

## Effect of Cu-Addition and Die-Upset Temperature on Texture in Die-Upset Nd-Lean Nd-Fe-B Alloys

H. W. Kwon<sup>1\*</sup> and J. H. Yu<sup>2</sup>

<sup>1</sup>*Pukyong National University, Busan 608-739, Korea*

<sup>2</sup>*Korea Institute of Materials Science, Changwon, Gyeongnam 641-831, Korea*

(Received 19 February 2010, Received in final form 10 March 2010, Accepted 11 March 2010)

The effects of Cu-addition and die-upset temperature on the texture in the die-upset Nd-lean  $\text{Nd}_x\text{Fe}_{93.5-(x+y)}\text{Cu}_y\text{Ga}_{0.5}\text{B}_6$  ( $x=9-12$ ,  $y=0-2$ ) alloys were investigated. The die-upset Cu-containing Nd-lean  $\text{Nd}_{12}\text{Fe}_{81.5-y}\text{Cu}_y\text{Ga}_{0.5}\text{B}_6$  ( $y=1, 2$ ) alloys showed a considerable texture. Texture in the Nd-lean alloys developed through basal plane slip deformation. The Cu-addition reduced the melting point of grain boundary phase facilitating grain gliding during the die-upsetting, and providing a greater chance for the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  grains to meet the deformation conditions. Die-upsetting at higher temperature facilitated grain gliding and plastic deformation, thus enhancing texture.

**Keywords:** permanent magnets, Nd-Fe-B, texture, die-upset

### 1. Introduction

Die-upsetting is a unique process by which the hard magnetic  $\text{Nd}_2\text{Fe}_{14}\text{B}$  grains in a melt-spun Nd-Fe-B alloy are aligned by simple deformation at an elevated temperature. The likelihood of texture formation by die-upset is heavily dependent upon alloy composition, particularly the Nd-content. Thus far, for an Nd-rich Nd-Fe-B alloy with an Nd-content well over the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  stoichiometric composition (11.8 at%), the die-upset induced a good texture. However, for an Nd-lean Nd-Fe-B alloy with an Nd-content near or under the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  stoichiometric composition, die-upset only led to a poor texture. The good texture in the die-upset Nd-rich alloy was closely related to the presence of an abundant Nd-rich grain boundary phase, which is liquid during die-upsetting. The texture in the Nd-rich alloy develops by taking advantage of the liquid via a stress-induced preferential grain growth mechanism [1-3]. Meanwhile, the Nd-lean alloys bear little to no Nd-rich grain boundary phase, and thus it would seem natural that in the Nd-lean alloys a good texture cannot be achieved by the die-upset. However, an appreciable texture has recently been achieved in a die-upset Nd-lean alloy [4-6]. In the present study, the effects

of Cu-addition and die-upset temperature upon the texture of the die-upset Nd-lean Nd-Fe-B alloys were investigated.

### 2. Experimental Work

The  $\text{Nd}_x\text{Fe}_{93.5-(x+y)}\text{Cu}_y\text{Ga}_{0.5}\text{B}_6$  ( $x=9-12$ ,  $y=0-2$ ) starting alloys were melt-spun, and the obtained ribbon briefly milled. The milled powder was then hot-pressed at 750 °C, and subsequently die-upset at various temperatures ranging from 750 to 900 °C to achieve an approximate 75% height reduction. Process details of the hot-pressing and die-upsetting have been previously published [7]. Texture in the die-upset magnets was evaluated by the ratio of  $M_{7(\parallel)}/M_{7(\perp)}$ , where,  $M_{7(\parallel)}$  and  $M_{7(\perp)}$  are the magnetization at 7 kOe in the first quadrant of the demagnetisation curve parallel and perpendicular to the pressing direction. The demagnetisation curves (corrected using a demagnetising factor of 0.33) were measured using VSM at room temperature after pre-magnetizing with a 50 kOe pulsing field. Differential thermal analysis (DTA) was performed in Ar gas using the hot-pressed sample (100 mg). The microstructure of the die-upset sample was examined by observing the fracture surface of the sample using SEM after brief cracking.

\*Corresponding author: Tel: +82-51-629-6352

Fax: +82-51-629-6353, e-mail: hwkwon@pknu.ac.kr

## Results and Discussion

Fig. 1 shows the demagnetisation curves of the die-upset Nd-lean alloys ( $\text{Nd}_{12}\text{Fe}_{81.5}\text{Ga}_{0.5}\text{B}_6$  and  $\text{Nd}_9\text{Fe}_{84.5}\text{Ga}_{0.5}\text{B}_6$ ) measured parallel and perpendicular to the pressing direction. The Nd-lean alloys die-upset at  $750^\circ\text{C}$  showed poor texture. The poor texture was not improved even at higher temperature as shown in Fig. 2. The grain shape in the die-upset sample evidenced the poor texture. As shown in Fig. 3, the grains were in equi-axed shape, with no grain texture observed. The negligible texture in these Nd-lean alloys was attributed primarily to the lack of an Nd-rich grain boundary phase. Fig. 4 shows the DTA results of the hot-pressed Nd-Fe-B alloys with different compositions. The endothermic event around  $650^\circ\text{C}$  corresponded to the melting of the Nd-rich grain boundary phase in the alloys. No endothermic event for the alloy with an Nd-content of 9 at% indicated an absence of the grain boundary phase (Fig. 4(a)). Although some grain boundary phase existed in the alloy with a 12

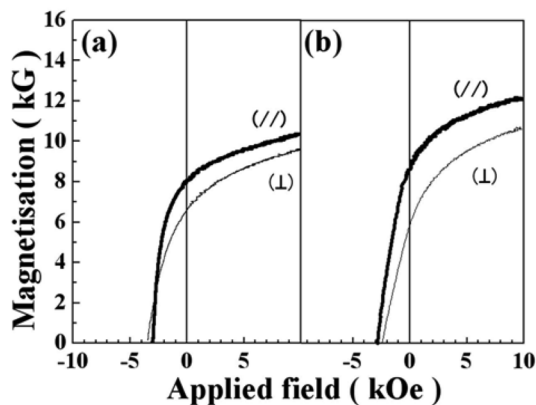


Fig. 1. Demagnetisation curves of the (a)  $\text{Nd}_{12}\text{Fe}_{81.5}\text{Ga}_{0.5}\text{B}_6$  and (b)  $\text{Nd}_9\text{Fe}_{84.5}\text{Ga}_{0.5}\text{B}_6$  alloys die-upset at  $750^\circ\text{C}$ .

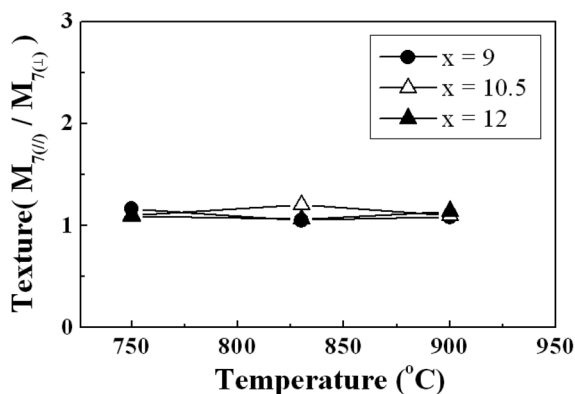


Fig. 2. Effect of die-upset temperatures on the texture of the die-upset  $\text{Nd}_x\text{Fe}_{93.5-x}\text{Ga}_{0.5}\text{B}_6$  alloys.

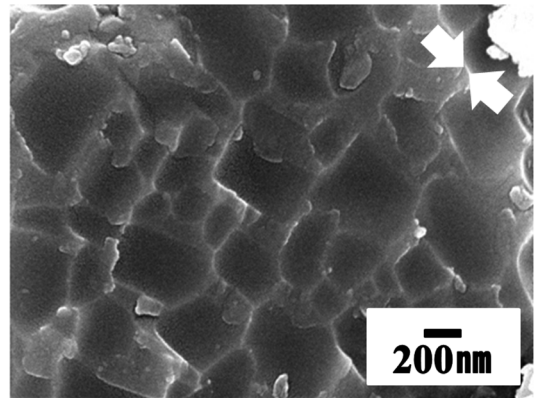


Fig. 3. SEM images showing the fracture surface of the  $\text{Nd}_{12}\text{Fe}_{81.5}\text{Ga}_{0.5}\text{B}_6$  alloys die-upset at  $830^\circ\text{C}$  (Arrows indicate pressing direction).

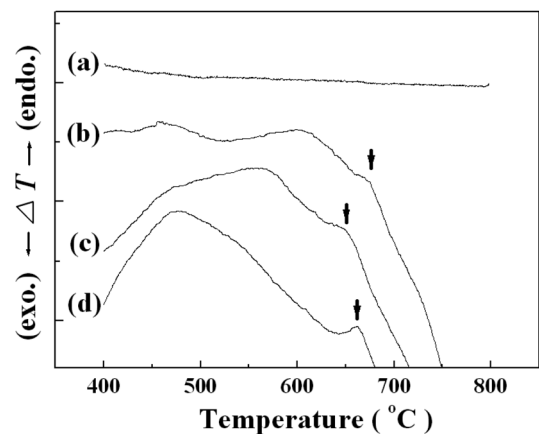


Fig. 4. DTA results on the hot-pressed compact of the (a)  $\text{Nd}_9\text{Fe}_{84.5}\text{Ga}_{0.5}\text{B}_6$ , (b)  $\text{Nd}_{12}\text{Fe}_{81.5}\text{Ga}_{0.5}\text{B}_6$ , (c)  $\text{Nd}_{12}\text{Fe}_{79.5}\text{Cu}_2\text{Ga}_{0.5}\text{B}_6$ , and (d)  $\text{Nd}_{13.5}\text{Fe}_{80}\text{Ga}_{0.5}\text{B}_6$  alloy.

at% Nd-content (Fig. 4(b)), it was much smaller compared to that for the Nd-rich alloy with a 13.5 at% Nd-content (Fig. 4(d)). Development of a texture in a die-upset Nd-Fe-B alloy was closely related to the Nd-rich grain boundary phase. The Nd-rich alloy (13.5 at% Nd) had a typical composition with good texture after the die-upset. This Nd-rich alloy inherently contained a considerable amount of Nd-rich grain boundary phase, became liquid during the die-upset. In the Nd-rich alloy, the texture developed via a stress-induced preferential growth of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  grain by taking advantage of the abundant liquid grain boundary phase [1-3]. During the die-upsetting, unfavourable grains, of which the c-axis is out of the pressing direction, are dissolved into the liquid grain boundary phase and precipitated preferentially on the lateral surface of the favourable grains, of which the c-axis is parallel to the pressing direction. The Nd-lean alloys studied herein bore little grain boundary phase,

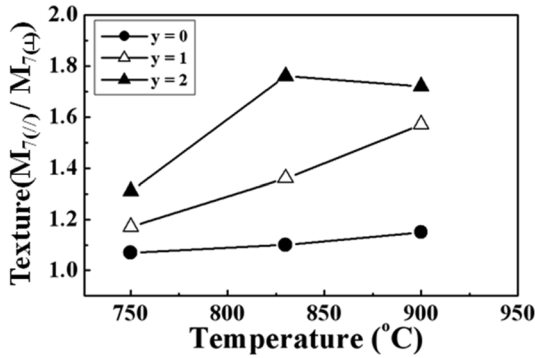


Fig. 5. Effect of Cu-addition and die-upset temperatures on the texture of  $Nd_{12}Fe_{81.5-y}Cu_yGa_{0.5}B_6$  alloys.

thus the stress-induced preferential grain growth could not operate.

Fig. 5 shows the effects of Cu-addition and die-upset temperature upon the texture in the Nd-lean  $Nd_{12}Fe_{81.5-y}Cu_yGa_{0.5}B_6$  alloys. The Cu-containing alloys showed textures in the samples die-upset at 750 °C much higher than the alloy without Cu. Moreover, unlike the alloy without Cu the Cu-containing alloys showed much improved texture when die-upset at higher temperatures. The induced texture in the Cu-added alloy was evidenced by the morphology of the grains in the sample die-upset at higher temperatures. As shown in Fig. 6(a), the grains in the Cu-added sample had a texture, whereby the grains were

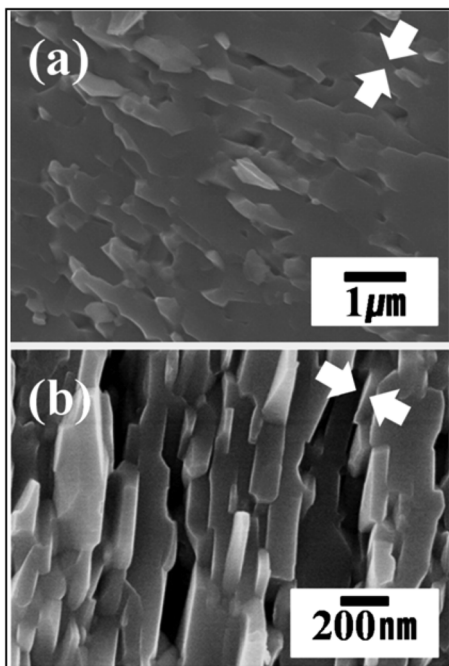


Fig. 6. SEM images showing the fracture surface of the (a)  $Nd_{12}Fe_{79.5}Cu_2Ga_{0.5}B_6$  alloy die-upset at 900 °C, and (b)  $Nd_{13.5}Fe_{80}Ga_{0.5}B_6$  alloy die-upset at 750 °C.

elongated and the long axis aligned perpendicular to the pressing direction. The texture in the Cu-containing Nd-lean alloy seems to have been induced by a slight enrichment of the grain boundary phase due to the Cu-addition. However, the DTA results showed that the increase in the amount of grain boundary phase by Cu-addition was insignificant (Fig. 4(b) and 4(c)). The lack of a grain boundary phase in the Cu-containing Nd-lean alloy could also be evidenced by the fracture surface morphology of the die-upset sample. In the Cu-added Nd-lean alloy the fracture generally occurred in a trans-granular mode (Fig. 6(a)), and this fracture mode was radically different from that in the Nd-rich alloy, in which the fracture occurred in an inter-granular mode along the abundant grain boundary (Fig. 6(b)). The trans-granular fracture in the Cu-added Nd-lean alloy indicated a lack of the grain boundary phase. Although the Cu-addition caused insignificant enrichment of the grain boundary phase, a considerable texture developed in the Cu-added alloys, indicating that the texture in the Cu-added Nd-lean alloy developed by a mechanism different from the stress-induced preferential grain growth operating in the Nd-rich alloy. Decisive evidence for a different texture formation mechanism in the Cu-added Nd-lean alloy was found in the temperature dependence of the texture. When the texture in a die-upset Nd-Fe-B alloy developed via the stress-induced preferential grain growth of crystallographically favourable grains, it usually decreased with increasing die-upset temperature [7]. In the present case, however, the reverse was true, in which the texture increased with increasing die-upset temperature. The texture in a die-upset Nd-lean alloy is known to develop by a basal plane slip deformation mechanism [7]. In this mechanism an easy grain boundary gliding during the die-upsetting facilitates texture inducement. As seen in Fig. 4(c), the Cu-addition significantly reduced the melting point of the grain boundary phase (from 680 to 650 °C approximately). A previous study reported that the Cu atoms added to the Nd-Fe-B alloy primarily segregated at the grain boundaries rather than dissolved into the  $Nd_2Fe_{14}B$  matrix grains [8]. The melted grain boundary region probably possessed higher fluidity, thus providing better lubrication, and facilitating grain gliding during die-upsetting. Therefore, more grains may have a chance where the magnitude of the resolved shear stresses along the basal plane reaches a critical value for plastic deformation. This effect may become more profound as the die-upset temperature increases because the lubrication of the grain boundary is enhanced. Simultaneously, the critical value for the slip deformation is reduced at higher die-upset temperatures and the  $Nd_2Fe_{14}B$  grains deform more readily. These two

factors of the Cu-addition and higher die-upset temperatures may possess synergy sufficient for inducing a texture in the die-upset Nd-lean Nd-Fe-B alloys.

### Conclusion

A considerable texture was developed in the die-upset Nd-lean  $\text{Nd}_{12}\text{Fe}_{81.5-y}\text{Cu}_y\text{Ga}_{0.5}\text{B}_6$  ( $y = 1, 2$ ) alloys through basal plane slip deformation. The Cu-addition to the Nd-lean Nd-Fe-Ga-B alloys significantly reduced the melting point of the grain boundary phase, facilitating grain gliding during the die-upsetting, more grains thus have a chance where the magnitude of the resolved shear stresses along the basal plane reach a critical value for plastic deformation. At higher die-upset temperatures grain gliding and plastic deformation were enhanced. Accordingly, this could come in to play for texture improvement.

### Acknowledgement

The present work was supported by a grant from the Fundamental R&D Program for Core Technology of

Materials funded by the Ministry of Knowledge Economy, Republic of Korea.

### References

- [1] R. K. Mishra, E. G. Brewer, and R. W. Lee, *J. Appl. Phys.* **63**, 3528 (1988).
- [2] L. Li and C. D. Graham Jr., *J. Appl. Phys.* **67**, 4756 (1990).
- [3] L. Li, and C. D. Graham Jr., *IEEE Trans. Magn.* **28**, 2130 (1992).
- [4] A. M. Gabay, Y. Zhang, and G. C. Hadjipanayis, *J. Magn. Magn. Mater.* **294**, 287 (2005).
- [5] A. M. Gabay, Y. Zhang, and G. C. Hadjipanayis, *J. Magn. Magn. Mater.* **302**, 244 (2006).
- [6] G. C. Hadjipanayis, J. F. Liu, A. M. Gabay, and M. Marinescu, *Proc. 19th International Workshop on Rare Earth Permanent Magnets and Their Applications, Beijing, China, Vol. I*, 12 (2006).
- [7] H. W. Kwon and J. H. Yu, *IEEE Trans. Magn.* **45**, 4435 (2009).
- [8] L. F. C. P. Lima, H. Takiish, and R. N. Faria, *J. Magn. Magn. Mater.* **285**, 112 (2005).