



Research articles

Magnetic and magnetocaloric properties of $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ compounds ($x = 0.25, 0.5, 0.75$)Santanu Pakhira^{a,b}, Chandan Mazumdar^{a,*}, R. Ranganathan^a^a Condensed Matter Physics Division, Saha Institute of Nuclear Physics, 1/AF, Bidhannagar, Kolkata 700064, India^b Ames Laboratory-USDOE and Department of Physics and Astronomy, Iowa State University, Ames, IA 50011, USA

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ABSTRACT

In this work, we report the magnetic dilution effect on the magnetic and magnetocaloric properties of $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0.25, 0.5, 0.75$) compounds by partial substitution of Y ions in Gd_2NiSi_3 . The X-ray diffraction studies indicate all these compounds form in AlB_2 type hexagonal crystal structure. The dc magnetic measurements reveal the compounds undergo antiferromagnetic like phase transition below 11.5, 7.2, and 3.1 K for $x = 0.25, 0.5$ and 0.75 compositions, respectively. The magnetic ground state of the compounds becoming increasingly fragile with the increase in Y concentration, and results in magnetic field induced ferromagnetic transition. Such change in magnetic spin configuration is responsible for moderately large magnetic entropy change ($\Delta S_M \sim 19.3 \text{ J/kg-K}$, 14.8 J/kg-K and 9.8 J/kg-K for $x = 0.25, 0.5$, and 0.75 , respectively, for $\Delta H = 7 \text{ T}$) in the cryogenic temperature region. The relative cooling power (RCP) has also been estimated for all the compounds that appears to be modest ($\sim 503 \text{ J/kg}$ for 7 T magnetic field change) for $x = 0.25$ analogue and reduces further with magnetic dilution effect caused by Y substitution.

1. Introduction

Rare earth based magnetic materials have attracted a significant attention of the researchers due to their complex phase transition among different magnetic phases under the influence of temperature, magnetic field and pressure. During the last few years, a group of rare earth based bulk intermetallic compounds have been extensively studied due to the exhibition of large magnetocaloric effect (MCE) [1–9]. MCE is an intrinsic thermodynamical phenomena of every magnetic materials where change in material temperature occurs under the influence of external magnetic field under adiabatic condition. MCE based magnetic refrigeration is of potential impact in both the field of science and technology due to their environment friendly and energy saving nature over conventional gas compression/expansion technique [1–3,10–12]. The studies of MCE in different rare earth based systems is not only considered to be important for their technological significance but also helps to understand different complex magnetic interactions in such systems [13–15]. Studies of MCE were primarily focussed on several ferromagnetic and some antiferromagnetic materials undergoing field induced ferromagnetic transition around their magnetic transition temperature [16–18]. However, in most of such materials, isothermal magnetic entropy change (ΔS_M), which is an important parameter to determine MCE in a magnetic system, decreases quite

faster as the sample temperature separates away from their respective magnetic transition temperature. Recently, quite a few different magnetically frustrated materials have been reported to exhibit large magnetic entropy change up to much higher temperature, in comparison to their phase transition temperature, which raised immense interest from practical point of view.

$R_2\text{TSi}_3$ -type of systems, with R = rare-earth ions, T = transition metals, is one of such magnetically frustrated systems where large MCE have been experimentally achieved [4,5,14,19–23]. In these systems, magnetic and magnetocaloric properties strongly depend on the local electronic environment among the magnetic rare earth ions [5,24–26]. Thus, one may expect different considerable changes in magnetic and magnetocaloric properties by tuning the local electronic interaction by means of external parameters, viz., external pressure [27], and chemical substitution [28,29]. To systematically investigate such change in magnetic and magnetocaloric properties, we have studied the effect of non-magnetic element yttrium (Y) substitution in one of the members, Gd_2NiSi_3 , of $R_2\text{TSi}_3$ family. It is reported that, the magnetic ground state of Gd_2NiSi_3 consists of coexistence of spin glass and antiferromagnetic interaction below 16.4 K and the compound exhibits large magnetic entropy change over a wide temperature scale [4]. In this work, we report the effect of magnetic dilution on the magnetic and magnetocaloric properties in $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0.25, 0.5, 0.75$) compounds.

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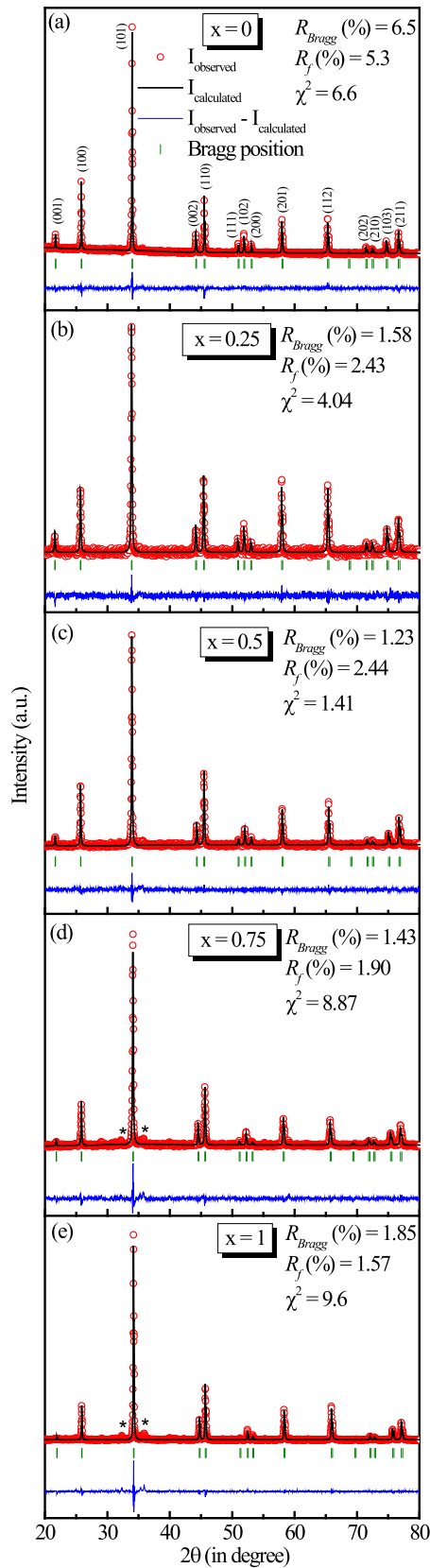


Fig. 1. Room temperature X-ray diffraction (XRD) pattern with full Reitveld refinement for (a) $x = 0$, (b) $x = 0.25$, (c) $x = 0.5$, (d) $x = 0.75$, and (e) $x = 1$ compositions along with fitting 'R'-factors. The additional peaks marked by '*' are the reflection for additional RNiSi_2 phase.

2. Experimental details

Polycrystalline $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0, 0.25, 0.5, 0.75, 1$) compounds were synthesized by standard arc-melting technique in argon atmosphere in an arc furnace. The ingots were melted repeatedly with flipping every time to ensure the homogeneity. Structural characterization was done by collecting the powder X-ray diffraction (XRD) pattern from a TTRAX-III diffractometer (M/s Rigaku, Japan) using $\text{Cu-K}\alpha$ radiation. The structural refinements were performed with the help of FULLPROF software package [30]. The temperature and magnetic field dependent magnetization measurements were carried out in a superconducting quantum interference device (SQUID) (M/s Quantum Design Inc., USA) in the temperature range 2–300 K and magnetic fields up to 7 T. Tiny cylindrical needle shaped samples having small average diameter (~ 0.5 mm) and large length (~ 4 mm) are used for magnetic measurements. The demagnetization factor of such geometry appear to be negligible, when the magnetic field is applied parallel to the length of the samples. The data for $x = 0$ compound have been reproduced from the earlier work reported in Ref. [4].

3. Results and discussions

Room temperature powder XRD patterns of $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0.25, 0.5, 0.75, 1$) compounds reveal that all the compounds form in AlB_2 type crystal structure with space group $P6/mmm$ (Fig. 1(a)–(e)). No significant structural change is observed with the increase of Y concentration. The samples with $x = 0, 0.25$ and 0.5 are single phase within the experimental resolution limit. However, for $x = 0.75$ and 1 substitution, one may find the presence of additional minor phase of stoichiometry RNiSi_2 . The estimated lattice parameters and the unit cell volume of the synthesized materials along with those of two end members Gd_2NiSi_3 and Y_2NiSi_3 are plotted in Fig. 2. The lattice parameter values of the Y-substituted compounds are also listed in Table 1. The stoichiometries estimated from full Rietveld analysis have also been provided in Table 1.

Fig. 3(a)–(d) depict the temperature dependence of magnetization at different applied magnetic fields for $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ compounds with $x = 0, 0.25, 0.5$, and 0.75 , respectively under both zero-field-cooled (ZFC) and field-cooled (FC) conditions. The magnetic transition appears to be in antiferromagnetic type in nature for all the compounds. However, the transition temperature shifts to lower temperature region with increase in Y substitution. The transition temperatures estimated from the $M(T)$ behaviour are 16.4, 11.5, 7.2, and 3.1 K for the compounds with $x = 0, 0.25, 0.5$, and 0.75 , respectively. In comparison to the magnetically undiluted compound Gd_2NiSi_3 ($T_N = 16.4$ K), the transition temperature reduces for the yttrium substituted compositions

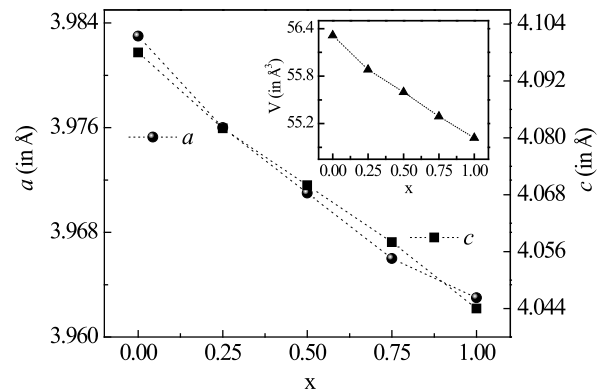


Fig. 2. The hexagonal lattice parameters (a and c) as a function of yttrium concentration (x). Inset shows corresponding unit cell volumes.

which can be easily understood as due to magnetic dilution in $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ compounds since Y has no significant role in magnetic interaction in these systems. The paramagnetic Curie–Weiss temperature (θ_p) and effective magnetic moment ($\mu_{\text{eff}}/\text{Gd ion}$) were determined for all these compounds using the relation $\frac{M}{H} = \frac{N\mu_{\text{eff}}^2\mu_0}{3k_B(T-\theta_p)} = \frac{C}{T-\theta_p}$, where, N is the number of magnetic atoms per unit volume, μ_0 is the permeability of free space, k_B is the Boltzmann constant, and C is the Curie constant. By fitting the temperature dependence of inverse susceptibility behaviour with linear Curie–Weiss behaviour in the paramagnetic region ($T > 55$ K for $x = 0.25$, $T > 50$ K for $x = 0.5$ and $T > 40$ K for $x = 0.75$) (Insets: Fig. 3(a)–(c)), we have estimated the values of θ_p and $\mu_{\text{eff}}/\text{Gd ion}$, which are listed in Table 1. The estimated θ_p values found to be positive in case of $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$ and GdYNiSi_3 revealing the presence of considerable ferromagnetic interaction along with dominating antiferromagnetic interaction. On the other hand, in case of $\text{Gd}_{0.5}\text{Y}_{1.5}\text{NiSi}_3$ compound, θ_p found to be negative which suggest that the ferromagnetic interaction strength also decreases with increasing Y substitution.

Isothermal magnetization measurements have been carried out at different temperatures to understand the magnetic field evolution of the magnetic ground state with the application of external magnetic field. Fig. 4(a)–(d) displays the $M(\mu_0H)$ behaviour at some selected temperatures for the compounds with x concentration 0, 0.25, 0.5, and 0.75, respectively. Though the low-field region of the magnetic isotherms found to be linear which is typical for any antiferromagnetic system, a considerable change in $M(\mu_0H)$ behaviour is observed at higher applied magnetic fields. As applied magnetic field strength increases, the $M(\mu_0H)$ isotherms deviate from linearity and exhibits field induced ferromagnetic like nature at higher fields. At 7 T field strength, $M(\mu_0H)$ behaviour almost saturates to a value of $\sim 7\mu_B/\text{Gd-ion}$, which is the theoretical saturation value expected for parallel alignment of all the Gd-ions. It may be mentioned here that, in case of the magnetically undiluted compound Gd_2NiSi_3 , full saturation moment was not achieved for 7 T magnetic field [4]. Our studies thus reveal that with the increase in Y substitution, the ground state of the compounds become more fragile and sensitive to perturbing magnetic field. From the Fig. 4(b)–(c), it is also evident that $M(\mu_0H)$ is not linear even at 50 K for $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$ and GdYNiSi_3 compounds. However, linearity is achieved at that temperature for $\text{Gd}_{0.5}\text{Y}_{1.5}\text{NiSi}_3$ compound and the results are in accordance with the inverse susceptibility behaviour. No magnetic hysteresis is observed in case of any of the compounds.

In order to evaluate the magnetocaloric properties of $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0.25, 0.5, 0.75$) compounds, isothermal magnetic entropy change (ΔS_M) were estimated from the magnetization isotherms measured at

different temperatures (below and above the transition temperatures) using Maxwell's relation,

$$\Delta S_M(T, \mu_0H) = \mu_0 \int_0^{\mu_0H} \left(\frac{\partial M}{\partial T} \right) dH \quad (1)$$

The temperature dependence of $-\Delta S_M$ for different magnetic field changes of the compounds have been presented in Fig. 5(a)–(c). All the compounds exhibit large value of $-\Delta S_M$ around the magnetic transition temperature. The maximum value found to be 19.3 J/kg-K, 14.8 J/kg-K and 9.8 J/kg-K in $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$, GdYNiSi_3 , and $\text{Gd}_{0.5}\text{Y}_{1.5}\text{NiSi}_3$ compounds, respectively, for 7 T applied magnetic field change. From the practical view point, it is always desirable to obtain large magnetic entropy change under low applied magnetic field. The maximum values of magnetic entropy change for 2, 5, and 7 T magnetic field changes are listed in Table 2. The values are moderate and particularly the value obtained for $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$ is quite comparable with that of different rare-earth based intermetallic compound reported to exhibit moderate MCE in this temperature region [18,22,14,33–37] (For detailed comparison with other reported good MCE materials please see ref [38]). The decrease in the $-\Delta S_M$ values with increase in Y substitution concentration could be well-understood by the decrease in ferromagnetic interaction due to Y substitution in the system. It should be mentioned here that, particularly in case of $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$ compound, moderately large value of $-\Delta S_M$ is observed above the ordering temperature which results in broadened peaks. Such type of broadening is generally observed for compounds having spin frustration or spin fluctuation that persists up to much higher temperature than the characteristic temperatures [4,13,31,32]. This type of broadening in $-\Delta S_M(T)$ behaviour also results in moderately large value of relative cooling power (RCP) in this compound. RCP is another important parameter to determine the refrigerant capacity of a magnetic material and is defined as $\text{RCP} = -\Delta S_M^{\text{max}} \times \delta T_{\text{FWHM}}$, where δT_{FWHM} is the full width at half maxima of $-\Delta S_M(T)$ curve. In case of $x = 0.75$ composition, the FWHM of the $-\Delta S_M(T)$ curve was estimated by extrapolation to lower temperature region using polynomial fit. From the Figs. 6(a)–(c), one may conclude that with the increase in Y substitution in the system, the spin fluctuation gets restricted to much lower temperature and results in faster decay of $-\Delta S_M$ values above the transition temperature. As a result, the FWHM of $-\Delta S_M(T)$ curve becomes much smaller in $\text{Gd}_{0.5}\text{Y}_{1.5}\text{NiSi}_3$ in comparison to lower Y-concentrated systems and is responsible for reduction in RCP values. However, as the spin fluctuation is higher in case $\text{Gd}_{1.5}\text{Y}_{0.5}\text{NiSi}_3$ compound, the estimated RCP value found to be modest, ~ 503 J/kg, 311 J/kg, and 66 J/kg for 7 T, 5 T, and 2 T magnetic field changes, respectively. The moderate RCP value for 2 T magnetic field change is comparable to different reported good magnetic refrigerant

Table 1

The hexagonal lattice parameters (a & c), unit cell volume (V), transition temperature (T_N), paramagnetic Curie–Weiss temperature (θ_p), effective moment (μ_{eff}), and the calculated stoichiometry from full Rietveld analysis for $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ compounds.

x	a (in Å)	c (in Å)	V (in Å ³)	T_N (in K)	θ_p	$\mu_{\text{eff}}/\text{Gd}$	Calculated stoichiometry
0	3.983(2)	4.098(3)	56.315(1)	16.4	14.3(0)	7.94(1)	$\text{Gd}_2\text{Ni}_{1.01(1)}\text{Si}_{2.98(1)}$
0.25	3.976(1)	4.082(2)	55.883(1)	11.5	8.53(1)	7.82(1)	$\text{Gd}_{1.5}\text{Y}_{0.48(2)}\text{Ni}_{0.98(1)}\text{Si}_{2.98(1)}$
0.5	3.971(1)	4.070(1)	55.597(1)	7.2	6.56(1)	7.83(1)	$\text{GdY}_{0.96(3)}\text{Ni}_{0.99(2)}\text{Si}_{3.0(1)}$
0.75	3.966(2)	4.058(1)	55.290(1)	3.1	-1.32(1)	7.86(1)	$\text{Gd}_{0.5}\text{Y}_{1.44(5)}\text{Ni}_{0.96(2)}\text{Si}_{2.97(3)}$
1	3.963(1)	4.044(1)	55.020(1)				$\text{Y}_2\text{Ni}_{0.99(2)}\text{Si}_{2.97(2)}$

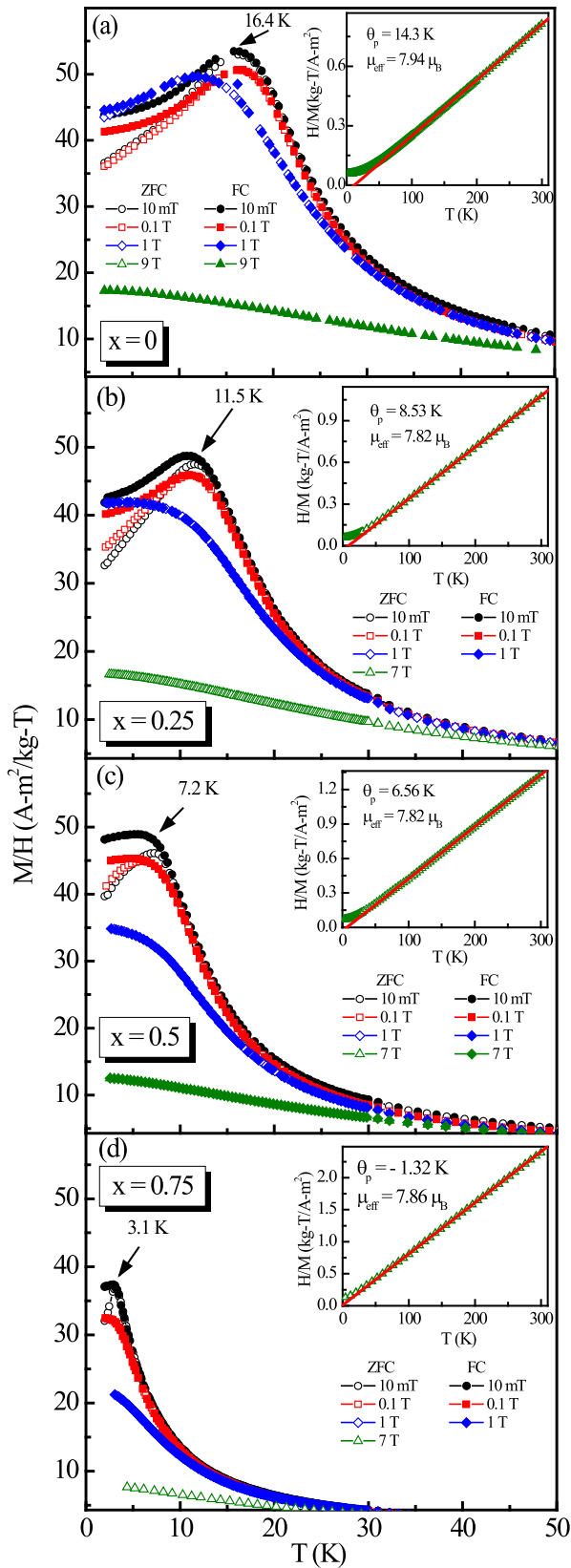


Fig. 3. Temperature dependence of dc magnetization at different applied magnetic fields under ZFC and FC conditions for (a) $x = 0$, (b) $x = 0.25$, (c) $x = 0.5$, and (d) $x = 0.75$ compositions. Insets show temperature dependence of inverse magnetic susceptibility with linear Curie-Weiss fit for the corresponding compounds.

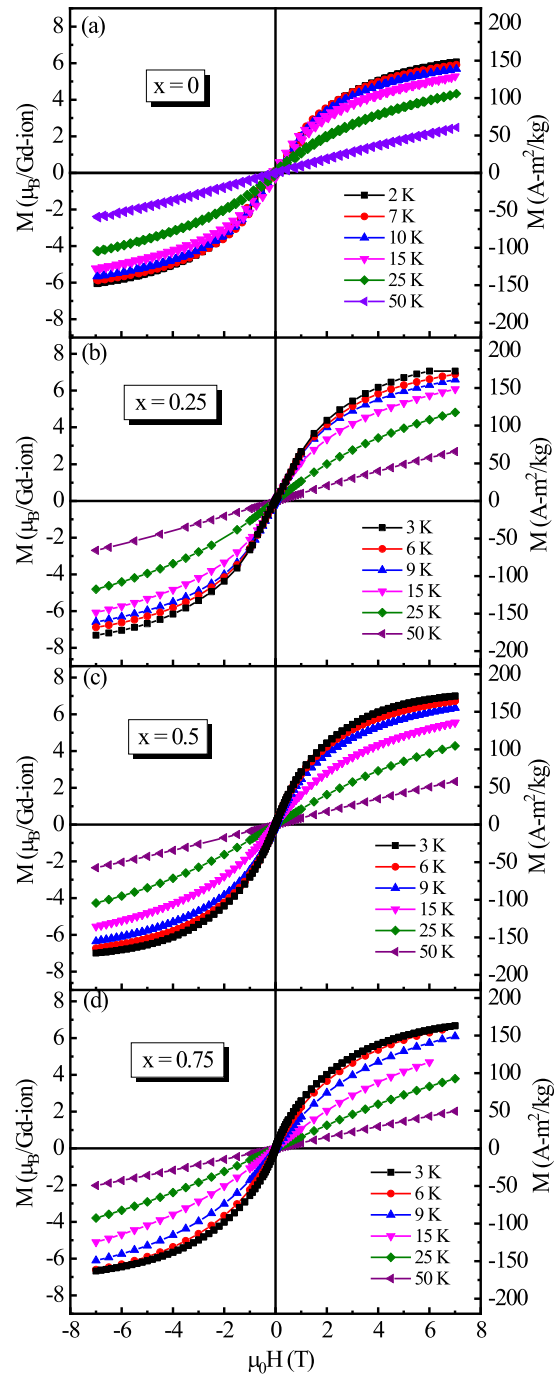


Fig. 4. Magnetic field evolution of isothermal magnetization at different temperatures for (a) $x = 0$, (b) $x = 0.25$, (c) $x = 0.5$, and (d) $x = 0.75$ Y substituting concentrations.

materials in the cryogenic temperature region, viz., GdCo₂B₂ (83.7 J/kg) [39], Dy_{0.9}Tm_{0.1}Ni₂B₂C (~ 44 J/kg) [40], ErNi₂ (39 J/kg) [41], TmZnAl (53 J/kg) [42], Ho₃Pd₂ (69.2 J/kg) [43], Nd(Co_{0.8}Fe_{0.2})₂ (77.9 J/kg) [44], etc. The RCP values for all the Y substituted compounds are less than that achieved for magnetically undiluted Gd₂NiSi₃. Thus, the effect of Y substitution found to be responsible for considerable change in magnetocaloric properties, particularly in determining the refrigerant capacity of the systems.

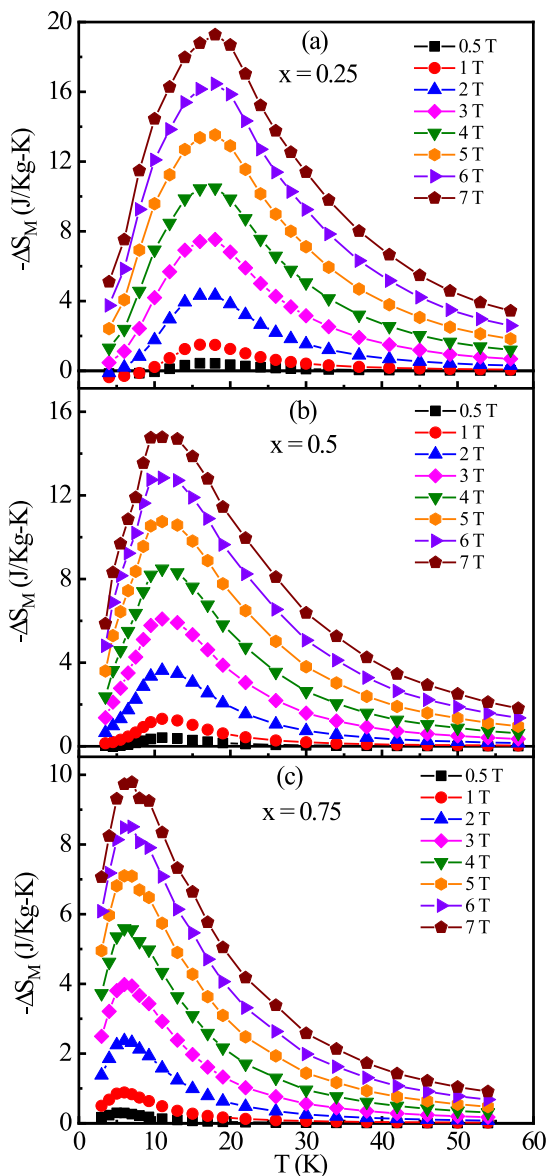


Fig. 5. Isothermal magnetic entropy change ($-\Delta S_M$) as a function of temperature for different magnetic field change for (a) $x = 0.25$ (b) $x = 0.5$ and (c) $x = 0.75$ compositions. The data for $x = 0$ sample had been presented earlier in Ref. [4].

Table 2
The maximum isothermal magnetic entropy change ($-\Delta S_M^{\text{max}}$) and RCP under the magnetic field change of 2, 5 and 7 T for $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ compounds.

x	$-\Delta S_M^{\text{max}}$ (J/kg-K)			RCP (J/kg)		
	2 T	5 T	7 T	2 T	5 T	7 T
0	4.08	12.9	18.4	66	316	525
0.25	4.3	13.5	19.3	66	311	503
0.5	3.7	10.8	14.8	49	222	346
0.75	2.4	7.1	9.8	25	104	171

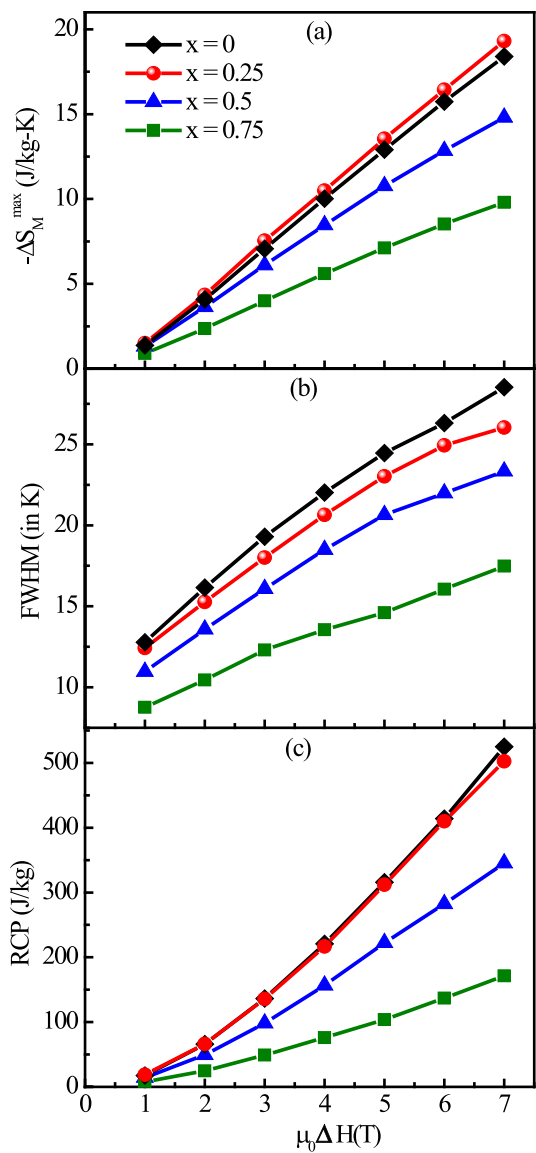


Fig. 6. (a) Maximum isothermal magnetic entropy change ($-\Delta S_M^{\text{max}}$) (b) FWHM of $-\Delta S_M(T)$ curve and (c) RCP of all the partially yttrium substituted compounds along with non-substituted reference Gd_2NiSi_3 system as a function of applied magnetic field change.

3.1. Summary

In summary, we have studied the structural, magnetic and magnetocaloric properties of partially Y substituted $(\text{Gd}_{1-x}\text{Y}_x)_2\text{NiSi}_3$ ($x = 0.25, 0.5, 0.75$) compounds by measuring XRD, dc magnetic susceptibility, isothermal magnetization and magnetic entropy change. It was found that lattice parameter of the compounds decrease with the increase of Y concentration. All of these materials commonly exhibit an anti-ferromagnetic phase transition at lower temperatures with an additional ferromagnetic-like contribution. Thermomagnetic irreversibility is also evidenced for all the compounds below their phase transition temperatures. The transition temperatures (11.5, 7.2, and 3.1 K for $x = 0.25, 0.5$ and 0.75 compounds) and the amount of ferromagnetic

coupling gets weakened with the magnetic dilution by increasing the Y concentration. The magnetization as well as the extent of short range magnetic correlation in the paramagnetic region decreases with increase in Y content. The magnetic entropy change and associated relative cooling power have been estimated for all the compositions. The maximum value of isothermal entropy change of 19.3 J/kg-K, 14.8 J/kg-K and 9.8 J/kg-K are observed for $Gd_{1.5}Y_{0.5}NiSi_3$ compound with relative cooling power ~ 503 J/kg, 311 J/kg, and 66 J/kg for 7 T, 5 T, and 2 T magnetic field changes, respectively. These magnetocaloric properties of $(Gd_{1-x}Y_x)_2NiSi_3$ compounds may find attraction as magnetic refrigerating materials in the cryogenic temperature region.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <https://doi.org/10.1016/j.jmmm.2019.04.021>.

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