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Anisotropic Mechanical Properties of $Pr(Co,In)_5$-type Compounds and Their Relation to Texture Formation in Die-upset Magnets

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Die-upset magnets from a mechanically-milled $Pr(Co,In)_5$-type alloy are known to have a peculiar texture; the easy magnetization axis (*c*-axis) is perpendicular to the pressing direction. This peculiar texture is thought to be linked closely to the anisotropic mechanical properties of $Pr(Co,In)_5$-type hexagonal compounds. The hardness of the $Pr(Co,In)_5$-type crystal was measured using selectively grown grains in an annealed $Pr_{17}Co_{82}In_1$ alloy button, and the crystallographic orientation was determined by observing the magnetic domain image. The hardness (549 VHN) on the plane with a 'cogwheel'-type domain image was significantly higher than that (510 VHN) on the plane with a 'cigar'-type domain image, indicating that the inter-layer bonding force between the (000l) basal planes is stronger than that between the (*hki*0) planes. This suggests that the most probable slip plane is the (*hki*0) plane parallel to the *c*-axis. During die-upsetting of the $Pr(Co,In)_5$-type alloys the deformation proceeds by (*hki*0) plane slip, and the *c*-axis rotates to ultimately become oriented perpendicular to the pressing direction. It is proposed that the peculiar texture in the die-upset $Pr(Co,In)_5$-type magnets is probably developed by slip deformation of the (*hki*0) plane of the $Pr(Co,In)_5$-type grains.

Keywords : $Pr(Co,In)_5$, die-upset, texture, slip deformation

1. Introduction

The alignment of hard magnetic grains (texture) is of utmost importance for the high performance of a permanent magnet. The hard magnetic grain texture in some rare-earth-transition metal magnetic alloys is commonly achieved by the die-upset technique [1-5]. It is known that die-upset magnets from a mechanically-milled $Pr(Co,In)_5$-type alloy have a peculiar texture; the easy magnetization axis (EMA) is perpendicular to the pressing direction (PD) [4,5]. This texture is quite different from that in die-upset Nd-Fe-B-type alloys, in which the EMA is parallel to the PD [6,7] (Fig. 1). The texture in die-upset $Pr(Co,In)_5$-type magnets is thought to have developed by a different mechanism from that (stress-induced preferential grain growth *via* dissolution and precipitation [6,7]) operating in the standard melt-spun Nd-Fe-B-type alloy, and is thought to be linked closely to the anisotropic mechanical properties of $Pr(Co,In)_5$-type hexagonal compounds. In the present study, the mechanical hardness of the $Pr_{17}Co_{82}In_1$ crystal was measured along different crystallographic directions, and the results were co-related to texture formation in die-upset $Pr(Co,In)_5$-type magnets.

2. Experimental Work

A $Pr(Co,In)_5$-type alloy button with the chemical composition $Pr_{17}Co_{82}In_1$ was prepared by arc-melting of the constituent elements, and the prepared button was homo-

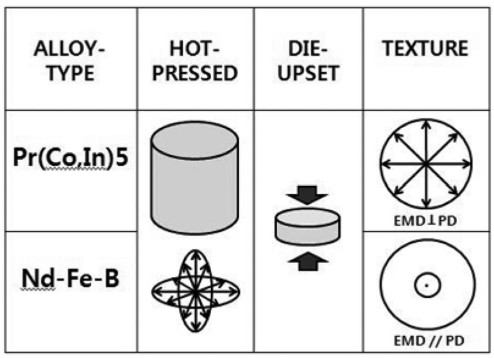

Fig. 1. Comparison of the textures induced in the die-upset $Pr(Co,In)_5$-type and Nd-Fe-B-type magnets.

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genized at 1050 °C for 3 days. Some of the Pr(Co,In)$_5$-type matrix grains in the annealed alloy were grown to sizes markedly over 500 μm in diameter. The mechanical hardness of the Pr(Co,In)$_5$-type crystal was measured by a Vickers micro-hardness tester (load = 100 g) using selectively grown grains in the annealed alloy. The pyramidal and pointed indenter was pressed on the polished flat surface of the grains. The crystallographic orientation of the Pr(Co,In)$_5$-type crystal was determined by observing the magnetic domain image of the polished flat surface of individual grains using Bitter solution. The magnetic domain structure on the specific plane of a crystal with uniaxial magnetocrystalline anisotropy shows a unique image depending on the crystallographic orientation. The Pr(Co,In)$_5$-type crystal has a hexagonal structure belonging to the CaCu$_5$-type (the *P*6/*mmm* space group), and its *c*-axis is the EMA. Therefore, the plane with the so-called 'cogwheel'-type domain image is the basal plane of the hexagonal crystal which is indexed as (000l) and is perpendicular to the EMA. Meanwhile, the plane with the 'cigar'-type image is the plane parallel to the EMA (*c*-axis) and indexed as (*hki*0). The mechanical hardness on the plane with the 'cogwheel'- and 'cigar'-type domain images shows the hardness on the basal plane perpendicular to the EMA (*c*-axis) and on the plane parallel to the *c*-axis, respectively. The average hardness value on a particular plane was obtained from approximately 200 indentations made on over 10 Pr(Co,In)$_5$-type crystal grains. Magnetic phase analysis of the alloy was performed by a magnetic balance-type thermo-magnetic analyser (TMA). The magnetic field applied to the sample in the TMA was approximately 400 Oe. Differential thermal analysis (DTA) of the alloy was carried out in order to see a phase change during heating. TMA and DTA were performed in Ar gas with a heating rate of 4 °C/min.

3. Results and Discussion

Fig. 2(a) shows the phase analysis results for the homogenized Pr$_{17}$Co$_{82}$In$_1$ alloy by TMA. There seem to be three magnetic transitions at 428 °C, 648 °C, and 908 °C, which correspond to the Curie temperatures of the Pr$_5$(Co,In)$_{19}$-type [10], Pr(Co,In)$_5$-type [11,12], and Pr$_2$(Co,In)$_{17}$-type phases [13], respectively. The homogenized Pr$_{17}$Co$_{82}$In$_1$ alloy consists mainly of the Pr(Co,In)$_5$-type phase with a minor phase of Pr$_2$(Co,In)$_{17}$-type. A trace of the Pr$_5$(Co,In)$_{19}$-type phase was also present in the alloy. It was noted that the Curie temperature (648 °C) of the matrix PrCo$_5$-type phase in the present study was higher than that (612-639 °C) of the stoichiometric PrCo$_5$ phase previously reported. This was attributed to the solu-

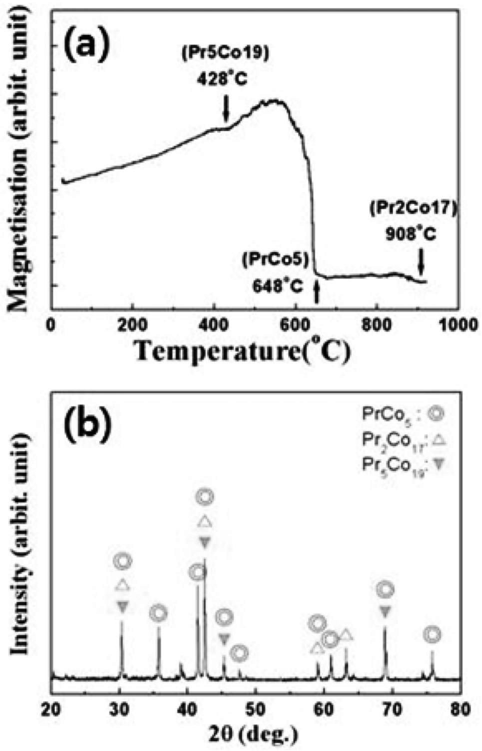

Fig. 2. Phase analysis results of the homogenized Pr$_{17}$Co$_{82}$In$_1$ alloy by (a) TMA and (b) XRD.

tion of the In into the PrCo$_5$ phase. This phase constitution was also confirmed by x-ray diffraction (XRD), as shown in Fig. 2(b). There was an unidentified peak at around 39°, which could not be indexed to a specific phase. The appearance of this unidentified phase was thought to be closely related to the addition of indium [14]. Fig. 3 shows the DTA results of the Pr$_{17}$Co$_{82}$In$_1$ alloy performance up to 950 °C which is the commonly used die-upset temperature for the die-upset Pr(Co,In)$_5$-type magnet. The DTA results showed no thermal event in the course of heating, indicating the absence of a low melting point phase. It is less likely, therefore, that the texture formation mechanism (stress-induced preferential grain growth *via* dissolution and precipitation) commonly operating in the die-upset Nd-rich Nd-Fe-B alloy containing a low melting point grain boundary phase may operate in this particular Pr(Co,In)$_5$-type alloy without a low melting point phase. The possible texture formation mechanism for the present die-upset Pr(Co,In)$_5$-type alloy would be slip deformation of the crystal along a specific plane [8,9]. This mechanism is known to operate in the Nd-lean Nd-Fe-B alloy, which contains little of the low melting point liquid grain boundary phases. As the present Pr(Co,In)$_5$-type alloy also contains no low melting point phase, it is worth discussing the likelihood that the

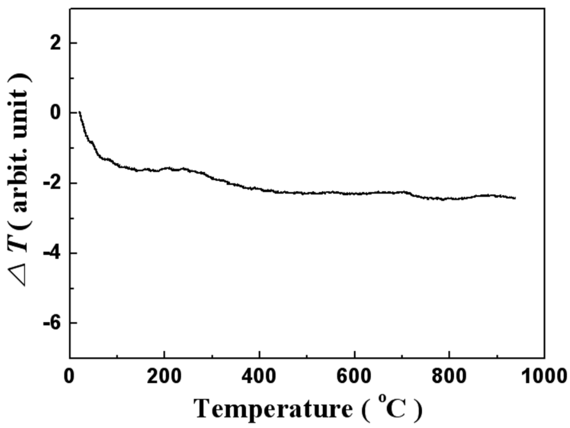

Fig. 3. DTA results for the $Pr_{17}Co_{82}In_1$ alloy.

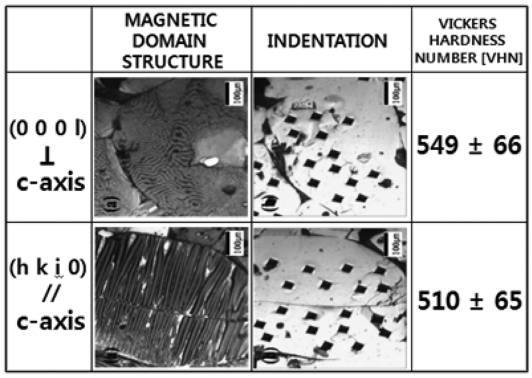

Fig. 4. Magnetic domain image and mechanical hardness values for the different crystallographic planes of the Pr(Co,In)$_5$-type crystal in the $Pr_{17}Co_{82}In_1$ alloy.

slip deformation mechanism may operate in the present alloy.

It is generally understood that in metal alloys a slip occurs most readily on the crystallographic planes of the greatest atomic density along the close-packed direction within the slip planes. In the hexagonal Pr(Co,In)$_5$-type alloy, the most probable slip system would be the combination of the (0001) basal plane and the <11-20> diagonal directions. If this slip system works during the die-upsetting of the Pr(Co,In)$_5$-type alloy, a texture with the *c*-axis being parallel to the pressing direction would develop. However, the texture in the die-upset Pr(Co,In)$_5$-type alloy is known to be different from this one [4,5]: in the die-upset Pr(Co,In)$_5$-type alloy, the easy magnetization axis (EMA = *c*-axis) is perpendicular to the pressing direction. Considering the harsh deformation conditions of die-upsetting [high temperature (> 900 °C) and extremely high strain rate ($> 5.8 \times 10^{-3}$/s)], it is acceptable that the deformation during the die-upsetting may not purely obey the classical slip system. In an effort to understand the texture formation mechanism in the die-upset Pr(Co,In)$_5$-type alloy, the present authors took notice of an anisotropic mechanical property of the alloy crystal. In a metallurgical practice, the most commonly used technique for the measurement of the mechanical properties of a metal crystal is the hardness test. For a given metal alloy the mechanical hardness is a measure of its resistance to plastic deformation by indentation, and is also a qualitative indication of its strength. In the present study, the anisotropic mechanical properties of the Pr(Co,In)$_5$-type crystal were examined by measuring the mechanical hardness. Fig. 4 shows the magnetic domain image, indentations and mechanical hardness values on the different crystallographic planes of the Pr(Co,In)$_5$-type crystal in $Pr_{17}Co_{82}In_1$ alloy. The domain observation and the indentations were made on the same crystal in the alloy. It can be seen that the mechanical hardness (549 VHN) on the plane with the 'cogwheel'-type domain image was significantly higher than that (510 VHN) on the plane with the 'cigar'-type domain image, indicating that the Pr(Co,In)$_5$-type alloy crystal has anisotropic mechanical properties. The relative hardness difference (39/549 × 100%) was around 7%. Considering that the small relative Young's modulus difference (around 2% at room temperature [15]) of the $Nd_2Fe_{14}B$ compound leads to good texture in the die-upset Nd-Fe-B type alloy, the hardness anisotropy of the Pr(Co,In)$_5$ compound in the present study is thought to be not insignificant. In order to assist the understanding of the relationship between texture formation and anisotropic mechanical properties, the material flow during the plastic deformation by an indentation needs to be understood. As the indenter of the Vickers micro-hardness tester is made of pyramidal diamond and is pointed, when a pyramid is pressed against a flat surface of the crystal a plastically deformed zone is formed mostly in the lateral region of the indenter rather than in the region beneath the indenter. According to the plastic-elastic model of indentation formation [16,17], the material displaced by the indenter is completely accounted for by the decrease in volume of the elastic material, as shown in Fig. 5. This removes the need for an upward flow of material around the indenter. It is generally accepted that little upward flow in the vicinity of the indentation is observed in a practical hardness measurement. This suggests that most of the deformation in the plastic zone is caused by slip of the atomic layers which are perpendicular to the pressing direction of the indentation. Therefore, the higher mechanical hardness on the plane with the 'cogwheel' domain image indicates that the inter-layer bonding force between the (000l) basal planes is stronger than that between the (*hki*0) planes. This suggests that the most probable slip

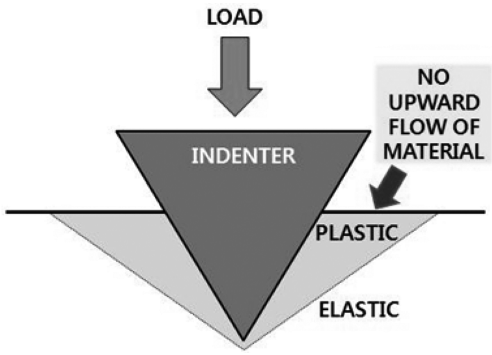

Fig. 5. Plastic zone under the Vickers indenter. No upward flow of material around the indenter suggests that the deformation in the plastic zone is caused by slip of the atomic layers which are perpendicular to the pressing direction of the indentation.

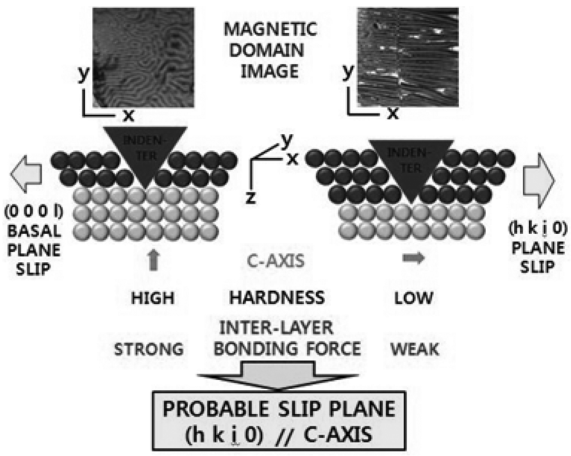

Fig. 6. Reasoning behind the decision regarding the probable slip plane in the Pr(Co,In)$_5$-type crystal based on the mechanical hardness measurements.

plane during plastic deformation of the Pr(Co,In)$_5$ compound may be the (*hki*0) plane parallel to the *c*-axis. Fig. 6 shows the reasoning behind the decision of the most probable slip plane in the Pr(Co,In)$_5$-type crystal based on the mechanical hardness measurement results.

Assuming that the anisotropic nature of the mechanical properties is still maintained at elevated temperature for the die-upsetting, when the Pr(Co,In)$_5$-type crystals are under uniaxial compression load during die-upsetting a shear stress is produced on particular geometric planes in the crystal in particular directions, and hence a shear strain is produced in the crystal. The magnitude of the resolved shear stress depends on the angle between the direction of the slip plane and the compression axis. As the most probable slip plane in the Pr(Co,In)$_5$-type grain is the (*hki*0) plane parallel to the *c*-axis, a plastic deformation of the Pr(Co,In)$_5$-type grain will occur through slippage along the (*hki*0) plane when the magnitude of the resolved shear stresses along the (*hki*0) plane during the die-upset reaches a critical value. As the deformation increases the (*hki*0) planes will rotate in such a manner that they tend to align perpendicular to the pressing direction, and at the same time the magnetization easy *c*-axis tends to orient perpendicular to the pressing direction. Thus, the Pr(Co,In)$_5$ grain texture, in which the magnetization easy *c*-axis orients perpendicular to the pressing direction, is developed. This texture formation mechanism is shown schematically in Fig. 7. It is proposed, therefore, that the peculiar grain texture in the die-upset Pr(Co,In)$_5$-type magnet, in which the magnetization easy *c*-axis orients perpendicular to the pressing direction, is developed *via* slip deformation of the (*hki*0) planes.

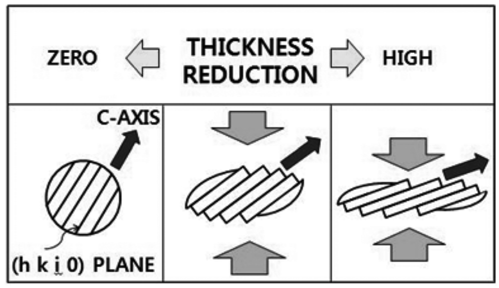

Fig. 7. Schematic of the proposed texture formation mechanism in the die-upset Pr(Co,In)$_5$-type magnet.

4. Conclusion

The Pr(Co,In)$_5$-type grain in the $Pr_{17}Co_{82}In_1$ alloy has anisotropic mechanical properties. The mechanical hardness on the basal plane of the hexagonal grain was higher than that on the plane parallel to the magnetization easy *c*-axis, this indicated that the inter-layer bonding force between the (000l) basal planes was stronger than that between the (*hki*0) planes. This suggested that the most probable slip plane during plastic deformation of the Pr(Co,In)$_5$ crystal may be the (*hki*0) plane parallel to the *c*-axis. It was proposed that the peculiar Pr(Co,In)$_5$ grain texture in a die-upset Pr(Co,In)$_5$-type alloy, in which the magnetization easy *c*-axis orients perpendicular to the pressing direction, is developed *via* slip deformation of the crystal along the (*hki*0) planes.

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