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(12) **United States Patent**
Nakamura et al.(10) **Patent No.:** **US 8,377,233 B2**
(45) **Date of Patent:** ***Feb. 19, 2013**(54) **PREPARATION OF RARE EARTH
PERMANENT MAGNET MATERIAL**(75) Inventors: **Hajime Nakamura**, Echizen (JP);
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claimer.(21) Appl. No.: **13/033,943**(22) Filed: **Feb. 24, 2011**(65) **Prior Publication Data**

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See application file for complete search history.

(56) **References Cited****U.S. PATENT DOCUMENTS**4,859,255 A * 8/1989 Fujimura et al. 148/302
4,933,009 A * 6/1990 Ghandehari 75/252
4,952,252 A * 8/1990 Ghandehari 148/105
4,954,186 A * 9/1990 Ghandehari 148/302
5,000,800 A * 3/1991 Sagawa 148/302
5,034,146 A 7/1991 Ohashi et al.
5,194,099 A 3/1993 Esper et al.
5,250,255 A 10/1993 Sagawa et al.
5,286,366 A * 2/1994 Mitsuji 205/176
5,405,455 A 4/1995 Kusunoki et al.
5,411,603 A 5/1995 Vial et al.
5,766,372 A 6/1998 Fujimura et al.
5,858,124 A 1/1999 Endo et al.6,296,720 B1 10/2001 Yamamoto et al.
6,302,939 B1 10/2001 Rabin et al.
6,606,019 B1 8/2003 Ohashi
6,777,097 B2 * 8/2004 Hamada et al. 428/469
6,960,240 B2 11/2005 Hirota et al.
7,053,745 B2 5/2006 Yoshimura et al.
7,163,591 B2 * 1/2007 Kim et al. 148/101
2004/0187970 A1 9/2004 Ishizaka et al.
2006/0278517 A1 12/2006 Machida et al.
2007/0017601 A1 1/2007 Miyata et al.
2007/0034299 A1 2/2007 Machida et al.
2007/0240789 A1 10/2007 Nakamura et al.
2008/0223489 A1 9/2008 Nagata et al.
2008/0245442 A1 10/2008 Nakamura et al.
2008/0247898 A1 10/2008 Nakamura et al.
2009/0098006 A1 4/2009 Nakamura et al.
2009/0226339 A1 9/2009 Nakamura et al.**FOREIGN PATENT DOCUMENTS**EP 0 255 939 A2 2/1988
EP 1 643 513 A1 4/2006
EP 1 705 668 A2 9/2006
EP 1 705 669 A2 9/2006
EP 1 705 670 A2 9/2006
EP 1 705 671 A2 9/2006
EP 1 830 371 A1 9/2007
JP 61-195954 A 8/1986
JP 62-256412 A 11/1987
JP 1-117303 A 5/1989
JP 1-155603 A 6/1989
JP 1-251704 A 10/1989
JP 3-188241 A 8/1991
JP 4-184901 A 7/1992
JP 4-328204 A 11/1992
JP 4-328804 A 11/1992
JP 5-021218 A 1/1993

(Continued)

OTHER PUBLICATIONSExtended European Search Report mailing date of Jan. 14, 2008,
issued in corresponding European Patent Application No. 06250542.
5.

(Continued)

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Daniels & Adrian, LLP(57) **ABSTRACT**A method for preparing a rare earth permanent magnet mate-
rial comprises the steps of: disposing a powder comprising
one or more members selected from an oxide of R², a fluoride
of R³, and an oxyfluoride of R⁴ wherein R², R³ and R⁴ each
are one or more elements selected from among rare earth
elements inclusive of Y and Sc on a sintered magnet form of
a R¹—Fe—B composition wherein R¹ is one or more ele-
ments selected from among rare earth elements inclusive of Y
and Sc, and then heat treating the magnet form and the powder
at a temperature equal to or below the sintering temperature of
the magnet in vacuum or in an inert gas. The result high
performance, compact or thin permanent magnet has a high
remanence and coercivity at a high productivity.**16 Claims, 3 Drawing Sheets**

FOREIGN PATENT DOCUMENTS

JP	5-31807 B	5/1993
JP	6-158238 A	6/1994
JP	6-244011 A	9/1994
JP	3143156 B2	3/2001
JP	2003-282312 A	10/2003
JP	3471876 B2	12/2003
JP	2004-296973 A	10/2004
JP	2004-304038 A	10/2004
JP	2005-11973 A	1/2005
JP	2005-285861 A	10/2005
JP	2007-53351 A	3/2007
RU	2 136 068 C1	8/1999
SU	1 513 738 A1	4/1995
WO	2004/114333 A1	12/2004
WO	2005/123974 A1	12/2005
WO	2006/003882 A1	1/2006
WO	2006/043348 A1	4/2006
WO	2007/119551 A1	10/2007

OTHER PUBLICATIONS

European Search Report mailing date of Mar. 31, 2008, issued in corresponding European Patent Application No. 07254503.

European Search Report mailing date of Jun. 4, 2008, issued in corresponding European Patent Application No. 07251603.2.

European Search Report mailing date of Jun. 26 2008, issued in corresponding European Patent Application No. 08250927.4.

International Search Report of PCT/JP2005/005134, mailing date of Jul. 12, 2005.

International Search Report of PCT/JP2007/056586, mailing date of Jun. 19, 2007.

International Search Report of PCT/JP2007/056594, mailing date of Jul. 10, 2007.

Russian Decision on Grant dated Mar. 18, 2009, issued in corresponding Russian Patent Application No. 2006 117529/02.

Translation of the Preliminary Report of Patentability mailed May 3, 2007 of International Application No. PCT/JP2005/005134.

2005 BM Symposium, abstract of presentation by The Japan Association of Bonded Magnet Industries held on Dec. 2, 2005.

S.P. Beeby et al., "Micromachined Silicon Generator for Harvesting Power From Vibrations", 4th Int'l Workshop on Micro and Nanotechnology for Power Generation and Energy Conversion Application (POWERMEMS 2004), Kyoto, Japan.

K. D. Durst et al., "The Coercive Field of Sintered and Melt-Spun NdFeB Magnets", Journal of Magnetism and Magnetic Materials 68 (1987), pp. 63-75.

R.W. Gao et al., "Effects of the Degree of Grain Alignment on the Hard Magnetic Properties of Sintered NdFeB Magnets", Applied Physics A, 67, Jan. 1998, pp. 353-356.

K. Hirota et al., "Coercivity Enhancement by the Grain Boundary Diffusion Process to Nd-Fe-B Sintered Magnets", IEEE Transactions on Magnetics, Oct. 2006, pp. 2909-2911, vol. 42, No. 10.

K. Hirota et al., "Coercivity Enhancement by Grain Boundary Diffusion Process to Nd-Fe-B Sintered Magnets", IEEE Int'l Magnetics Conference 2006—Technical Digest, May 8-12, 2006, p. 910.

K. Hirota et al., "Production of Nd-Fe-B Sintered Magnet with Higher Coercive Force by Grain Boundary Diffusion", Abstracts of Autumn Meeting of Japan Society of Powder and Powder Metallurgy, (held) Nov. 14-16, 2005, p. 143.

D.H. H Hwang et al., "Development of High Coercive Powder from the Nd-Fe-B Sintered Magnet Scrap", IEEE Transactions on Magnetics, Jul. 2004, vol. 40, No. 4, pp. 2877-2879, New York, NY (USA).

K. Machida et al., "Grain Boundary Modification and Magnetic Properties of Nd-Fe-B Sintered Magnets", Proceedings of the 2004 Spring Meeting of Japan Powder and Powder Metallurgy Society, 2004, 1-47A, pp. 202.

H. Nakamura et al., "Magnetic Properties of Extremely Small Nd-Fe-B Sintered Magnets", IEEE Transactions on Magnetics, Oct. 2005, pp. 3844-3846, vol. 41, No. 10.

H. Nakamura et al., "Magnetic Properties of Extremely Small ND-Fe-B Sintered Magnet", InterMag Asia 2005: Digest of the IEEE Int'l Magnetics Conference, (held) Apr. 4-8, 2005, p. 476.

H. Nakamura, "Magnetic Properties of Miniature Nd-Fe-B Sintered Magnets", The Journal of the Institute of Electrical Engineers of Japan, Nov. 1, 2004, pp. 699-702, vol. 124, No. 11.

H. Nakamura et al., "Microstructures of High Coercivity Nd-Fe-B Sintered Magnets Produced by the Grain Boundary Diffusion Process", Digests of the 30th Annual Conference on Magnetics in Japan, (held) Nov. 11-14, 2006, pp. 417-418.

H. Nakamura, "Nd-Fe-B Sintered Magnets Produced by the Grain Boundary Diffusion Process", Bulletin of Topical Symposium of the Magnetic Society of Japan, Mar. 14, 2006, pp. 13-18.

Nikkei Net, press release, Mar. 24, 2005.

K.T. Park et al., "Effect of Metal-Coating and Consecutive Heat Treatment on Coercivity of Thin Nd-Fe-B Sintered Magnets", Proceedings of the 16th Int'l Workshop on Rare-Earth Magnets and Their Applications, Sendai, (2000), pp. 257-264.

C.D. Qin et al., "The Protective Coatings of NdFeB Magnets by Al and Al(Fe)", Journal of Applied Physics, American Institute of Physics, Apr. 15, 1996, pp. 4854-4856, vol. 79, No. 8.

K. Sato, Techno-Frontier Symposium 2005, (held) Apr. 20, 2005 by JMA, pp. B1-2-1 to B1-2-12.

"Shin-Etsu Chemical develops new high-performance technology for neodymium rare-earth magnets", Shin-Etsu News, press release in Tokyo, Mar. 24, 2005.

The Dempa Shinbun, press release, Mar. 25, 2005.

The Kagaku Kogyo Nippo (The Chemical Daily), press release, Mar. 25, 2005.

The Nikkan Chemical News, press release, Mar. 25, 2005.

The Nikkan Kogyo Shimbun, press release, Mar. 25, 2005.

The Sekiyu Kagaku Shimbun Nikkan Tsushin, press release, Mar. 25, 2005.

U.S. Appl. No. 11/916,498, filed Dec. 4, 2007 entitled, "Method for Preparing Rare Earth Permanent Magnet Material".

* cited by examiner