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### ACCEPTED MANUSCRIPT

## Magnetic and magnetocaloric properties of $Gd(Ni_{1-x}Fe_x)_2$ quasi-binary Laves phases with $x = 0.04 \div 0.16$

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<u>Abstract</u> In this paper the results of specific magnetization (M), heat capacity (C<sub>P</sub>) and magnetocaloric effect (MCE) measurements for Gd(Ni<sub>1-x</sub>Fe<sub>x</sub>)<sub>2</sub> system over the Ni substitution by Fe range of x = 0÷0.16 are presented. Phase composition was controlled by X-ray diffraction analysis. Heat capacity was measured in the temperature range 77÷320 K. MCE has been studied within the temperature range 5÷400 K in magnetic fields up to 70 kOe by the entropy magnetic contribution change calculation ( $\Delta S_m$ ) and by direct  $\Delta T_{ad}$  measurements at the adiabatic conditions for external magnetic field change  $\Delta H = \pm 17.5$  kOe. It was found that the Fe concentration increase causes both the C<sub>P</sub> maxima disappearing at Curie temperature point and emergence of magnetic contribution to C<sub>P</sub> in a wide temperature range below this point. Moreover, in compounds with iron, a plateau-like temperature dependence of the MCE was observed for both magnetic entropy change ( $\Delta S_m$ ) and direct  $\Delta T_{ad}$  data which are independent on Fe concentration. The possible reasons of such behavior are discussed.

**Keywords:** Magnetic properties, magnetocaloric effect, magnetic entropy change, Laves phase, heat capacity.

#### 1. Introduction

Adiabatic magnetizing or demagnetizing of the RNi<sub>2</sub> or RCo<sub>2</sub> Laves phase type bulk samples (R is a heavy rare earth metal) at temperatures (T) close to their Curie temperature (T<sub>C</sub>) may abruptly change their temperature on a significant amount ( $\Delta T_{ad}$ ), i.e. they possess a high magnetocaloric effect (MCE) [1]. Since the T<sub>C</sub> values of these compounds are substantially lower the room T, they had no the interest yet from the developers of magnetic refrigeration devices. Nevertheless, the study of the magnetic properties and MCE in the R(Me<sub>1-x</sub>Fe<sub>x</sub>)<sub>2</sub> (Me = Al, Ni, Co) quasi-binary systems showed that a partial substitution of 3d-sublattice by Fe leads to T<sub>C</sub> increase, and the emergence of significant MCE in a wide temperature range below its T<sub>C</sub> [2, 3, 4, 5]. The latter is an important material property when it is used for the production of magnetic refrigerator working bodies.

Our recent MCE measurements for some  $Dy(Co_{1-x}Fe_x)_2$ ,  $Ho(Co_{1-x}Fe_x)_2$ , and  $Er(Co_{1-x}Fe_x)_2$  [6] compounds with Co substitution by Fe confirmed these results and allowed to suggest the reasons of MCE peak widening in the temperature range below their  $T_{C}$ .

It is well known that the RFe<sub>2</sub> type compounds with a heavy rare earth elements (R) having a non-zero R<sup>3+</sup>-ion orbital momentum, possess enormous magnetocrystalline anisotropy (MCA) at low temperatures originating from crystal field (CF) mechanism [7]. Such MCA mechanism can produce the "umbrella"-like magnetic structure in the R-ions subsystem due to local MCA fluctuations caused by a random substitution of Me-atoms by Fe ones at the nearest to R-ions positions in R(Me<sub>1-x</sub>Fe<sub>x</sub>)<sub>2</sub> (Me = Ni, Co) intermetallics [3]. In order to find out whether this factor is a key one for the MCE phenomena in the compounds with a large orbital moment of the 4f-electron shell R-ions (Tb, Dy, Ho, Er), we in this study examined the magnetic and magnetocaloric properties of Gd(Ni<sub>1-x</sub>Fe<sub>x</sub>)<sub>2</sub> (x ≤ 0.16) compounds. As far as the orbital magnetic moment of the Gd<sup>3+</sup>-ions in pure metal or in compounds with 3d metals is close to zero (S-state 4f-shell), the Gd-sublattice does not possess a high MCA from CF-mechanism [8]. In addition, the another motivation was due to the fact that magnetic and magnetocaloric properties of the Gd(Ni<sub>1-x</sub>Fe<sub>x</sub>)<sub>2</sub> compounds system have not been studied previously (excepting only the GdNi<sub>1,85</sub>Fe<sub>0,15</sub> compound [9]).

The started GdNi<sub>2</sub> binary compound is crystalized to the cubic C15 Laves phase superstructure. The crystal lattice has a doubled lattice parameter in comparison with the C15 one and is described by F-43m space group belonging to the TmNi<sub>2</sub> type structure [10]. The similar superstructure is formed in the Gd<sub>1</sub>.  $_xY_xNi_2$  [11] and Gd<sub>1-x</sub>Sc<sub>x</sub>Ni<sub>2</sub> [12] compounds, as well as in other R - Ni binary compounds [13]. But the iron

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