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Effects of Nd- and Zr- substitution for Sm on magnetic and structural properties in SmFe_{11}V 1:12 compound

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Abstract

Structural and magnetic properties of polycrystalline alloys of $\text{Sm}_{1-x}\text{Y}_x\text{Fe}_{11}\text{V}$ [$\text{Y}=\text{Nd}$ ($0 \leq x \leq 0.4$), Zr ($0 \leq x \leq 0.6$)] were investigated. Substitution of Nd for Sm promoted the formation of α -(Fe,V) and 3:29 phases. In the case of Zr-substitution, all the optimally heat-treated alloys crystallize into tetragonal ThMn_{12} -type structure 1:12 phase and bcc α -(Fe,V) phase. For high Zr content of $x = 0.4$ and 0.6 , Laves phase ZrFe_2 was also detected with Curie temperature of 709 and 697 K, respectively. The substitution of Zr resulted in a contraction of 0.7% of the 1:12 cell volume. Oriented powder XRD patterns indicated that the easy magnetization direction was along (002) c -axis. As Zr content increased from $x = 0$ to 0.6 , anisotropy field decreased from 11 to 9 T and Curie temperature from 625 to 583 K. On the other hand, the saturation magnetization increased from 114 to 138 $\text{Am}^2 \text{kg}^{-1}$. Among the alloys investigated in the present study, the $\text{Sm}_{0.61}\text{Zr}_{0.40}\text{Fe}_{11.03}\text{V}_{0.96}$ with saturation magnetization of 129 $\text{Am}^2 \text{kg}^{-1}$, anisotropy field of 9.4 T and Curie temperature of 599 K is the most favorable candidate for permanent magnet applications.

Keywords: Hard magnets, ThMn_{12} -type structure, iron alloys, rare-earth lean magnets.

1. Introduction

In response to the increasing limitations on critical rare-earth (R) metals supplies [1, 2], a global effort is being devoted to find R-lean/free magnetic phases suitable for permanent magnet applications [3]. As a result, there has been a renewed interest in $\text{R}(\text{Fe},\text{M})_{12}$ (M is transition metal, such as Ti, V, Mo, Si, used as stabilizer of the 1:12 phase) compounds because of their reduced R concentration (7.7 at.% as compared with 11.8 at.% in $\text{Nd}_2\text{Fe}_{14}\text{B}$, 16.7 at.% in SmCo_5 and 10.5 at.% in $\text{Sm}_2\text{Co}_{17}$), excellent intrinsic magnetic properties and a simple crystal structure. The structure is tetragonal ThMn_{12} -type, $I4/mmm$ space group, with three nonequivalent sites ($8i$, $8j$ and $8f$) for Fe and M together and one site ($2a$) for R [4]. The most promising candidate materials are those with $\text{R} = \text{Nd}$ and Sm . Figure 1 shows Curie temperature (T_C), saturation magnetization ($\mu_0 M_s$) and magnetocrystalline anisotropy ($\mu_0 H_A$) of the most investigated 1:12 compounds [5–21].

Previous studies have shown that the SmFe_{12} -based compounds as potential candidates for permanent magnet applications as it has been possible to get coercivities up to 1.1 T [22–28] in Sm-Fe-V alloy (bulk and ribbons), and 1.32 T [29] in $\text{Sm}(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$ alloy (thin films). However, for bulk alloys, the remanence observed has been relatively low (< 0.5 T). It is necessary to increase the remanence via texture or increasing saturation magnetization along with the stability of 1:12 phase. In this direction, there had been investigations where Sm was partially or fully substituted by other R elements or non R elements such as Zr. Substitution of Sm by

Ce, Pr, Gd, Tb or Dy in $\text{SmFe}_{11}\text{Ti}$ resulted in decreasing $\mu_0 H_A$ and $\mu_0 M_s$ [27, 30, 31]. More recently Zhao et al. [32] studied the core-shell structure (Sm-rich shell and Y-rich core) in $(\text{Sm}_{0.75}\text{Y}_{0.25})\text{Fe}_{0.8}\text{Co}_{0.2})_{11.25}\text{Ti}_{0.75}$ alloy, as Y substitution not only can stabilize the ThMn_{12} -type structure, but also suppress the precipitation of the α -Fe phase. On the other hand, when Sm is substituted by Nd the $\mu_0 M_s$ either remained unchanged [31] or increased [33]. Kim et al. [31] reported that $\mu_0 H_A$ decreased more than 75% when half of Sm was substituted by Nd in $\text{SmFe}_{11}\text{Ti}$, while Niarchos et al. [33] reported a decrease of 50%.

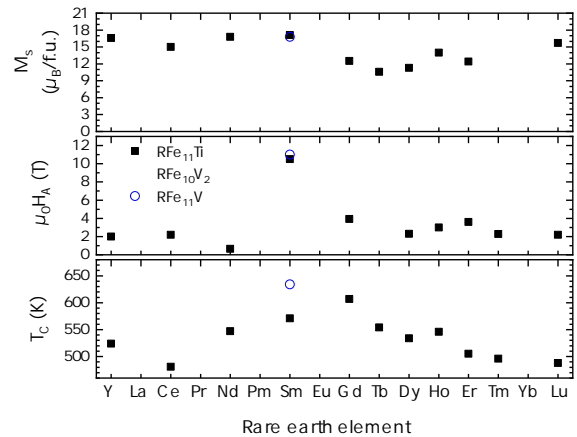


Figure 1: Intrinsic magnetic properties of $\text{R}(\text{Fe},\text{Ti})_{12}$ and $\text{R}(\text{Fe},\text{V})_{12}$ for different R elements [5–21].

Tozman et al. [34] investigated the effect of V on the phase stability and intrinsic magnetic properties of $\text{SmFe}_{12-x}\text{V}_x$ ($0 \leq x \leq 1.9$) thin films demonstrating V-substituted compounds with 7 at.% phase stabilizers have more merit than Ti-substituted ones.

The last decade there have been reports investigating the stability of the $\text{R}(\text{Fe},\text{M})_{12}$ phase by replacing the R (R = Nd or Sm) with less expensive R metals such as Ce [35, 36] or by non R metals such as Zr [37–39]. Among them, R-Fe-Ti and R-Fe-Si systems have been widely investigated. According to the previous studies [40, 41], Zr atoms occupy only the rare-earth 2a sites. Sakurada et al. [42] reported that substitution of Nd with Zr stabilized the 1:12 phase up to $x \approx 0.75$ in $\text{Nd}_{1-x}\text{Zr}_x\text{Fe}_{10}\text{Si}_2$. More recently Gabay et al. [43] synthesized $\text{Zr}(\text{Fe},\text{Si})_{12}$, the first R-free 1:12 compound. They reported a $\text{ZrFe}_{10.4}\text{Si}_{1.6}$ alloy with μ_0M_s of 1.15 T but with a very weak $\mu_0H_A \approx 1.9$ T. Partial substitution of Zr with R = Y, La, Ce, Pr and Sm has been investigated by Gabay et al. [44]. In terms of μ_0H_A , the effect of Sm was the strongest with a value of 4.07 T in $\text{Zr}_{0.7}\text{Sm}_{0.3}\text{Fe}_{10}\text{Si}_2$. In the same direction, Tozman et al. [39] attained the 1:12 phase in $\text{Sm}_{1-y}\text{Zr}_y(\text{Fe}_{0.8}\text{Co}_{0.2})_{11.5}\text{Ti}_{0.5}$ alloys with small concentration of Zr ($y = 0.1$ to 0.3). However, with increase in Zr content from $y = 0.1$ to 0.3, μ_0M_s decreased from 1.61 to 1.52 T and μ_0H_A from 9.2 to 7.9 T. A maximum T_C of 830 K was reported for $\text{Sm}_{0.8}\text{Zr}_{0.2}(\text{Fe}_{0.8}\text{Co}_{0.2})_{11.5}\text{Ti}_{0.5}$ alloy.

Up to now, no study has been reported on the intrinsic magnetic properties of $(\text{Sm},\text{R})\text{Fe}_{11}\text{V}$ compounds. This paper studies the effect on the structural, microstructural, thermal and magnetic properties of the Sm-substitution by Nd in $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ ($0 \leq x \leq 0.4$) and by Zr in $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ ($0 \leq x \leq 0.6$) alloys. The SmFe_{11}V alloy was chosen as it has a strong μ_0H_A , a relatively high μ_0M_s and T_C , which are suitable for permanent magnet applications [20].

2. Experiment

Polycrystalline samples were prepared by arc melting the constituent elements under argon atmosphere in a water-cooled copper crucible. The ingots were turned over and remelted three times to ensure their homogeneity. The Sm losses were compensated by adding an appropriate excess of Sm. The as-cast ingots were sealed in evacuated quartz tubes filled with Ar, heat-treated at temperatures ranging from 1000 to 1100°C for 3–4 days and quenched in water.

X-ray diffraction (XRD) and room-temperature magnetic measurements were performed on powders prepared by hand grinding. The XRD patterns were taken using a Rigaku Ultima IV diffractometer with $\text{Cu-K}\alpha$ ($\lambda = 1.5418 \text{ \AA}$) radiation in backscattering Bragg-Brentano geometry. For structural characterization, analysis of the XRD patterns was performed using a FullProf suite [45] based on the Rietveld method [46]. Easy-axis magnetization was found from the intensities of lines on diffractograms of magnetically oriented powder samples.

Magnetization was measured with a VersaLab 3 T cryogen-free PPMS from Quantum Design and a high field VSM with a superconducting magnet (up to 14 T) cooled with He closed circuit refrigerator (CFMS from Cryogenic Ltd). The μ_0H_A

estimation was performed on oriented samples which were prepared by mixing the powders ($< 44 \mu\text{m}$) with a cold-curing epoxy and dried under a 2 T magnetic field. The demagnetization factor of the aligned powders was considered negligible as the μ_0H_A was large enough. The T_C was obtained from $M(T)$ measurements at an applied field of 0.05 T for small ($\approx 15 \text{ mg}$) heat-treated samples.

Microscopic analysis of polished unetched alloy samples was carried out by using Hitachi TM3000 Tabletop scanning electron microscope (SEM) and the composition analysis was performed with a Quantax70 energy dispersive X-ray spectroscopy (EDX) detector.

3. Results and Discussion

Figure 2 presents XRD patterns of $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ ($x = 0, 0.2$ and 0.4) alloys heat-treated at 1100°C for 4 days.

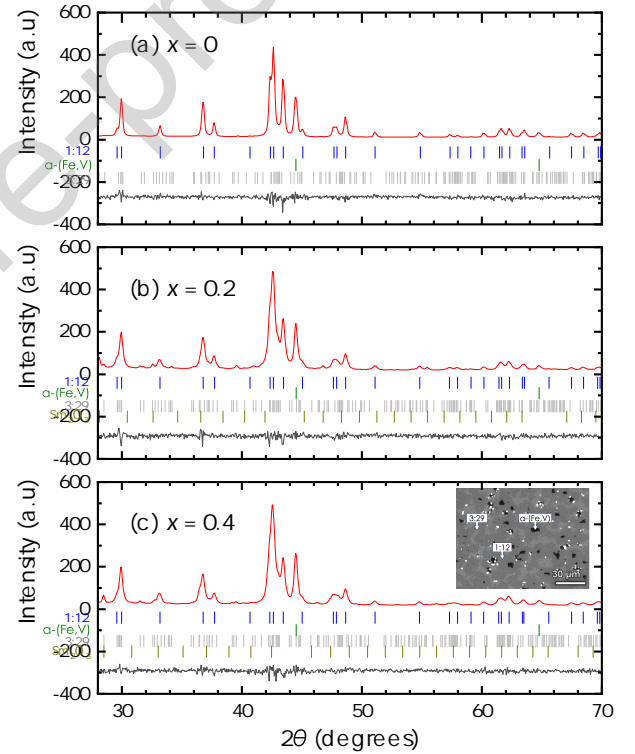


Figure 2: Experimental (black dots) and calculated (continuous line) XRD patterns of $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ alloys heat-treated at 1100°C for 4 days (a) $x = 0$, (b) $x = 0.2$ and (c) $x = 0.4$. Boxes include the error and small vertical lines refer to Bragg peak positions of the phases present. Inset in Fig. 2(c) corresponds to the BSE-SEM micrograph of $x = 0.4$ alloy.

The diffraction patterns of all the samples present characteristic lines corresponding to the 1:12, 3:29 and α -(Fe,V) phases. For the case of $x = 0.2$ and 0.4 small amount of Sm_2O_3 is also observed. Although the 1:12 phase is maintained throughout the Nd-substitution, Nd promotes the formation of the 3:29 phase ($\text{Nd}_3\text{Fe}_{29}$)-type structure with the monoclinic symmetry and C2/m space group). Observation of the 3:29 phase is consistent with the existing ternary equilibria of Sm-Fe-V at 1100 and 1200°C (both for 20 h) [47]. From the phase diagram

(1100°C) in the reference, we can see that there are two phase fields near pure 1:12 phase, 3:29+ α -(Fe,V) and 1:12 + α -(Fe,V). Considering our slightly different annealing conditions (longer durations), formation of 3:29 phase along with 1:12 is possible in $\text{Sm}_{7.7}\text{Fe}_{84.6}\text{V}_{7.7}$ and in $(\text{Sm,Nd})_{7.7}\text{Fe}_{84.6}\text{V}_{7.7}$.

Volume fractions and lattice parameters of each phase derived from the Rietveld analysis are listed in Table 1. The SEM-EDX composition analysis confirmed the results obtained by XRD. For $x = 0.4$ sample (inset in Fig. 2c) the matrix in dark gray corresponds to the 1:12 phase, this exists together with α -(Fe,V) in black, and a minor phase in light gray which is identified as 3:29. The small bright spots correspond to free Sm. This phase was not observed in the XRD due to its low volume fraction. The chemical composition of the 1:12 phase obtained by SEM-EDX analysis are listed in table 2.

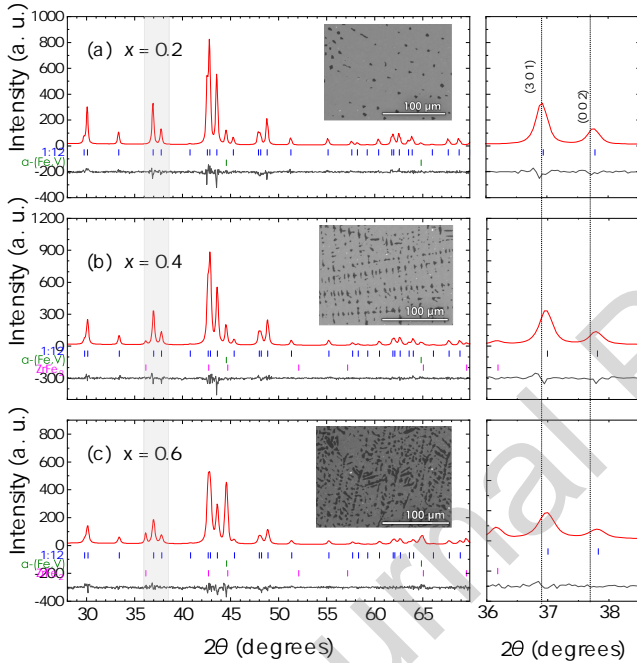


Figure 3: Experimental (black dots) and calculated (continuous line) XRD patterns of optimally heat-treated $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ alloys (a) $x = 0.2$ (b) $x = 0.4$ and (d) $x = 0.6$. Small vertical lines refer to Bragg peak positions of phases present and the error is shown below them. Enlarged peaks from the gray regions are shown at the right side. Insets correspond to the SEM-BSE images of the respective samples.

Figure 3 displays the XRD patterns of the optimally heat-treated $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ ($x = 0.2, 0.4$ and 0.6) alloys. The major phase in all these samples is 1:12, followed by α -(Fe,V)

and the cubic Laves phase ZrFe_2 (MgCu₂-type, $Fd\bar{3}m$ space group). As Zr content increases, the α -(Fe,V) phase increases and the Laves phase precipitates for $x \geq 0.4$. This is in agreement with the frequent formation of Laves phase compounds in alloys involving transition metals [48]. Volume fractions and lattice parameters of each phase present derived from the Rietveld analysis are listed in Table 1. As Zr has a smaller atomic radius (1.60 Å [49]) than Sm (1.80 Å [49]), the increase in Zr results in lattice contraction as observed from the 1:12 peaks shifting towards higher 2θ angles as shown in Fig. 3 enlarged (301) and (002) peaks at the right side.

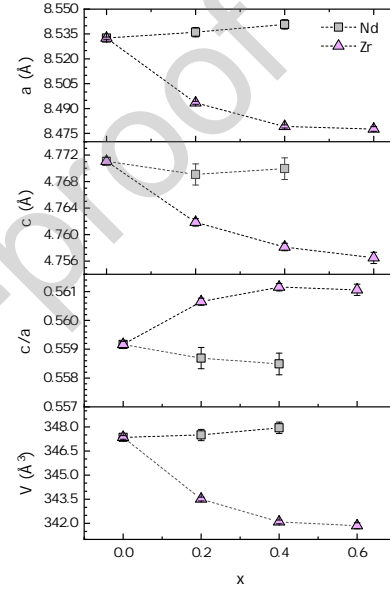


Figure 4: Lattice parameters (a and c), c/a ratio and unit cell volume (V) of the 1:12 phase of heat-treated $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ and $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ alloys.

Lattice parameters a and c , c/a ratio and cell volume (V) of the 1:12 phase for Zr- and Nd-substitution are shown in Fig. 4. The parameter a decreases from 8.532 to 8.478 Å and c from 4.771 to 4.756 Å with increasing Zr from $x = 0$ to 0.6. An increase in c/a ratio can also be seen. The unit cell contraction comes with a 0.7% of volume reduction. Tozman et al. [39] reported a contraction of lattice parameter a from 8.53 to 8.50 Å in $\text{Sm}_{1-y}\text{Zr}_y(\text{Fe}_{0.8}\text{Co}_{0.2})_{11.5}\text{Ti}_{0.5}$ alloys with increasing y from 0 to 0.3. In contrast, expansion of the lattice with increasing Nd content occurs predominantly in the basal plane, resulting in a decrease of c/a ratio. The expansion comes with a 0.2%

Table 1: Volume fractions and lattice parameters of the phases present in $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ and $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ heat-treated alloys.

x	composition	Volume fraction					Lattice parameters							
		1:12	α -(Fe,V)	3:29	Sm_2O_3	ZrFe_2	1:12		α -(Fe,V)	3:29			Sm_2O_3	ZrFe_2
						$a(\text{Å})$	$c(\text{Å})$	$a(\text{Å})$	$a(\text{Å})$	$b(\text{Å})$	$c(\text{Å})$	β	$a(\text{Å})$	$a(\text{Å})$
0.0	$\text{Sm}_{0.95}\text{Fe}_{10.97}\text{V}_{1.08}$	0.796	0.152	0.052		8.532	4.771	2.879	10.568	8.523	9.709	96.855		
0.2	$\text{Sm}_{0.80}\text{Nd}_{0.20}\text{Fe}_{11.00}\text{V}_{1.00}$	0.695	0.135	0.156	0.014	8.536	4.769	2.878	10.612	8.546	9.720	96.877	10.986	
0.4	$\text{Sm}_{0.59}\text{Nd}_{0.40}\text{Fe}_{10.99}\text{V}_{1.02}$	0.667	0.146	0.176	0.011	8.541	4.770	2.878	10.629	8.561	9.710	96.817	10.851	
0.2	$\text{Sm}_{0.77}\text{Zr}_{0.20}\text{Fe}_{10.98}\text{V}_{1.05}$	0.95	0.05			8.494	4.762	2.876						
0.4	$\text{Sm}_{0.61}\text{Zr}_{0.40}\text{Fe}_{11.03}\text{V}_{0.96}$	0.91	0.06			8.479	4.758	2.877						7.021
0.6	$\text{Sm}_{0.40}\text{Zr}_{0.60}\text{Fe}_{11.02}\text{V}_{0.98}$	0.70	0.23			8.478	4.756	2.875						7.022

of volume increment from $x = 0$ to 0.4. In the case of Nd-substitution in $\text{SmFe}_{11}\text{Ti}$ reported by Niarchos et al. [33], there is an increase of a from 8.5572 to 8.5639 Å and a decrease of c/a ratio from 0.5609 to 0.5592 with increasing Nd from $x = 0$ to 0.5.

Insets in Fig 3(a), (b) and (c) show SEM-BSE micrographs of the heat-treated alloys $x = 0.2$, 0.4 and 0.6 respectively. Narrow dendrites of α -(Fe,V) phase between 5 and 20 μm are formed and uniformly distributed, the matrix phase is identified as 1:12 phase, and the light gray areas as the ZrFe_2 Laves phase, the latter is only visible for $x = 0.6$. A SEM-BSE micrograph and EDX elemental mappings for $x = 0.6$ are shown in Fig. 5. Segregation of α -(Fe,V) and ZrFe_2 phases can be seen where, the former are represented by the green-red dendrites and the latter by the blue-greens. The chemical composition of the 1:12 phase determined by SEM-EDX are listed in Table 2.

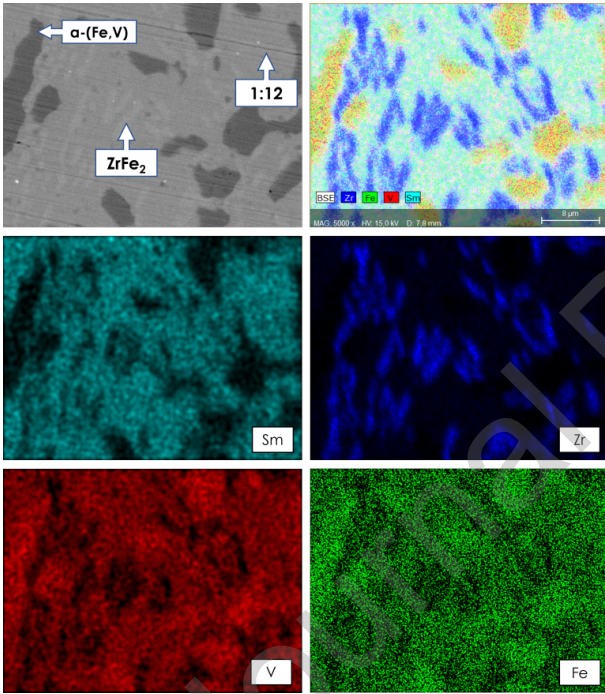


Figure 5: SEM-BSE micrographs and EDX composition maps of Sm, Zr, V and Fe elements from the polished surface of the optimally heat-treated $\text{Sm}_{0.4}\text{Zr}_{0.6}\text{Fe}_{11}\text{V}$ alloy.

Table 2: Intrinsic magnetic properties of 1:12 phase in $\text{Sm}_{1-x}\text{Y}_x\text{Fe}_{11}\text{V}$ ($\text{Y}=\text{Nd}, \text{Zr}$) alloys and their composition determined by SEM-EDX. Magnetization values given in parenthesis are in T.

x	Composition	M_s ($\text{Am}^2 \text{kg}^{-1}$)	$\mu_0 H_A$ (T)	T_C (K)
0.0	$\text{Sm}_{0.95}\text{Fe}_{10.97}\text{V}_{1.08}$	114 (1.11)	11.0	625
0.2	$\text{Sm}_{0.80}\text{Nd}_{0.20}\text{Fe}_{11.00}\text{V}_{1.00}$	111 (1.08)	10.6	608
0.4	$\text{Sm}_{0.59}\text{Nd}_{0.40}\text{Fe}_{10.99}\text{V}_{1.02}$	109 (1.06)	7.4	593
0.2	$\text{Sm}_{0.77}\text{Zr}_{0.20}\text{Fe}_{10.98}\text{V}_{1.05}$	115 (1.13)	10.2	608
0.4	$\text{Sm}_{0.61}\text{Zr}_{0.40}\text{Fe}_{11.03}\text{V}_{0.96}$	129 (1.28)	9.4	599
0.6	$\text{Sm}_{0.40}\text{Zr}_{0.60}\text{Fe}_{11.02}\text{V}_{0.98}$	138 (1.37)	9.0	583

Figure 6 (left) shows room-temperature initial magnetization curves of the oriented powders of $\text{Sm}_{1-x}\text{Y}_x\text{Fe}_{11}\text{V}$ ($\text{Y}=\text{Nd},$

Zr) alloys measured \parallel and \perp to the magnetic field orientation direction. On the right side, corresponding XRD patterns of the oriented powders are shown. The M_s values for the 1:12 phase were deduced by subtracting the contribution from the secondary phases (magnetization at room temperature: $M_{3:29}=78.8 \text{ Am}^2 \text{kg}^{-1}$ [50], $M_{\alpha\text{-Fe}}=218 \text{ Am}^2 \text{kg}^{-1}$ [51] and $M_{\text{ZrFe}_2}=43.2 \text{ Am}^2 \text{kg}^{-1}$ [52]) taking into account their volume fractions from the XRD analysis (Table 1). In the case of Zr-substitution, M_s at room temperature increases from 114 to 138 $\text{Am}^2 \text{kg}^{-1}$ (1.11 to 1.37 T) with increasing Zr from $x = 0$ to 0.6. Kuno et al. [53] reported an increase in $\mu_0 M_s$ of 1:12 phase from 1.50 to 1.58 T with increase in Zr from $y = 0$ to 0.2 in $\text{Sm}_{1-y}\text{Zr}_y(\text{Fe}_{0.8}\text{Co}_{0.2})_{11.5}\text{Ti}_{0.5}$ alloys. Also Tozman et al. [54] reported an increase in $\mu_0 M_s$ from 1.78 to 1.90 T with increasing Zr-substitution from $x = 0$ to 0.26 in $\text{Sm}_{1-x}\text{Zr}_x(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$ thin films. The enhancement in $\mu_0 M_s$ was recently explained (by using XAFS technique) as a result of electron transfer from Zr to the Fe 3d atoms which causes an increase in the magnetic moment and magnetic interactions in the Fe 3d site [55]. For Nd-substitution, M_s at room temperature decreases from 114 $\text{Am}^2 \text{kg}^{-1}$ (1.11 T) in $x = 0$ to 109 $\text{Am}^2 \text{kg}^{-1}$ (1.06 T) in $x = 0.4$. A similar behavior was reported by Kim et al. [31] when half of Sm was substituted for Nd in $\text{SmFe}_{11}\text{Ti}$ alloy, the $\mu_0 M_s$ decreased from 1.27 to 1.21 T.

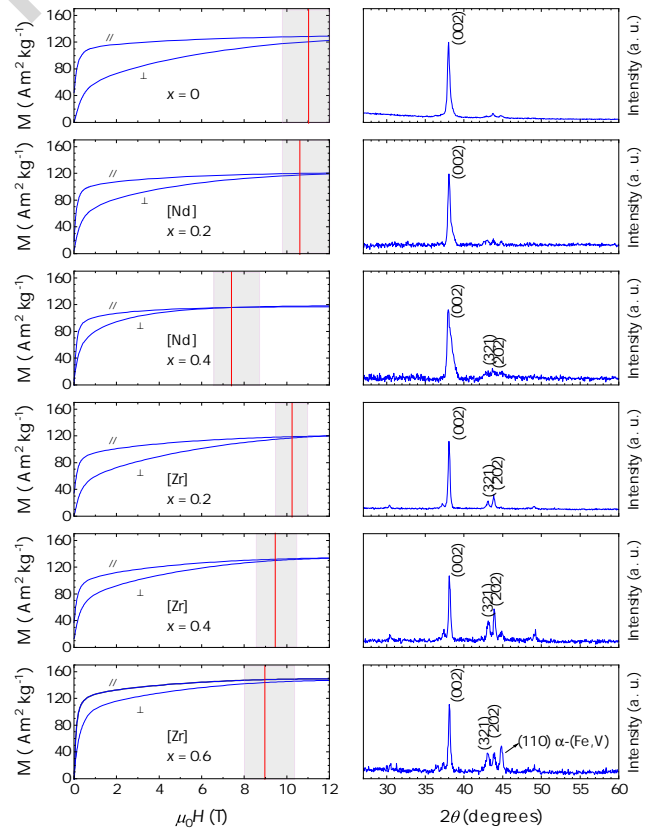


Figure 6: (Left) Magnetization at room temperature measured \parallel and \perp to the orientation direction as a function of applied field for optimally heat-treated $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ and $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ alloys. (Right) Corresponding XRD data of oriented optimally heat-treated samples.

The $\mu_0 H_A$ was determined by plotting M in the perpendicular direction as a function of $[(\mu_0 H)^{-2}]$ [56] from Fig. 6, and it was obtained from the minimum of the derivative [20, 39, 57]. The highlighted region in the plots shows the uncertainty in determining the anisotropy field (see supplementary data). Table 2 summarizes the intrinsic magnetic properties. As Zr content is increased, $\mu_0 H_A$ decreases from 11 T ($x = 0$) to 9 T ($x = 0.6$). Even for highest Zr content the $\mu_0 H_A$ is still higher than NdFeB anisotropy field (6.7 T [58]). Tozman et al. [39, 54] reported a decrease in $\mu_0 H_A$ on substitution of Zr for Sm in $\text{Sm}_{1-y}\text{Zr}_y(\text{Fe}_{0.8}\text{Co}_{0.2})_{11.5}\text{Ti}_{0.5}$ bulk alloys and in $\text{Sm}_{1-x}\text{Zr}_x(\text{Fe}_{0.8}\text{Co}_{0.2})_{12}$ thin films. This decrease in $\mu_0 H_A$ with increasing Zr is attributed to the decrease of R content. The high anisotropy is mainly due to the R element, this property is linked to the magnetic coupling between the R and M sublattices [59]. In accordance with the anisotropy field, XRD measurements of field oriented powder samples indicate that the easy-axis of magnetization is along the c -axis (002). The other reflexions such as (321) and (202) are attributed to the presence of the secondary phases or misoriented grains. In case of Nd-substitution, $\mu_0 H_A$ decreases from 11 to 7.4 T with increasing Nd from $x = 0$ to 0.4, respectively. Kim et al. [31] and Niarchos et al. [33] reported a similar decrease in $\mu_0 H_A$ on Nd-substitution in $\text{SmFe}_{11}\text{Ti}$ alloy system.

All the magnetization curves measured in direction \perp to the orientation have a curvature in the low-field region. This can be due to the combination of two effects, the presence of a soft phase of high susceptibility α -(Fe,V), and poor alignment, as the particles (size $< 44 \mu\text{m}$) may not all be single crystals and also have irregular external shapes that prevent their perfect alignment.

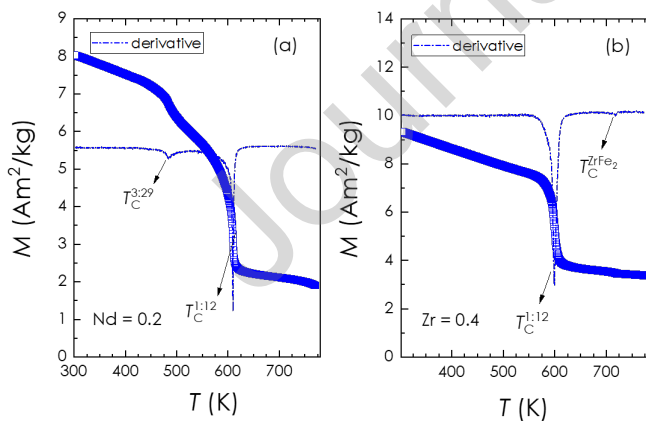


Figure 7: Typical thermomagnetic curves depicting T_C of different phases present in optimally heat-treated (a) $\text{Sm}_{0.8}\text{Nd}_{0.2}\text{Fe}_{11}\text{V}$ and (b) $\text{Sm}_{0.6}\text{Zr}_{0.4}\text{Fe}_{11}\text{V}$ alloys.

Values of T_C of 1:12 phase for $\text{Sm}_{1-x}\text{Y}_x\text{Fe}_{11}\text{V}$ ($\text{Y}=\text{Nd}$, Zr) are summarized in Table 2. The optimally heat-treated $\text{Sm}_{1-x}\text{Nd}_x\text{Fe}_{11}\text{V}$ alloys show that the T_C of 1:12 phase decreases from 625 to 593 K with increase of Nd from $x = 0$ to 0.4. In the case of $\text{Sm}_{1-x}\text{Zr}_x\text{Fe}_{11}\text{V}$ alloys, T_C of 1:12 phase decreases from 625 to 583 K with the Zr-substitution from $x = 0$ to

0.6. Figure 7(a) and (b) shows typical thermomagnetic scans for heat-treated $\text{Sm}_{0.8}\text{Nd}_{0.2}\text{Fe}_{11}\text{V}$ and $\text{Sm}_{0.6}\text{Zr}_{0.4}\text{Fe}_{11}\text{V}$ alloys, respectively. The T_C is strongly determined by the transition metals M-M interaction. When Sm is substituted by Nd or Zr, the interatomic distances between the near neighbors of Fe atoms at the different sites are affected resulting in reduced total interaction causing a decrease in T_C .

The 1:12 is not the only magnetic phase present in these alloys, but there is also a small quantity of 3:29 phase in Nd-substituted alloy and ZrFe_2 in Zr-substituted alloy, which are also observed in XRD analysis. The T_C at 483 K in Fig. 7(a) corresponds to 3:29 phase and the T_C at 709 K in Fig. 7(b) corresponds to ZrFe_2 Laves phase. The T_C of 3:29 decreases from 483 to 473 K with increasing Nd from $x = 0.2$ to 0.4. The T_C of Laves phase decreases from 709 to 697 K with increasing Zr from $x = 0.4$ and 0.6. Brückner et al. [52] found that T_C of the Laves phase increases from 610 to 798 K as the Fe content increases from 66.7 to 72.3 at.%. Based on this dependency of the T_C on Fe concentration for ZrFe_2 , we can say that Fe content in the Laves phase in the alloys studied in the current work is between 68.7 and 70.6 at.%.

4. Conclusions

Effects of Zr- and Nd-substitution for Sm on magnetic and structural properties in SmFe_{11}V alloy were studied. For all the optimally heat-treated alloys, the main phase was found to be 1:12. As Zr content increased from $x = 0$ to 0.6, the $\mu_0 H_A$ decreased from 11 to 9 T and the T_C from 625 to 583 K while M_s increased from 115 to 138 $\text{Am}^2 \text{kg}^{-1}$. For high content of Zr ($x = 0.4$ and 0.6) Laves ZrFe_2 phase was formed. The volume fraction of ZrFe_2 and α -(Fe,V) increased with increasing Zr content. In the case of Nd-substitution, $\mu_0 H_A$, M_s and T_C decreased with increasing Nd content, the substitution of even small quantities of Sm by Nd promotes the formation of the 3:29 phase, which ruin the desired magnetic properties. The $\text{Sm}_{0.61}\text{Zr}_{0.40}\text{Fe}_{11.03}\text{V}_{0.96}$ alloy exhibits suitable intrinsic magnetic properties for permanent magnet applications, $\mu_0 M_s = 1.28 \text{ T}$, $\mu_0 H_A = 9.4 \text{ T}$ and $T_C = 599 \text{ K}$. Also it contains only 4.6 at.% of rare-earth metal compared to the 7.7 at.% used in the SmFe_{11}V alloy.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Highlights

- We investigated the effects of substitution of Sm by RE element Nd and non RE element Zr on structure and magnetic properties in $\text{Sm}_{1-x}\text{Y}_x\text{Fe}_{11}\text{V}$ (Y=Nd, Zr) alloys.
- The 1:12 phase is maintained throughout the Nd- and Zr- substitution.
- Substitution of Nd promotes the formation of α -(Fe,V) and 3:29 phase.
- As Zr content increases from $x = 0$ to 0.6, anisotropy field and Curie temperature decreases while saturation magnetization increases.
- The 1:12 phase with composition $\text{Sm}_{0.61}\text{Zr}_{0.40}\text{Fe}_{11.03}\text{V}_{0.96}$ exhibits optimum combination of magnetic properties of $\mu_0 M_s = 1.28$ T, $\mu_0 H_A = 9.2$ T and $T_C = 599$ K. It contains only 4.6 at. % of rare-earth metal compared to the 7.7 at. % used in the SmFe_{11}V alloy.