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Magnetic and Magnetocaloric Properties of $Pr_2Fe_{17-x}Cu_x$ Intermetallic Compounds

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In this study, Cu-doped intermetallic compounds with the chemical composition of $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) were prepared by the arc-melting process. SEM analysis revealed that the samples consist of irregularly shaped and strongly connected micron-scale particles range from 0.5 μm to 4 μm. EDX analysis showed that all the samples consist of a single phase. The magnetic measurements revealed that all the samples exhibit a ferromagnetic to the paramagnetic phase transition. The Curie temperature was determined to increase linearly with increasing Cu substitution from 286 K for x = 0 to 303 K for x = 0.2. The maximum value of magnetic entropy change |ΔSm| was estimated from isothermal magnetization curves and it decreases with increasing Cu content from 5.63 J/kg·K for x = 0 to 4.21 J/kg·K for x = 0.2 at 5 T magnetic field change. At the same magnetic field change, the relative cooling power values range from 404.8 J/Kg for x = 0 to 248.3 J/Kg for x = 0.2. Arrott plots and Landau analyses revealed that all the samples show second-order magnetic phase transition.

Keywords : 2:17 type intermetallic compounds, Cu substitution, magnetic entropy change, magnetocaloric effect

1. Introduction

The magnetocaloric effect (MCE) is an intrinsic property to magnetic materials and defined as the temperature change of a magnetic material associated with an external magnetic field change. In recent years, magnetocaloric materials have been intensively studied due to the possible practical application in magnetic refrigeration instead of conventional vapor-cycle refrigeration [1, 2]. In recent years, the majority of the studies on the magnetocaloric effect have focused on finding the most suitable magnetocaloric materials as room temperature magnetic refrigerants. In the early stages of studies, due to the large magnetic entropy changes, most of the attentions have been paid to materials with first-order magnetic phase transition [3, 4]. Nowadays, it has been understanding that not only the large magnetic entropy change for high cooling efficiency but also the refrigeration capacity (RC), which is a key parameter for the application of a material as a refrigeration material, must also be considered [5, 6]. Recent studies have verified that materials with second-order magnetic phase transitions also show relatively large magnetic entropy change over a wide temperature range, which is important for high RC [7, 8].

In recent years, among the various magnetocaloric materials with second-order phase transition, Pr_2Fe_{17} intermetallic compounds have attracted interest as magnetocaloric materials based on their large magnetization values, large MCE close to room temperature, relatively more broader magnetic entropy peak and low production cost due to their high Fe content [9-25]. All these properties make these compounds promising materials for room temperature magnetic cooling technology. Previous investigations have shown that Pr_2Fe_{17} crystallizes in the rhombohedral Th_2Zn_{17}-type crystal structure and exhibits ferromagnetic order with relatively high values for the saturation magnetization and a Curie temperature nearly at 285 K [10, 26]. Although Pr_2Fe_{17} compounds possess high saturation magnetization, the relatively low Curie temperature (T_c) limits their use as a magnetic refrigerant at room temperature and above. Therefore, most of the previous works have focused on improving magnetocaloric properties and the Curie temperature towards to room temperature. In this context, many studies have been done to improve the magnetocaloric properties and the Curie temperature by replacing the Fe atoms with many other atoms such as Al, Si, Ga, V, Mo, Nb, W, Zr, Co, Ni, Cr, Mn, Ni [9, 11, 13, 16, 25, 27, 28]. Dan [9] *et al.* have reported that $Pr_2Fe_{16}Si$ compound possesses high relative cooling power (RCP) (87 J/kg at 1.5 T magnetic

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field change), high operation temperature (390 K) and moderate magnetic entropy change (1.61 J/kg·K at 1.5 T magnetic field change) with second-order magnetic phase transition. Pospisil [11] *et al.* have concluded the Cr doping effect in $Pr_2Fe_{17-x}Cr_x$ compounds and found that Cr yields enhancement of the Curie temperature by 50 K. The maximum ΔS_m value for $Pr_2Fe_{17-x}Cr_x$ was reported to increase with Cr content. Zhong [13] *et al.* have investigated the magnetocaloric properties of $Pr_2Fe_{17-x}Mn_x$ compound and reported that the Curie temperature increases from 283 K at x = 0 to 294 K at x = 1, and then decreases to 285 K at x = 2. Magnetic entropy change has reported to decrease with Mn content. The refrigeration capacity (RC) has reported to range from 458 to 364 J/kg with typical second-order phase transition. Guetari [16] *et al.* has reported for $Pr_2Fe_{17-x}Al_x$ that Curie temperature increases with the Al content. The magnetic entropy change has determined to decrease with the increasing Al content from 5.5 J/kg·K (x = 0) to 4.2 J/kg·K (x = 1) at 5T. As can be seen from reviewed results, although R_2Fe_{17} compounds do not have a significantly large magnetic entropy change, comparatively high RC values make these compounds advantageous from the point of view of application [9, 29]. Moreover, these compounds also show negative thermal expansion below T_c due to the magnetovolume effect. The previous studies have shown that the substitution of Fe atoms by magnetically weaker or non-magnetic atoms reduces the value of the negative thermal expansion coefficient by weakening the magnetovolume effect, resulting an increase of T_c [30, 31]. In the present work, the effect of the substitution of Cu for Fe on magnetic and magnetocaloric properties in $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) intermetallic compounds have been investigated.

2. Experimental

$Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) intermetallic compounds have been prepared by arc melting of high purity elements Fe, Pr and Cu in a water-cooled Cu mould under a high-purity argon atmosphere. In order to offset the losses due to evaporation of Pr, 15 wt% excess of Pr was added to the each one sample composition. All ingots of the polycrystalline alloys were re-melted four times with being turned over each time to ensure homogeneity the compositions. The resulting buttons were sealed in a quartz tube under high vacuum and annealed at 1323 K for 1 week to homogenize the materials. After the annealing process the alloys were rapidly quenched in iced water.

Grain structure and micro structural analysis was observed using a LEO-EVO-40 scanning electron microscope (SEM) system attached to a Röntec 3000 detector and an energy-dispersive X-ray (EDX) system. Color mapping and phase formation results were investigated using electron probe microscopy analysis (EPMA) results from the same detector. The magnetic measurements were performed using a PPMS9 (Quantum Design) in a temperature range 100-350 K and 6 T maximum magnetic field was applied.

3. Results and Discussion

Fig. 1 shows the X-ray diffraction patterns of $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at room temperature. In order to determine the lattice parameters and existing phases, the experimental X-ray diffraction patterns were compared with the simulated patterns by Rietveld refinement method using GSAS-II software. The Rietveld analysis of the X-

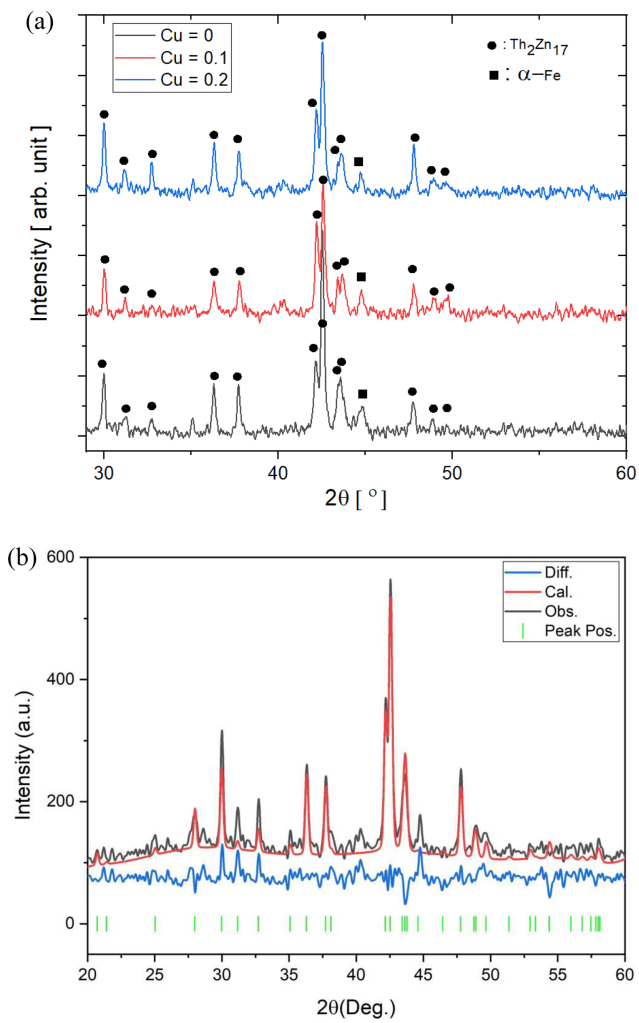


Fig. 1. (Color online) (a) X-ray diffraction patterns of the $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at room Temperature, (b) Results of Rietveld refinement for x = 0.2.

Table 1. Lattice parameters, Curie temperature (T_c), magnetization value (M) determined at 6 T magnetic field and 260 K, maximum magnetic entropy change ($|\Delta S_m|$) and relative cooling power (RCP).

	a = b (Å)	c (Å)	V (Å^3)	T_c (K)	M (emu/g)	$\|\Delta S_m\|$ (J/kg·K)	RCP (J/Kg)
x = 0	8.584	12.462	795.347	286	118.8	5.63	404.8
x = 0.1	8.583	12.458	794.937	289	126.6	5.28	373.6
x = 0.2	8.565	12.435	790.080	303	134	4.21	288.3

ray diffraction has indicated that all the compounds have nearly single rhombohedral Th_2Zn_{17} type structure with R-3m space group. Additionally, a small amount of secondary α-Fe phase has also been detected. We have tried to determine the percentage of α-Fe phase using peak analysis of XRD spectrum and results showed nearly all samples have less 5 % of α-Fe phase. The determined lattice parameters were listed in Table 1. It is clearly seen that lattice parameters (*a*, c and V) slightly decrease with increasing Cu content. The similar behavior has also observed in some Th_2Zn_{17} type rhombohedral compounds [28, 33].

Fig. 2 shows the typical SEM imagines for $Pr_2Fe_{16.8}Cu_{0.2}$ sample with magnifications of 5000 and 20000. The sample is composed of irregularly shaped and strongly connected micron sized grains with a broad size distribution. The SEM picture showed that the grains were dispersed randomly and most of the grains seem to be in the range from 0.5 µm to 4 µm. In order to get a deeper insight in the structure of the sample, EDX analyses have been also performed. Fig. 3 shows the EDX analysis of $Pr_2Fe_{16.8}Cu_{0.2}$ sample. The element percentage distribution has showed only Fe, Pr and Cu elements exist no other impurity elements were observed. Further microstructural analysis using a color mapping technique was also carried out. Color mapping of $Pr_2Fe_{16.8}Cu_{0.2}$ sample is shown in Fig. 4. The top picture clearly shows the uniform distribution of the constituent elements. In color mapping, Fe was represented by red, Pr by green and Cu by blue. All the results the small pictures show the distribution of each element in the picture: a dark area shows that the element represented does not exist or is present only in trace amounts.

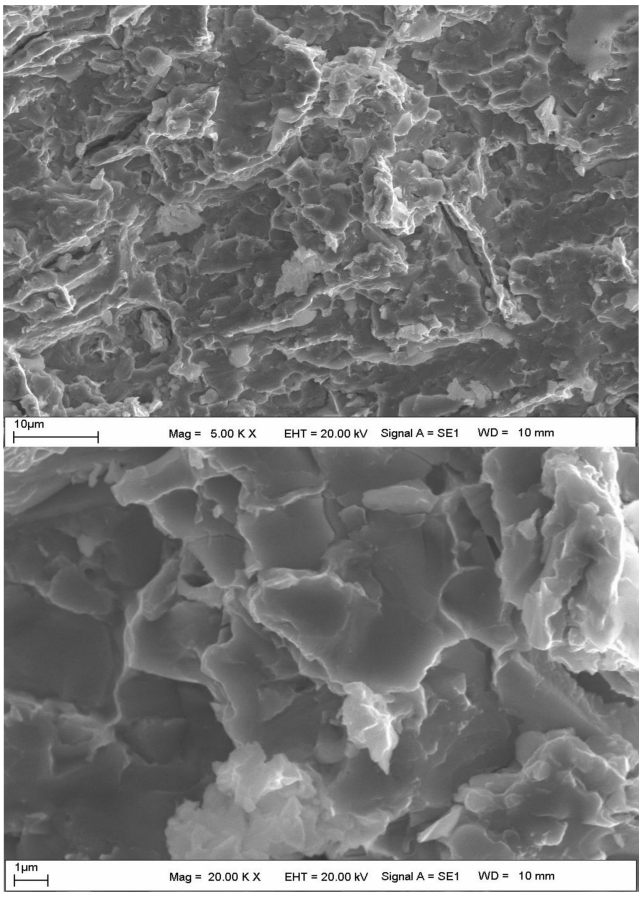
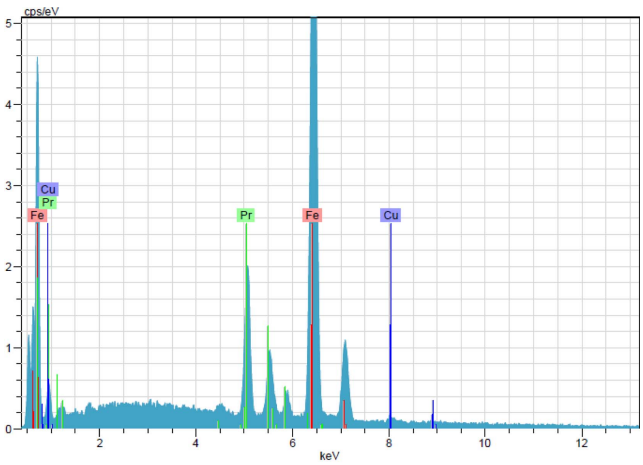


Fig. 2. The typical SEM imagines for $Pr_2Fe_{16.8}Cu_{0.2}$ sample with magnifications of 5000 and 20000.



El	AN	Series	unn. C [wt.-%]	norm. C [wt.-%]	Atom. C [at.-%]	Error [%]
Fe	26	K-series	73.37	74.52	86.87	2.0
Pr	59	L-series	22.72	23.07	10.66	0.7
Cu	29	K-series	2.37	2.41	2.47	0.1
		Total:	98.46	100.00	100.00	

Fig. 3. (Color online) EDX of $Pr_2Fe_{16.8}Cu_{0.2}$ sample.

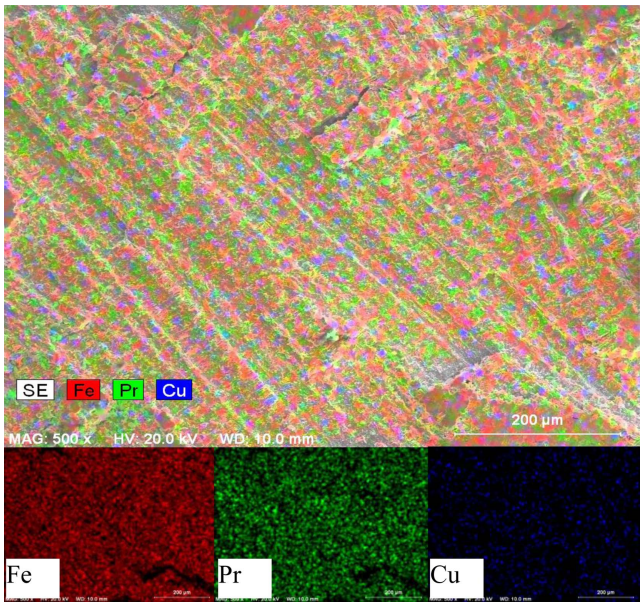


Fig. 4. (Color online) Color mapping of the $Pr_2Fe_{16.8}Cu_{0.2}$ sample.

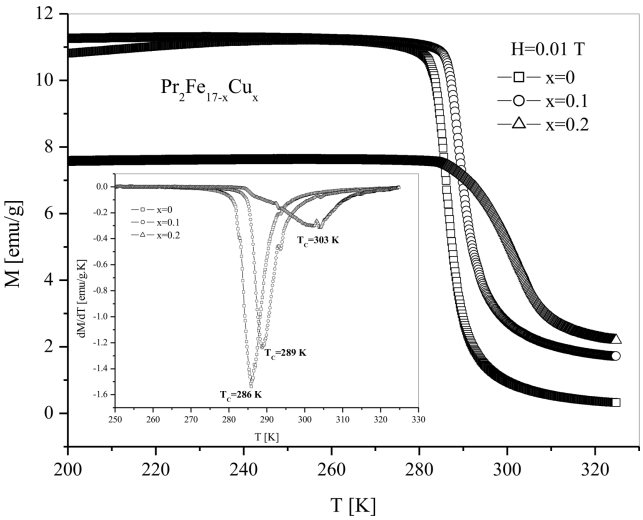


Fig. 5. Temperature dependence of magnetization for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at 0.01 T. The inset presents the dM/dT curves.

Fig. 5 shows the temperature dependence of magnetization for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds. All data were taken under an applied field of 0.01 T. As can be seen all the samples exhibit a paramagnetic to ferromagnetic phase transition. The Curie temperature, T_c, can be defined as the temperature of maximum value of |dM/dT| as shown inset in Fig. 5. The determined Curie temperatures were given in Table 1. For x = 0 sample, we have found a T_c equal to 286 K. For the further Cu concentrations, T_c was observed to increase with increasing x. The value of T_c for x = 0 and the variation of T_c with increasing Cu content is in agreement with the literature [10, 12, 16, 17]. As clearly seen in Fig. 5, while the x = 0 and 0.1 samples exhibit a narrow minimum in the the |dM/dT| vs T curves, for x = 0.2 sample, the curve becomes more broaden. Such a similar behavior in |dM/dT| curves of bulk and ball milled Pr_2Fe_{17} samples was interpreted by Llamazares [10] that the ferro-to-paramagnetic transition is not well defined due to the structural disorder so we assume that observed anomaly in the |dM/dT| curve for x = 0.2 could be due to the structural disorder.

As can be seen in Table 1, T_c has observed to increase from 286 K for x = 0 to 303 K for x = 0.2. The value of Curie temperature is closely related with the ferromagnetic exchange interaction. In such intermetallic compounds (Pr_2Fe_{17}), magnetic interactions, consequently the Curie temperature is determined by three different exchange interactions: the Pr-Pr (4f-4f) exchange interaction between the magnetic moments of rare earth sublattice, the intersublattice Pr-Fe (4f-3d) exchange interaction between the magnetic moments of rare earth sublattice and magnetic Fe sublattice, the Fe-Fe (3d-3d) exchange interaction between magnetic Fe moment sublattices [16]. The Pr-Pr interactions are in nearly negligible due to the localized character of the electrons. The Pr-Fe exchange interaction has also relatively small value and moreover it hardly varies with doping concentration [32]. As a result, it could be inferred that Pr-Pr and Pr-Fe interactions have almost no effect on T_c. That means for intermetallic compounds, the Fe-Fe interaction is dominant and the magnetic behaviors is mainly determined by the Fe-Fe sublattice. The nature of magnetic interaction of Fe magnetic moments depends on the Fe-Fe interatomic distance. For distance lower than 2.45 Å, the exchange interaction favors an antiferromagnetic alignment of the magnetic moments, while for longer distances a ferromagnetic alignment is preferred [30, 31, 33, 34]. As mentioned above, due to the decrease in unit cell volume with increasing Cu content, the increase in T_c could not be attributed to increase in the unit cell volume and in the Fe-Fe interatomic distance. In this case, the increase in T_c is most likely due to the competition of ferromagnetic and antiferromagnetic exchange interaction between Fe-Fe atoms. As discussed frequently in many studies, in rhombohedral R_2Fe_{17} type intermetallic compounds, the Fe(6c)-Fe(6c) bond length is the shortest and the interaction between the Fe(6c) atoms is antiferromagnetic [9-34]. In the present case, the doped Cu atoms are most likely substituted with the Fe (6c) atoms and consequently the strength of the antiferromagnetic interaction between two Fe(6c) atoms reduces with increasing Cu content. In this case, ferromagnetic Fe-Fe exchange interaction at crystallo-

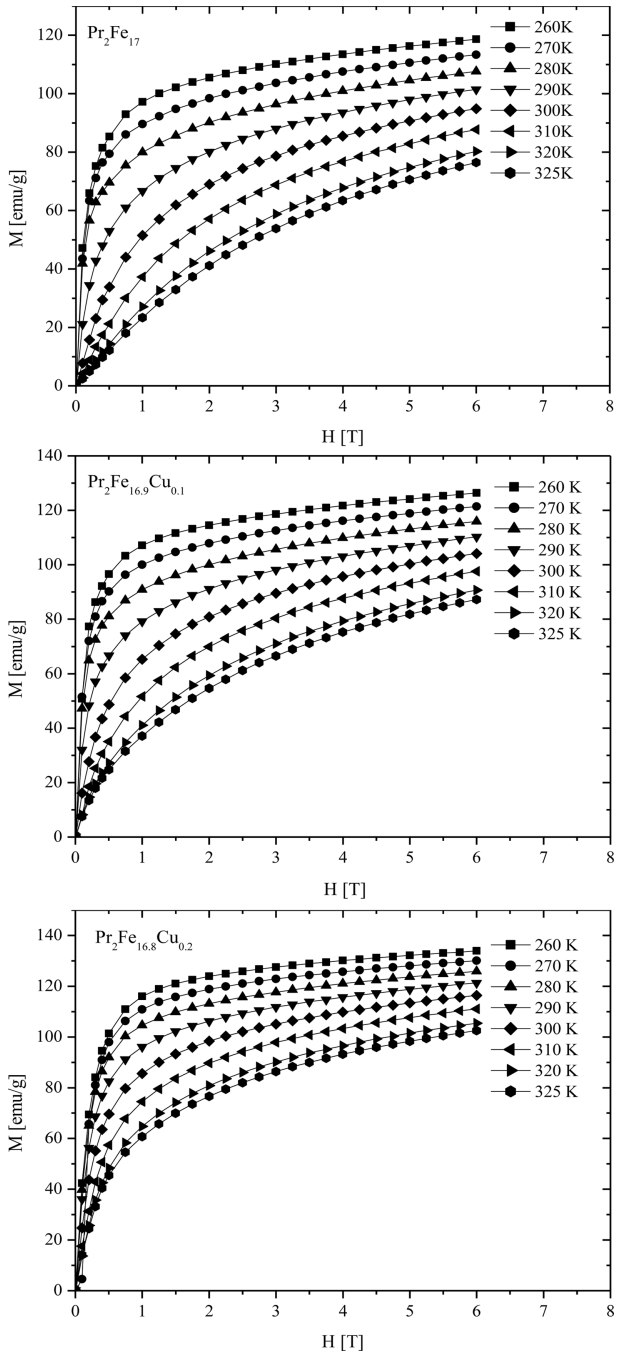


Fig. 6. Isothermal magnetization as a function of applied field up to 6 T for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at various temperatures.

graphic sites of 9d, 18*f* and 18*h* becomes more dominant, leading to an increase in Curie temperature.

Fig. 6 shows the isothermal magnetization as a function of applied field up to 6 T for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at various temperatures. The magnetization isotherms for Pr_2Fe_{17} compound show typical ferromagnetic behavior. Below the Curie temperature, the magnetization curves increase sharply and saturated at nearly 1 T magnetic field. Above the Curie temperature, magnetization curves increase slowly with the applied magnetic field which is a typical signature of the paramagnetic state.

The magnetization curves of Cu doped samples ($Pr_2Fe_{16.9}Cu_{0.1}$, $Pr_2Fe_{16.8}Cu_{0.2}$) also exhibit typical ferromagnetic behavior. But, the saturation is achieved at lower than 1 T magnetic field. This means that Cu substitution with Fe promotes ferromagnetic interactions. The maximum magnetization value determined from magnetization curves at 6 T magnetic field and at 260 K for $Pr_2Fe_{17-x}Cu_x$ samples has increased from 118.8 emu/g for x = 0 to 134 emu/g for x = 0.2. The determined magnetization values were given in Table 1. The results are in agreement with that of $Pr_2Fe_{17-x}Al_x$ [16]. As discussed by Guetari [16], the magnetization values of Al doped compound generally result from competition between the magnetovolume effect caused by the expansion of the unit cell volume and magnetic dilution caused by nonmagnetic element. At low concentrations, the magnetovolume effect is more dominant and responsible in increase of magnetization values well as the increase of T_c as in present $Pr_2Fe_{17-x}Cu_x$ compound. The decrease of magnetization at higher Al concentration (x > 0.75) rate in $Pr_2Fe_{17-x}Al_x$ compound has been attributed to magnetic dilution [16]. For maximum Cu concentration rate of x = 0.2 in present $Pr_2Fe_{17-x}Cu_x$ compound, the magnetic dilution is not expected to be dominant.

The magnetic entropy change, which is associated with the magnetocaloric effect, can be calculated from the isothermal magnetization curves (Fig. 6) under the influence of a magnetic field. According to the classical thermodynamic theory, the magnetic entropy change ΔS_m produced by the variation of a magnetic field from 0 to H_{max} is given by,

$$\Delta S_m(T,H) = \int_0^{H_{max}} \left(\frac{\partial M}{\partial T}\right)_H dH \qquad (1)$$

By using Eq. (1), the magnetic entropy change was calculated from isothermal magnetization curves (Fig. 5). Fig. 7 shows the magnetic entropy change for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at various magnetic fields. In all the samples a peak of $|\Delta S_m|$ around the transition temperature was found. The peak value and peak temperature increase with increasing magnetic field. The results were summarized in Table 1. The largest magnetic entropy change has been observed to be 5.63 J/kg·K at 5 T magnetic field change for Pr_2Fe_{17} sample and the value of $|\Delta S_m|$ for Pr_2Fe_{17} is in agreement with literature [13, 16,

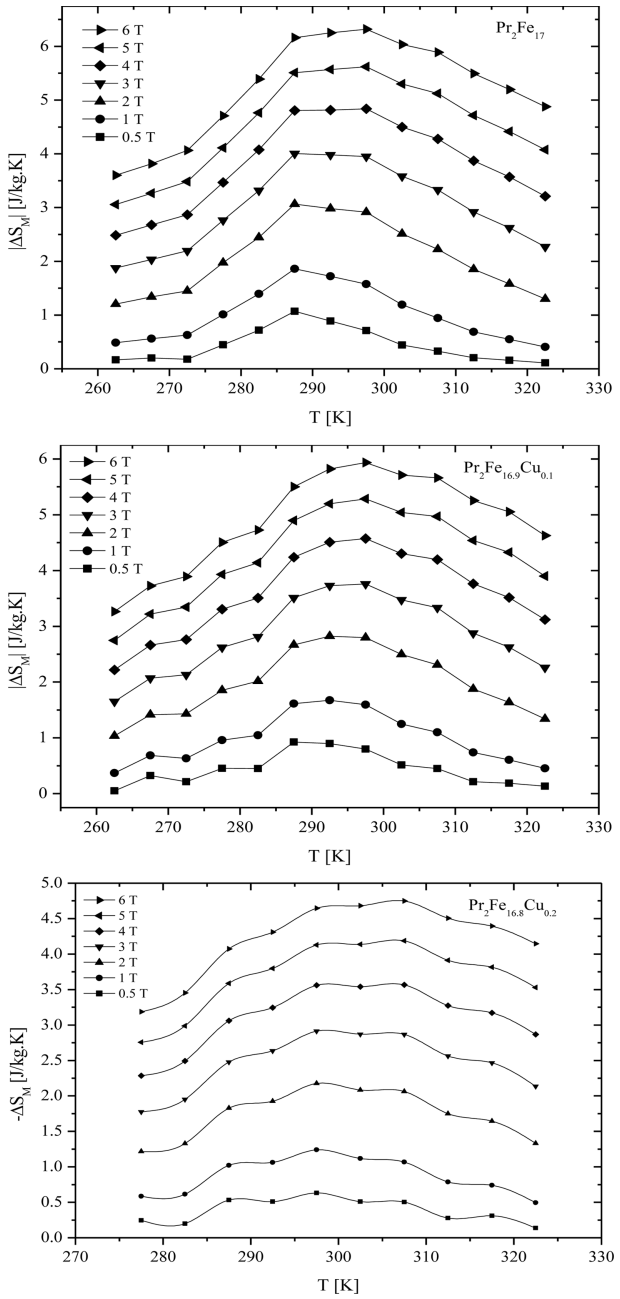


Fig. 7. Temperature dependence of magnetic entropy change at various magnetic fields for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds.

17]. Fig. 8 shows the magnetic entropy change for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1 and 0.2), compounds at 1 and 5 T magnetic field change. The decrease in peak value of $|\Delta S_m|$ with increasing Cu content is clearly seen. The same situation was also observed for very similar $Pr_2Fe_{1-x}Al_x$ compound. While the saturation magnetisation slightly increase, the decrease in |ΔSm| was discussed by Guetari [16]. It has ben concuded that the maximum change of magnetic entropy, |ΔSm|, depends not only on the saturation

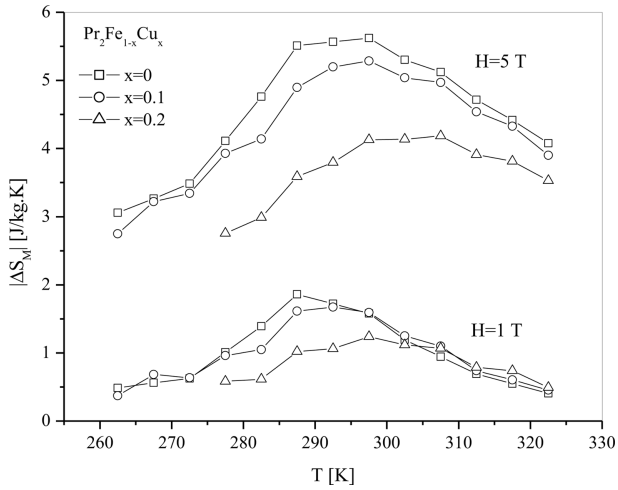


Fig. 8. Magnetic entropy change for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at 1 and 5 T magnetic fields.

magnetization, M_S, but depends also on the difference $\Delta M_T(H)$ between two consecutive isotherms at a given magnetic field around the transition temperature. This may explain the slowly decrease of |ΔSm|, although, M_S increases slightly with Al content. In addition to above mentioned discussion, the decrease of the magnetic entropy change with Cu content could be due to the magnetic inhomogeneity existing in sample, which causes a more slower magnetic transition (one of the key parameters that determine the value of magnetic entropy change) at transition temperature. Nevertheless, values of $|\Delta S_m|$ for Cu doped samples are moderate compared to literature [3-8]. Although the magnetic entropy value decreases with Cu doping, the temperature range in which the entropy change remains at its maximum is quite high. As can be clearly seen from Fig. 7 and Fig. 8, while the magnetic entropy change has decreased around 25 % with increasing Cu content to x = 0.2, the temperature range at half maximum has decreased only around 5 %. This large temperature interval (average 70 K) is a great advantage for magnetic cooling applications.

For the practical application of the magnetocaloric effect in magnetic cooling, not only the maximum value of $|\Delta S_m|$ but also the temperature range in which $|\Delta S_m|$ remains the maximum is important. Magnitude of this temperature range determines the value of one of the critical criterion called as relative cooling power (RCP) which evaluates the refrigeration efficiency The RCP measures the amount of heat transferred from a hot reservoir to cold reservoir in a thermodynamic cycle. In this context, RCP was calculated by taking the direct product of the maximum entropy change and the full width at half maximum of the peak ($=|\Delta S_m|\cdot\delta T_{FWHM}$). The

estimated RCP values at 5 T magnetic field change were listed in Table 1.

The RCP values for $Pr_2Fe_{17-x}Cu_x$ have been determined to be 404.8, 373.6 and 288.3 J/kg for x = 0, 0.1 and 0.2 respectively at 5 T magnetic field change. Although the Cu doped samples have relatively low magnetic entropy changes, the RCP values are larger due to the wider magnetic entropy curves. The magnetocaloric propeties of produced samples are comparable or greater than those of similar compounds as $Pr_2Fe_{17-x}Mn_x$ [13] (ΔS_m = 5.66-4.31 J/kg·K, RCP = 458-364 J/kg), $Pr_{1.5}Ce_{0.5}Fe_{17}$ [14] (ΔS_m = 5.3 J/kg·K, RCP = 238 J/kg), $Pr_2Fe_{17-x}Al_x$ [16] (ΔS_m = 5.5-4.2 J/kg·K, RCP = 540-375 J/kg), $Pr_2Fe_{16}Si$ [9] (ΔS_m = 1.61 J/kg·K, RCP = 87 J/kg) and the other magnetocaloric materials as $La_{0.67}Sr_{0.33}MnO_3$ [7] (ΔS_m= 5.15 J/kg·K, RCP = 252 J/kg), $Gd_5Ge_2Si_2$ [7] (ΔS_m = 18.5 J/kg·K, RCP = 425 J/kg), $MnFeP_{0.45}As_{0.5}$ [7] (ΔS_m = 14.5 J/kg·K, RCP = 188 J/kg). Sufficiently high RCP values and higher broader magnetic entropy peak allow operation over a very wide temperature interval (average 70 K) for magnetic cooling applications. One of the most important points is the substitution of Fe atoms by magnetically weaker or non-magnetic atoms reduces the value of the negative thermal expansion coefficient by weakening the magnetovolume effect, resulting an increase of Tc. This situation allows the Tc to be adjusted by Cu doping. In addition, the use of Fe and Cu in material production provides a significant advantage in terms of application as it significantly reduces production costs.

Generally such a broad magnetic entropy change (ΔS_m) and relatively large RCP values are associated with the second-order magnetic phase transition of materials. To ensure, the nature of phase transition of samples could be investigated by using Landau theory. The magnetic free energy F(M,T) can be expanded as a Landau expansion in the powers of the magnetization M, neglecting higher order contributions as

$$F(M,T) = \frac{c_1(T)}{2} M^2 + \frac{c_3(T)}{4} M^4 - M\,H \qquad (2)$$

Here, $c_1(T)$ and $c_3(T)$ are called as Landau coefficients and the sign of the $c_3(T)$ of M^4 term determines the type of the magnetic phase transition. The first-order phase transition is expected in the case $c_3(T) < 0$ and the second-order phase transition in the case $c_3(T) > 0$. From the condition of equilibrium $\partial F(M, T)/\partial M = 0$, we obtain

$$H = c_1(T)M + c_3(T)M^3 \qquad (3a)$$

$$\frac{H}{M} = c_1(T) + c_3(T)M^2 \qquad (3b)$$

In order to get a deeper insight in the nature of magnetic phase transition, M^2 versus H/M, which is called as Arrott plot, was plotted in a critical temperature interval. The slope of the resulting curves denotes whether a magnetic transition is of first-order or second-order. According to the Banerjee criterion [35], the positive slope of H/M vs M^2 curves indicates a second-order phase transition. On the other hand, if several of the curves show a negative slope at some point, then the magnetic

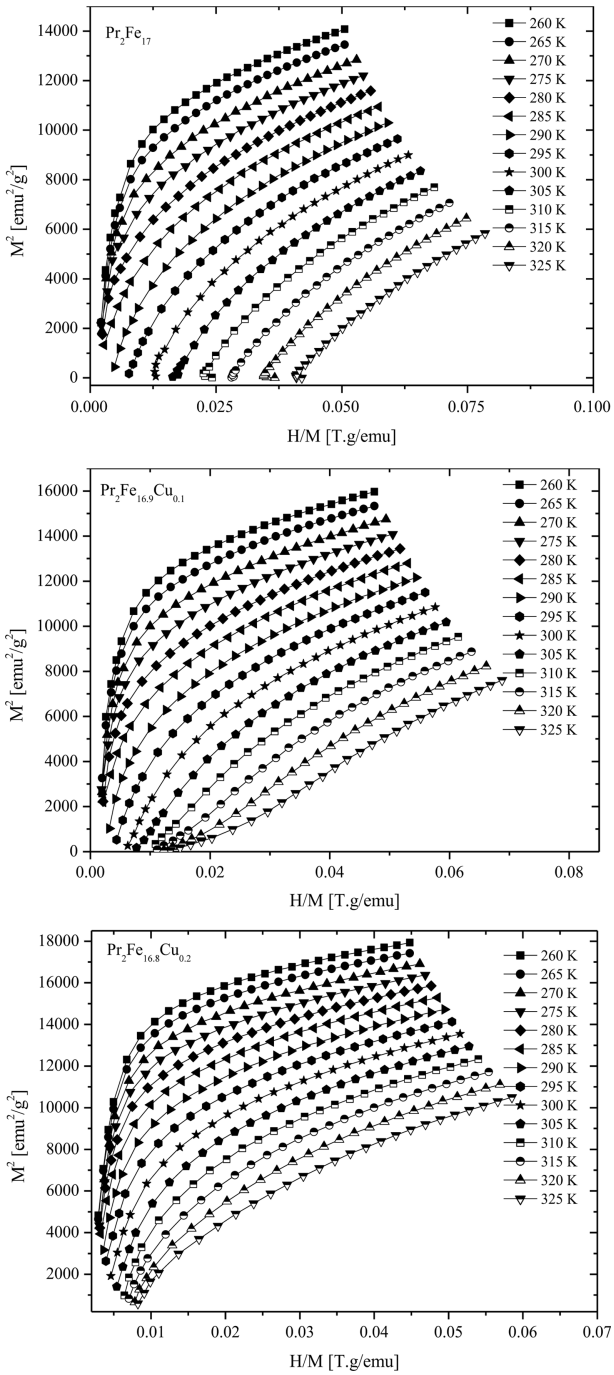


Fig. 9. Arrott plots of $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds at various temperatures.

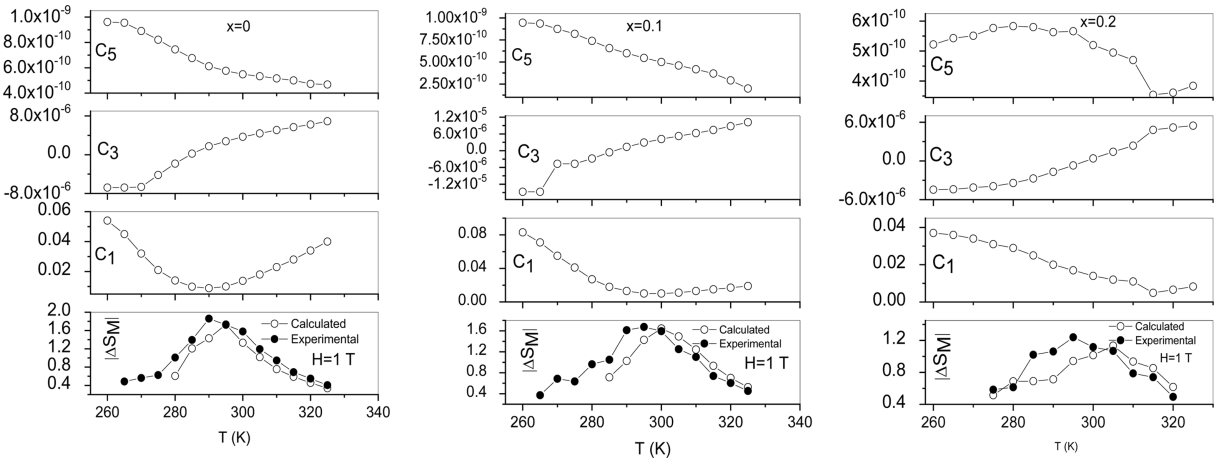


Fig. 10. The temperature dependence of c_1 (T.g/emu), c_3 (T.g^3/emu^3) and c_5 (T.g^5/emu^5) and experimental and theore tical magnetic entropy change for x = 0, 0.1 and 0.2 samples at 1 T magnetic field.

transition is first-order. Fig. 9 shows the Arrott plots for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds. As shown in Fig. 9, all the curves for samples show a positive slope confirming the second-order phase transition. The Landau coefficients c_1(T), c_3(T) and c_5(T) could be determined by fitting Eq. (3a) to magnetic field H against magnetization M (Fig. 6) or fitting Eq. (3b) to Arrott plots (Fig. 9). The temperature dependence of the Landau coefficients determined from above mentioned fits is shown in Fig. 10 for $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) compounds. As can be seen in Fig. 10, the value of c_3(T) at Tc is positive for three samples indicating that the magnetic phase transitions are second-order.

By differentiation of the magnetic part of the magnetic free energy with respect to temperature, the magnetic entropy change is obtained,

$$\Delta S_m(T,H) = -\frac{1}{2}\frac{\partial c_1}{\partial T}M^2 - \frac{1}{4}\frac{\partial c_3}{\partial T}M^4 - \frac{1}{6}\frac{\partial c_5}{\partial T}M^6 \qquad (4)$$

c_1(T), c_3(T) and c_5(T) parameters are given in Fig. 10 and their derivative in Eq. (4), the magnetic entropy change was calculated theoretically under a magnetic field change of 1 T for x = 0, 0.1 and 0.2 samples. Fig. 10 shows the calculated and experimental magnetic entropy changes for x = 0, 0.1 and 0.2 samples at 1 T magnetic field. As can be seen in Fig. 10, while the agreement between the experimental and theoretical data for three samples is satisfactory at above the Curie temperature, there is a small deviation between the experimental and theoretical data at lower temperatures for x = 0.2 sample. As concluded in literature, good agreement between theoretical and experimental results demonstrates that the magnetoelastic coupling, which is interaction between the magnetization and the strain of a magnetic material, and electron interaction have strong influence in determining the magnetocaloric effect [36, 37]. Otherwise, when a small deviation is observed between the experimental and theoretical data as in x = 0.2 sample, it has been concluded that the magnetoelastic coupling and electron interaction do not contribute directly to the magnetic entropy. This small deviation observed between the experimental and theoretical data is generally attributed to the existence of mixed phases at lower temperatures [38].

4. Conclusions

In this study, Cu doped intermetallic compounds with chemical composition of $Pr_2Fe_{17-x}Cu_x$ (x = 0, 0.1, 0.2) were prepared by arc-melting process. SEM analysis revealed that the samples consist of irregularly shaped and strongly connected micronic particles range from 0.5 μm to 4 μm. EDX analysis showed that all the sample consist of single phase. No any impurity phase was observed Magnetic and magnetocaloric properties of the samples were systematically investigated. The study of magnetic properties reveals that all the samples exhibit a ferromagnetic to paramagnetic phase transition. The value of Curie temperatures estimated from the temperature of maximum value of |dM/dT| are 286, 289 and 303 K for x = 0, 0.1 and 0.2 respectively. The increase of T_c in $Pr_2Fe_{17-x}Cu_x$ compounds with increasing Cu content was attributed to cell volume expansion. The maximum value of magnetic entropy change $|\Delta S_m|$ was estimated from isothermal magnetization curves and it decreases with increasing Cu content from 5.63 J/kg·K for x = 0 to 4.21 J/kg·K for x = 0.2 at 5 T magnetic field change. At same magnetic field change RCP values range from 404.8 J/Kg for x = 0 to 248.3 J/Kg for x = 0.2. Arrott plots and

Landau analyses revealed that all the samples show second-order magnetic phase transition. Relatively broad magnetic entropy peaks at a large temperature interval, large RCP values, the operation near room temperature and the relatively low production cost due to the low raw-materials (Fe) price make these intermetallic compounds potential candidates for room temperature magnetic refrigerants.

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