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Phase diagram of Sm−Nd−Fe ternary system

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Abstract: Phase equilibria of the Sm−Nd−Fe ternary system were determined in an isothermal section of 500 °C and vertical sections of $SmFe_2$–$NdFe_2$ and $(Sm_{0.86}Nd_{0.12})Fe_x$ ($1.6\leq x\leq 2.4$) by optical microscopy, X-ray diffraction analysis, electron probe microanalysis (EPMA), and differential thermal analysis (DTA) techniques. There are four intermetallic phases: $(Sm,Nd)Fe_2$, $(Sm,Nd)Fe_3$, $(Sm,Nd)_5Fe_{17}$ and $(Sm,Nd)_2Fe_{17}$ in the Sm−Nd−Fe ternary system, and no $(Sm,Nd)_6Fe_{23}$ phase exists in this system. The isothermal section of 500 °C possesses 7 single-phase regions, 8 two-phase regions, and 4 three-phase regions. The vertical section of $SmFe_2$−$NdFe_2$ contains 2 single-phase regions, 4 two-phase regions, and 7 three-phase regions, and that of $(Sm_{0.88}Nd_{0.12})Fe_x$ consists of 2 single-phase regions, 5 two-phase regions, and 2 three-phase regions. The peritectic temperatures for $(Sm_{1-x}Nd_x)Fe_2$ alloys decrease with increasing the Nd content when $x\leq 0.5$, and the substitution of Nd for Sm makes the stability of $(Sm,Nd)Fe_2$ phase decrease.

Key words: Sm−Nd−Fe system; phase diagram; structure; compound

1 Introduction

Intermetallic compounds of rare-earth and transition metal have been widely utilized as permanent magnets, magnetostrictive materials, and magnetic recording media. Among these systems, the Sm–Fe system has been the focus of considerable attention and extensive studies because its compounds have high spontaneous magnetization and remain magnetic at relative high temperatures [1,2]. In actual magnets, other rare-earth elements are added in order to induce special magnetic properties. For example, the replacement of Sm by Dy or Nd in $SmFe_2$ compound can make the magnetocrystalline anisotropies of RFe_2 compounds reduced because the magnetocrystalline anisotropies are dominated by the rare-earth ions. The investigation of the magnetostriction and anisotropy compensation for $Sm_{1-x}Nd_xFe_2$ [3] and $Sm_{0.88}Nd_{0.12}(Fe_{1-x}Co_x)_{1.93}$ [4] has found that the $(Sm,Nd)Fe_2$ compounds possess very high saturation magnetostriction. Structure, magnetic properties and magnetostriction of $Sm_{1-x}Nd_xFe_{1.55}$ alloys were studied in Ref. [5], and it was found that the alloys with $0\leq x\leq 0.48$ contain mainly cubic Laves phase, besides minor rare earths. However, the metallurgical process of Sm−Fe alloys is very complex [6] and it is necessary to have the knowledge of solidification process of Sm−Nd−Fe alloys. Therefore, it is interesting to investigate the phase diagram of the Sm−Nd−Fe ternary system in order to develop a new type of magnetostrictive material and permanent magnetic rare earth-iron alloys.

The Sm−Fe binary system contains $SmFe_2$, $SmFe_3$ and Sm_2Fe_{17} phases [7,8]. Two phases, Nd_5Fe_{17} and Nd_2Fe_{17}, exist in the Nd−Fe system [7,8], nevertheless, the cubic Laves compound $NdFe_2$ has been successfully prepared by high pressure technique. There are α-Nd and α-Sm phases in the Sm−Nd system at room temperature [7]. The crystallographic data of their compounds are summarized in Table 1 [7−9]. Up to now, the phase diagram of the Sm−Nd−Fe ternary system has not been published. In this work, the isothermal section of 500 °C, and vertical sections of $SmFe_2$−$NdFe_2$ and $(Sm_{0.86}Nd_{0.12})Fe_x$ ($1.6\leq x\leq 2.4$) are investigated for the Sm−Nd−Fe ternary system.

2 Experimental

Alloys were prepared from materials with the

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Table 1 Crystallographic data of compounds in Sm−Fe and Nd−Fe binary systems

Compound	Pearson symbol	Space group	Structure type	Refs.
$SmFe_2$, $NdFe_2$	*cF*24	$Fd\bar{3}m$	$MgCu_2$	[7,8]
$SmFe_3$	*hP*24	$R\bar{3}m$	$PuNi_3$	[7,8]
Sm_6Fe_{23}	*cF*116	$Fd\bar{3}m$	Th_6Mn_{23}	[7,8]
$SmFe_7$	*tP*68	$P4_2/mnm$	−	[7,8]
Nd_5Fe_{17}	*hP*264	$P6_3/mcm$	Nd_5Fe_{17}	[7−9]
Sm_2Fe_{17}, Nd_2Fe_{17}	*hR*19	$R\bar{3}m$	Th_2Zn_{17}	[7,8]

following purities: Sm, Nd, 99.9% (mass fraction) and Fe 99.8%. Pure metals are made into small pieces, mixed according to the stoichiometry and compacted into pellets. The pellets were heated in an alumina crucible to 400 °C and kept at 400 °C for 10 h under high-purity argon in order to obtain composition-homogenized samples. To compensate for evaporation loss of Sm during melting, 10% more of Sm was added in excess of the stoichiometric amount. Then, they were melted three times in an arc furnace under high-purity argon. During the melting, the melting current should be suitably controlled in order to reduce the loss of Sm. 54 specimens were prepared and the mass loss of each sample was kept below 1%. The as-cast samples wrapped in Mo foil were sealed in silica tube filled with high-purity argon. Specimens were homogenized at different temperatures (Table 2) on the basis of the Sm−Fe and Nd−Fe binary phase diagrams [6−8] and then quenched in water.

Table 2 Composition and heat treatment conditions of alloys in Sm−Nd−Fe system

No.	Alloy	Composition	Heat treatment conditions
1	$(Sm_{1-x}Nd_x)Fe_2$	x=0, 0.1, 0.12, 0.2, 0.3, 0.4, 0.5	800 °C, 10 d, then 500 °C, 10 d
		x=0.6, 0.7, 0.8, 0.9, 1.0	700 °C, 10 d, then 500 °C, 10 d
2	$(Sm_{1-x}Nd_x)Fe_3$	x=0, 0.2 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 1.0	750 °C, 10 d, then 500 °C, 10 d
3	$(Sm_{1-x}Nd_x)_5Fe_{17}$	x=0, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 1.0	750 °C, 10 d, then 500 °C, 10 d
4	$(Sm_{1-x}Nd_x)_6Fe_{23}$	x=0, 0.15, 0.3, 0.5, 0.75, 1.0	800 °C, 10 d, then 500 °C, 10 d
5	$(Sm_{1-x}Nd_x)_2Fe_{17}$	x=0, 0.15, 0.3, 0.5, 0.75, 1.0	1000 °C, 5 d, then 500 °C, 15 d
6	$(Sm_{0.86}Nd_{0.12})Fe_x$	x=1.6, 1.7, 1.8, 1.85, 1.9, 1.95	650 °C, 15 d
		x=2.0, 2.05, 2.1, 2.2, 2.3, 2.4	800 °C, 15 d

Characterization of the specimens was performed using optical microscopy. The etchant used was 2% nital. Differential thermal analysis (DTA) was carried out with heating and cooling rates of 10 K/min by using an LCP-1-type high temperature differential thermal dilatometer. The DTA furnace was evacuated with a pressure of 5×10^{-3} Pa and then kept under high-purity argon atmosphere in order to prevent the oxidation of samples in the alumina crucibles during the experiment. Pure Cu (99.99%) was used to check the temperature. X-ray diffraction analysis was carried out in a D/max-rA diffractometer equipped with a pyrolytic graphite monochromator. Cu K_α radiation was used. Software Jade 5.0 was used to analyze the XRD data of all samples. Electron probe microanalysis (EPMA) was performed in a Camebax-micro analyzer.

3 Results and discussion

3.1 Intermetallic phases

Metallographic examination, X-ray diffraction analysis, and EPMA confirm that there are four intermetallic phases: $(Sm,Nd)Fe_2$, $(Sm,Nd)Fe_3$, $(Sm,Nd)_5Fe_{17}$ and $(Sm,Nd)_2Fe_{17}$.

3.1.1 $(Sm,Nd)Fe_2$

According to Sm−Fe and Nd−Fe systems, $SmFe_2$ phase is stable, and the $NdFe_2$ phase does not exist. Metallographic examination and X-ray diffraction analysis confirm that the structure of as-cast $(Sm_{1-x}Nd_x)Fe_2$ alloys consists of the majority of $(Sm,Nd)Fe_2$ phase with $MgCu_2$-type cubic structure when $x\leq0.55$. Small amounts of phases are the $(Sm,Nd)Fe_3$ and rare earth-rich phase when $x\leq0.4$ and the $(Sm,Nd)_5Fe_{17}$ phase occurs when x=0.5 (Fig. 1).

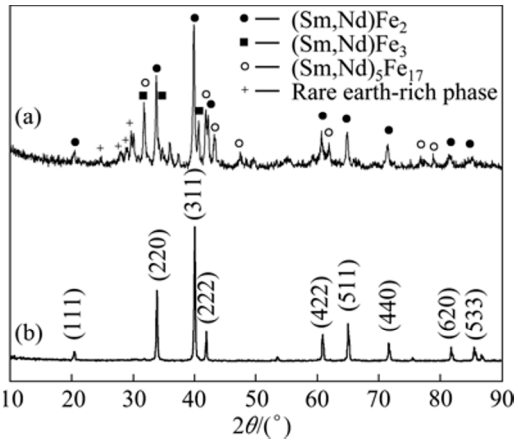

Fig. 1 X-ray diffraction patterns of as-cast (a) and annealed (b) $(Sm_{0.5}Nd_{0.5})Fe_2$ alloy

After being annealed, the $(Sm_{1-x}Nd_x)Fe_2$ alloys are the single $(Sm,Nd)Fe_2$ phase when $x\leq0.5$. As we know, the $NdFe_2$ compound does not exist in the equilibrium