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DISTRIBUTION OF Al IN Nd-Fe-Al-B PERMANENT MAGNETS

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Abstract. – ⁵⁷Fe Mössbauer spectroscopy and electron microprobe analysis are used to determine the Al-distribution in the Nd-Fe-Al-B alloys; Mössbauer spectroscopy gives the Al distribution between the J₂ and K₂ sites of Nd₂Fe₁₄B-like-phase; microprobe analysis gives the composition of both matrix and minor phases and their evolution with sintering.

Substitution of Al for Fe in the Nd-Fe-B permanent magnets increases H_c and decreases B_r and T_c [1, 2]. A ⁵⁷Fe Mössbauer study and a microprobe investigation allowed us to determine the distribution of Al atoms in as-cast and sintered alloys of nominal weight composition

Nd_{33.7}(Fe_{1-y}Al_y)_{65.15}B_{1.15} and

Nd_{35.8}(Fe_{1-y}Al_y)_{63.1}B_{1.1} for $y = 0.007$;

$y = 0.017$; $y = 0.027$ which correspond to a nominal atomic ratio (Al/Al + Fe) of 0.015; 0.035; 0.055.

In addition to the magnetic Nd₂Fe₁₄B type phase Mössbauer spectra show the paramagnetic phase Nd₁₁Fe₄B₄ (weight 6 %); no other phase was detected except iron traces in as-cast alloys; high and low Nd alloys show similar spectra.

A low substitution of Al (1 to 3 atoms per unit cell) in the Nd₂Fe₁₄B-like phase dramatically changes the Mössbauer spectra: decreasing of mean magnetic fields at iron sites and asymmetric line-broadening typical of a hyperfine field distribution (Fig. 1); this seems to give evidence that substitution takes place mainly in the most magnetic sites (i.e. those with the higher number of nearest-neighbour-iron atoms), to say J₂ and K₂, which are also the largest sites [3, 4]. Assuming this preferential occupation the spectra are fitted with a near-neighbour-model, which takes into account the decreasing of hyperfine field ΔH_i at every site i (due to

Fig. 1. – ⁵⁷Fe Mössbauer spectra of Nd₂(Fe_{1-x}Al_x)₁₄B at room temperature.

Table I. – Distribution of n Al atoms between J₂ and K₂ per unit cell in Nd₂(Fe_{1-x}Al_x)₁₄B. (n_e : nominal concentration, n_m : microprobe analysis, n : Mössbauer fits).

x	n_e	n_m	n	z J ₂	$n - z$ K ₂
0.015	0.84	1.0	0.85	0.85	0
0.035	1.96	1.9	2.0	1.1	0.9
0.055	3.08	3.1	3.0	1.5	1.5
		±0.2	±0.1	±0.1	

an Al atom near-neighbour of a Fe atom) and this leads to the determination in the Nd₂Fe₁₄B-like phase, of the Al concentration, the Al distribution between the sites (Tab. I) and the iron hyperfine fields.

In all the samples, we found (in the magnetic phase) the Al nominal atomic concentration.

Fits show that Al substitution occurs first in the J₂ sites up to a concentration corresponding to one substituted atom per cell. Further Al substitution occurs also in the K₂ site up to an equipartition between the two sites for $n = 3$ ($x = 0.055$).

Hyperfine fields decrease with n but ΔH_i remains fairly constant (about 20 kOe) except for J₂ sites where it is about 10 kOe (probably due to a greater distance).

Electron microprobe analysis allowed us to study both the matrix and the minor phases.

In the matrix phase, we found a homogeneous Al distribution, with composition close to that determined with Mössbauer spectroscopy (Tab. I).

The B-rich $\text{Nd}_{1.1}\text{Fe}_4\text{B}_4$ phase, however is observed to have a near-zero Al concentration. In the Nd-rich phase, the Al distribution is not homogeneous and generally very low compared with the distribution in the matrix phase (Figs. 2a and 2b).

Fig. 2. - Electron microprobe micrographs for $\text{AlK}\alpha$: a) as-cast ingot; b) sintered magnet ($100 \times 100 \mu\text{m}$). The arrows indicate the Al-rich phase.

Moreover, in the as-cast samples, we detected Al concentrations up to 5 % weight together with Nd and Fe, suggesting the existence of an Al-rich phase [5]

which is located in the Nd-rich intergranular phase in the ingots or in the triple junction phase in the sintered samples. The latter seems to disappear partially during sintering.

Fig. 3. - Calculated and experimental saturation magnetization [7] (dashed line) versus Al-content.

At last, the saturation magnetization *versus* Al concentration was calculated assuming proportionality of the mean hyperfine field (given by Mössbauer spectroscopy) with the mean magnetic moment at iron sites [6]. A good agreement is observed with the experimental values (Fig. 3).

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