An influence of Si on the structural and magnetic properties of Dy(Fe,V,Si)\(_{12}\) compounds is studied. X-ray diffraction shows that these materials crystallise in the tetragonal Th\(_{12}\)-type structure. An increase in the saturation magnetization, the Curie temperatures and magnetocrystalline anisotropy in the Si substitutional systems is observed when compared with DyFe\(_{12-x}V_x\). The magnetic properties depend on the preferential site occupation and concentration of Si and V atoms in these compounds.

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1. Introduction

Iron-rich intermetallic compounds of the Th\(_{12}\)-type structure have been proposed as inexpensive alternatives for permanent magnetic materials. It is known that the rare earths and iron do not form the binary RFe\(_{12}\) phase, a third element is necessary to stabilize the tetragonal Th\(_{12}\)-type structure [1]. These compounds have the composition RFe\(_{12-x}M_x\), in which M represents a nonmagnetic 3d element, for instance Ti, V, Cu or metalloid Si. Apart from the R elements and 3d elements, which mainly determine the magnetic properties of the compounds, also stabilizing elements contribute to the magnetic properties. The aim of this work is to study the influence of the substitution of Si instead of V on the magnetic properties of DyFe\(_{12-x}M_x\) compounds.

2. Experimental

Samples of nominal composition DyFe\(_{10}V_{2-x}Si_x\), DyFe\(_{9}V_{3-x}Si_x\) and DyFe\(_{8}V_{4-x}Si_x\) (\(x = 0.05, 1.0, 1.5, 2.0\)) were prepared by arc-melting. The samples were prepared in the same way as those described in [2].

The structure and homogeneity of these compounds were checked by X-ray diffraction and thermomagnetic measurements. Lattice parameters were measured.
using X-ray powder diffractometer Siemens 5000 with Cu $\text{K}_\alpha$ radiation. Recorded lines were fitted with X-Profiler (written by A. Kachel) and the HIX61 programme was applied to find exact values of the lattice parameters.

Magnetic moment was studied at fields up to 5 T in the temperature range 1.7 K to room temperature. Magnetocrystalline anisotropy was investigated by means of ac susceptibility and torsion measurements in the temperature range 77–293 K. The easy direction of magnetization (EMD) was determined from X-ray diffraction of aligned powders. The most of the samples showed the easy $c$-axis at room temperature. Thermomagnetic curves at high temperatures were measured using a Faraday balance.

3. Results and discussion

All investigated compounds have the tetragonal ThMn$_{12}$-type structure. Slight decrease in the $a$ and the $c$ parameters with the increase in Si content were observed but the Vegard law was not fulfilled (Fig. 1).

![Graph showing lattice parameters, $T_c$, and average $\mu(\text{Fe})$ for DyFe$_{12-x}$(V, Si)$_x$ versus Si concentration.]

Fig. 1. Lattice parameters, $T_c$ and average $\mu(\text{Fe})$ for DyFe$_{12-x}$(V, Si)$_x$ versus Si concentration.

The investigated compounds are ferrimagnets with the Curie temperatures above room temperature. Plot of $T_c$ as a function of silicon concentration is presented in Fig. 1. The values of $T_c$ are almost unchanged for DyFe$_{10}V_{2-x}$Si$_x$ but increase in silicon-rich compounds of DyFe$_9V_{3-x}$Si$_x$ and DyFe$_8V_{4-x}$Si$_x$. The Curie temperature for DyFe$_8V_2$Si$_2$ is about 60 K higher than for DyFe$_8V_4$. Moreover, as shown in Fig. 2, the resultant magnetization increases with the Si content. Assuming that the magnetic moment of the Dy ions takes its free-ion value (10μ$_B$) the magnetic moment of the Fe atoms can be calculated from the values of the saturation magnetization obtained at 1.7 K. The estimated moment on Fe atom increases
markedly in DyFe$_{12-x}$(V, Si)$_x$ systems when compared with DyFe$_{12-x}$V$_x$. This is attributed to the enhancement of the exchange interaction due to the replacement of V by Si atoms. The effect of substituted Si atoms is similar to that observed in DyFe$_{12-x}$(V, C)$_x$ under carbon doping [2].

The ac-initial magnetic susceptibility versus temperature curves of DyFe$_{10}V_{2-x}$Si$_x$ (Fig. 3) shows strong anomalies which are shifted towards higher temperatures with increase in $x$. These peaks of ac-susceptibility correspond to spin reorientation due to magnetocrystalline anisotropy competition between the 3$d$ and rare-earth sublattices. At room temperature the EMD is determined by the Fe sublattice but at low temperatures the negative Dy anisotropy contributions cause the spin reorientation transition. The torsion measurements confirm the tilting of the EMD from the $c$-axis with lowering temperature. A cone ordering of the magnetic moments is observed for all studied compositions at 77 K. Generally, silicon content increases temperature of the beginning of the spin reorientation transition. The anisotropy constants reported for YFe$_{10}$V$_2$ and YFe$_{10}$Si$_2$ are nearly the same [3]. It follows that magnetocrystalline anisotropy of dysprosium sublattice is enhanced because any essential changes in the 3$d$ sublattices are not expected.

The analysis of spin reorientation in compounds with lower concentration of iron (e.g. DyFe$_9$V$_{2-x}$Si$_x$ and DyFe$_8$V$_2$Si$_2$) is more complicated. It should be noticed that metamagnetic behavior of resultant magnetic moment is observed at low temperatures (Fig. 2). Changes in the magnetization under the action of an external field seem to indicate the spin-flip. It follows from Fig. 2 that the critical field values depend on vanadium and silicon content in DyFe$_{12-x}$(V, Si)$_x$ series. The results of torsion measurements also demonstrate an influence of the applied magnetic field on the magnetic arrangement in the studied compounds.
The EMD in DyFe$_{9}$V$_{3-\text{x}}$Si$_{\text{x}}$ determined from torsion curves at 77 K in sufficiently small magnetic field is still the c-axis but markedly tilts from the c-axis in magnetic fields higher than 0.4 T. Temperature and field dependence of the spin reorientation transition are due to the varying the nonmagnetic M element concentration.

The obtained results indicate that magnetic properties of DyFe$_{12-\text{x}}$M$_{\text{x}}$ depend on the stabilizing element M. In these compounds, three different crystallographic sites are available for the Fe and M atoms. The V atoms prefer the 8i sites [3] whereas the Si atoms share the 8j and 8f positions with the Fe atoms [4]. The deeper analysis taking into account the various distribution of the Fe and M atoms and its influence on the exchange and crystal field interactions responsible for magnetic behavior in the studied compounds will be done elsewhere.

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References