218REVERSIBLE LOGIC SYNTHESIS OF FAULT TOLERANT CARRY SKIP BCD A VARIETY OF MAGNETISM IN CuMPt₆ (M=3d TRANSITION METALS) TERNARY ALLOYS

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ABSTRACT

The magnetic properties of polycrystalline CuMPt₆ (M= Ti, V, Cr, Mn, Fe, Co and Ni) ternary alloys were studied by Superconducting Quantum Interference Device (SQUID) over the temperature range 4-300 K. Magnetization measurements revealed that the compounds containing M=Ti, V and Ni are paramagnetic. A spin glass type magnetic behavior appeared in the compounds with M= Cr and Mn in their annealed state and the corresponding temperatures for freezing the magnetic moments were T_g = 15 and 40 K, respectively. Positive values of the paramagnetic Curie temperature (Q) indicated that the ferromagnetic interaction is dominant in these spin-glass compounds. The compounds with M= Fe and Co showed ferromagnetic behavior having Curie temperatures (T_c) = 230, and 190 K, respectively. Hysteresis study showed that the alloy with M=Co was magnetically softer than that with M=Fe.

Keywords: CuMPt₆ ternary alloys, Magnetic behavior, Effective magnetic moments (P_{eff}), Spin glass type.

1. INTRODUCTION

Magnetic materials alloyed with 3d-transition metals attracted world-wide scientific and technical interest both for their unique physical properties and for constantly increasing number and variety of their recent applications as sensors, attenuators, storage and control systems, etc. [1, 2]. It is well known that 3d electrons are responsible for the magnetic moment of the transition elements and partake in the cohesion of transition metals. The correlation between the structure and magnetism of 3d transition-metal alloys has been an important topic in condensed matter physics. Different chemical compositions of an element in an alloy might lead to various crystallographic structures, which in turn can modify its magnetic behavior [3]. The effort to investigate such a correlation is helpful in principle to reach a better understanding of the itinerant magnetism or even to manipulate these materials for specific magnetic properties [4].

The compounds CuMPt_6 (M= Ti, V, Cr, Mn, Fe, Co and Ni) have been studied by Das et al. using X-ray and Electron diffraction methods [5]. They have explained that the specimens consisting of M= Cr, Mn, Co and Ni, quenched from 1000°C, have got the face-centered cubic (fcc) structure while in alloys with M= Ti, V and Fe ordering occurred with the structure of Cu₃Au type. On annealing at lower temperatures ordering has been induced in the compounds with M= Cr, Mn and Co and the structure is of Cu₃Au type. Among others only the CuMnPt₆ compound showed two-step ordering as ABC₆ type B Cu₃Au type B FCC with increasing temperature. Takahashi et al. have also studied the structural behavior of CuMnPt₆ single crystal using in-situ neutron diffraction technique earlier [6]. They described a secondary ordering phenomenon (2-step ordering) in this compound.

There is no report on the magnetic behaviors of $CuMPt_6$ (M= Ti, V, Cr, Mn, Fe, Co and Ni) ternary compounds in the existing literature [7]. It is, therefore, our interest to study the magnetic properties of $CuMPt_6$ compounds. In this paper, we report the results of in-situ magnetization measurement obtained from Superconducting Quantum Interference Device (SQUID). The dc magnetic susceptibility (C), Curie temperature (T_c) and the effective magnetic moment (P_{eff}) have also been determined.

2. EXPERIMENTAL

Polycrystalline specimens having the stoichiometric composition CuMPt_6 (M= Ti, V, Cr, Mn, Fe, Co and Ni) were prepared using conventional arc melting furnace in argon gas atmosphere. The starting materials were 99.9 to 99.99% pure. In order to obtain homogeneity in the specimens, each alloy ingot was re-melted at least 12 times by turning it over consecutively at the time of melting. Then the ingots were cut into pieces of rectangular

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shape having the dimension » 4'2'1 mm³ and weight of about 0.5 gm suitable for the SQUID measurements. Heat treatments were performed by putting the specimens individually in the evacuated quartz tube and putting them into the electric furnace. To attain the ordered state in the specimen annealing was performed by keeping the sample at 900°C for 12 hours then at 700°C for 12 hours and at 500°C for two weeks and was then gradually cooled down to room temperature. Quenching was accomplished by keeping the sample at 900°C for 24 hours then dropping into iced water rapidly for obtaining the disordered state. The chemical compositions of all the specimens were verified by Electron Probe Micro Analysis (EPMA) method. The homogeneity variation of the alloys was found to be less than 1 at. %. All the experimental measurements have been performed in the Institute of Materials Science in the University of Tsukuba, Japan.

3. EXPERIMENTAL RESULTS AND DISCUSSIONS

The magnetization measurements were carried out using SQUID in the temperature range 5-300 K and an applied magnetic field of 100 Oe. Fig. 1 shows the temperature dependence of dc magnetic susceptibility (c) of the compounds containing M= Ti, V and Ni in CuMPt₆ annealed at 500 ^oC for two weeks. The nature of the susceptibility curves reveal that a paramagnetic behavior exists in these compounds. Fig. 2 shows the susceptibility curves taken from CuCrPt₆ compound annealed at 500°C for two weeks. A spin glass type magnetic behavior appears in this compound, as the cusp type anomalies are observed in the susceptibility curves for both the zero field-cooled and field-cooled data [8]. The temperature for freezing the magnetic moments has been estimated as T_g= 15 K. The paramagnetic Curie temperature, Q =53 K, has been determined form the slope of the high temperature region of - T plot as shown in the inset of Fig. 2. The positive value of Q indicates that ferromagnetic interaction is dominant in this compound [9]. Fig. 3 shows the susceptibility curves taken from the same specimen quenched from 1000°C. A very similar nature of magnetism as that of annealed one has been found with minor change in T_g and Q as shown in the inset of Fig. 3.



Fig. 1. Temperature dependence of dc magnetic susceptibility (c) for M= Ti, V, Ni in CuMPt₆ compounds annealed at 500°C for 2 weeks, showing the paramagnetic behavior.



Fig. 2

Fig. 2. Temperature dependence of dc magnetic susceptibility (c) of CuCrPt₆ annealed at 500°C for 2 weeks, showing the spin glass type behavior with $T_g = 15$ K. Inset: 1/c - T plot showing the positive value of paramagnetic Curie temperature Q=53 K.



.Fig. 3. Temperature dependence of dc magnetic susceptibility (c) of CuCrPt₆ quenched from 1000°C, showing the spin glass type behavior with $T_g = 13$ K. Inset: 1/c - T plot showing the positive value of paramagnetic Curie temperature Q=51 K.

The compound CuMnPt₆ has got two-step structural phase transitions, observed in the X-ray and neutron diffraction studies [5, 6]. At 750 0 C the compound transforms its structure from ABC₆ type to Cu₃Au type and then at 970 $^{\circ}$ C, from Cu₃Au type to fcc structure. SQUID measurements have also been performed to observe the

magnetic nature for either of the crystalline states. Fig. 4 shows the temperature dependence of dc magnetic susceptibility for the specimen annealed at 500°C for two weeks. A spin glass type magnetic behavior appears in this compound. The temperature for freezing the magnetic moments was found as T_g = 40 K. Also the same nature has appeared in the case of quenched (from 1000°C) specimen but the temperature for freezing the magnetic moments was found as T_g = 35 K for this state of the compound as shown in Fig. 5. The paramagnetic

Curie temperature Q has been determined form the $\frac{1}{c}$ - T plot for either crystalline states as shown in the

insets of the Figs. 4 and 5. For Cu₃Au type structural state Q = 137 K and for FCC state Q = 135 K, respectively. The positive value of Q indicates that the ferromagnetic interaction is dominant in CuMnPt₆ compound for both of Cu₃Au type and fcc type crystalline state.



Fig. 4. Temperature dependence of dc magnetic susceptibility (c) of CuMnPt₆ annealed at 500°C for 2 weeks, showing the spin glass type behavior with $T_g = 40$ K. Inset: 1/c - T plot showing the positive value of paramagnetic Curie temperature Q=137 K.

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Fig. 5. Temperature dependence of dc magnetic susceptibility (c) of CuMnPt₆ alloy quenched from 1000°C, showing the spin glass type behavior with $T_g = 35$ K. Inset: 1/c - T plot showing the positive value of paramagnetic Curie temperature Q=135 K.

The alloy with M= Fe annealed at 500°C for two weeks shows the ferromagnetic behavior along with the Curie temperatures T_C = 230 K as shown in Fig. 6. The alloy with M= Co also shows the ferromagnetic behavior having T_C = 190 K (Fig. 6). The effective numbers of Bohr magnetons (p_{eff}) of all the compounds have also been calculated from the susceptibility measurements data. According to the quantum theory of paramagnetism [10] the effective number of Bohr magnetons is given by the equation



Fig. 6. Temperature dependence of dc magnetic susceptibility (c) for M= Fe and Co in CuMPt₆ compounds annealed at 500°C for 2 weeks, showing the ferromagnetic behavior.

$$p_{eff} = \sqrt{\frac{3k_B C}{Nm_B^2}}$$
(1)

where the constant values k_B , N and m_B are the Boltzmann constant, Avogadro's number and Bohr magneton, respectively. The Curie constant, C, was calculated from the slope of the 1/c vs. T plot of the corresponding compounds. The calculated p_{eff} values of alloys with M= Mn, Fe and Co show a good consistency with the standard values [10], and justify that the 3d elements in the alloys contributed to the magnetic properties. A little discrepancy has been found in case of the alloy with M= Cr. This discrepancy is not very unexpected because Cr is a special type of antiferromagnet and its nature is explained not by conventional antiferromagnetism of sublattices but rather by its spin density wave, first proposed by Overhouser [11] and experimentally established by E. W. Lee and M.A. Asgar [12]. Since antiferromagnetic nature of magnetic atoms diluted by non magnetic atoms like Cu and Pt can give rise to frustration in respect of magnetic ordering of spins, it is likely that the spin glass nature of Cr-substituted alloys have developed in our samples.

For the two ferromagnetic alloys (M = Fe and Co) the M-H hysteresis data have been taken varying the magnetic field from 0 to $\pm 5' 10^4$ Gauss at a constant temperature 5 K. Fig. 7 and Fig. 8 show the M-H curves for CuFePt₆ and CuCoPt₆ alloys, respectively. The saturated magnetic moments are M_s=18 and 12 emu/gm for CuFePt₆ and CuCoPt₆ alloys, respectively, determined from the curves. The effective moment for the CuFePt₆ alloy is calculated as 4.55 m_b and for CuCoPt₆ alloy 2.78 m_b per formula unit for Cu₃Au type structural state. Both the alloys show the characteristics of the soft magnetic materials. The hysteresis loop obtained from CuCoPt₆ is comparatively narrower than that of CuFePt₆ alloy. Apparently it means that CuCoPt₆ is magnetically softer than CuFePt₆ alloy, although the hysteresis parameters are not solely the intrinsic properties but are dependent on grain size, domain state, stresses and temperature [13]. The results of the magnetic measurements along with the structural phases for CuMPt₆ compounds are summarized in Table 1.



Fig. 7. M vs H hysteresis curve for CuFePt₆ ferromagnetic alloy annealed at 500°C for 2 weeks. Data have been taken at a constant temperature 5 K varying the magnetic field from 0 to $\pm 5 \times 10^4$ Gauss.



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Fig. 8. M vs H hysteresis curve for CuCoPt₆ ferromagnetic compound annealed at 500°C for 2 weeks. Data have been taken at a constant temperature 5 K varying the magnetic field from 0 to $\pm 5 \times 10^4$ Gauss.

Table 1

Structural phases and the corresponding magnetic states along with the Curie temperature (T_c) , freezing temperature (T_s) and the effective number of Bhor magnetons (p_{eff}) of the CuMPt₆ compounds

M in CuMPt ₆	Structural phases ⁰	Magnetic state	$T_{C}(K)$	T _g (K)	$p_{eff}(m_{B})$
Ti	Cu ₃ Au type	Para			1.52
V	Cu ₃ Au type	Para			2.61
Cr	fcc	Spin-glass		13	2.65
	Cu ₃ Au type	Spin-glass		15	2.17
Mn	fcc	Spin-glass		35	5.28
	Cu ₃ Au type	Spin-glass		40	4.86
	ABC ₆ type	Spin-glass			
Fe	Cu ₃ Au type	Ferro	230		4.55
Co	fcc	Ferro	185		3.43
	Cu ₃ Au type	Ferro	190		2.78
Ni	fcc	Para			2.69

 \overline{O} Data have been taken from Ref. 5.

4. SUMMARY

A variety of magnetic behaviors have been found in the CuMPt_6 (M= Ti, V, Cr, Mn, Fe, Co and Ni) ternary alloy system. The fundamental nature of magnetism for all the specimens remains the same whether it is quenched from high temperature or annealed at low temperatures. Only the critical temperatures T_C and T_g are changed a little due to order-disorder structural fluctuations. This fact suggests that the magnetic property is less sensitive on the atomic arrangement in the present system. The probable reasons may be the major composition (75 at.%) of the specimens is Pt, a paramagnetic element of fcc structure, and due to the fact that structural modifications do not produce any significant effect on the spin-alignment of these Pt-based compounds. It has also been observed that the magnetic behaviors change from paramagnetic to spin-glass type to ferromagnetic and again to paramagnetic with increasing the atomic number of the M elements. This is undoubtedly due to the unpaired electrons in the alloys, but further study is needed to clarify these phenomena in detail.

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