994-2006		
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.
Y	JP 2003-126943 A (Nippon Kinzoku Co., Ltd.) , 08 May, 2003 (08.05.03) , Par. No. [0013] (Family: none)	1-4
Y	JP 05-070880 A (Ken MASUMOTO) , 23 March, 1993 (23.03.93) , Par. Nos. [0014] , [0015] (Family: none)	1-4
☐ 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 13 June, 2006 (13.06.06)		Date of mailing of the international search report 20 June, 2006 (20.06.06)
Name and mailing address of the ISA/ Japanese Patent Office		Authorized officer
Facsimile No.		Telephone No.

Form PCT/ISA/210 (second sheet) (April 2005)

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 H06279890 A [0002]
- JP 2003305554 A [0002]
- JP H05070880 A [0002]
- JP 2004099941 A [0002]
- JP 2005072308 A [0027]