Thermal Expansion in Ferromagnetic Fe-Ni INVAR Alloy

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Abstract : The INVAR effect in 3D-transition metal $Fe_{1-x}Ni_x$ alloys with x=0.35, 0.50,0.70 is studied. Experiments have been made on thermal and structural properties to study the physical origin of INVAR characteristics such as thermal expansion atomic ordering, phase transition and curie temperature (Tc) which are closely related to its ferromagnetism.

$Fe_{0.65}Ni_{0.35}$ shows an anomalously negative thermal expansion coefficient below the curie temperature 528K with Fcc structure. This behavior is not found in other combination. The specific heat results shows an anomalous temperature dependant where a strong decrease in curie temperature as well as the magnetic heat of transformation occur with decreasing   Ni% content.

Keywords: *Curie temperature INVAR, , Phase transition, Thermal expansion*

I. INTRODUCTION

Fe-Ni alloys with Ni concentration of around 35% have an extraordinarily low thermal expansion coefficient and their dimension is nearly "*invar*iable" near the room temperature. Therefore, they are called INVAR alloys. These alloys show various anomalies in magnetic properties such as the deviation of the magnetization from the Slater–Pauling curve by decreasing the number of outer shell electrons, and the strong dependence of the Curie temperature T_C (the transition temperature between ferromagnetic and paramagnetic phases) on the mean distance of the constituent atoms in alloys [1]. These anomalies can be interpreted as due to the instability of the 3d-ferromagnetism in fcc lattice [2] [3].Experimentally, this instability has been observed, for example, as a rapid decrease in Curie temperature under the high pressure [4] [5], and a large change in magnetic properties by changing the Ni concentration in alloys [4]. Nickel is a non-carbide-forming element, which is soluble in iron in all properties. Nickel helps to prevent excess grain growth at high temperatures and enables fine grain steels to be produced more easily. It tends to stabilize austenite and thus lowers the critical temperature. This makes the heat treatment a little less severe. Nickel could be present in the steel up to 50 percent. In the range of 2 to 5 percent, nickel contributes great strength and hardness with high elastic limit, good ductility, and good resistance and decrease machinability. In the range of 30 to 40 percent nickel lowers the coefficient of thermal expansion, and in the range of 50 percent and above it, increases magnetic permeability. Large amounts of Ni give resistance to oxidation at high temperature. $Fe_{0.65}Ni_{0.35}$ invar alloys are well known as the material which has a very small thermal expansion coefficient near room temperature(<2 x 10^{-6} k^{-1} compared to most metallic materials which have a thermal expansion coefficient of 10-20x $l0^{-6}$ k^{-1}) and is widely used in industrial application, such as telecommunications, aeronautical and aerospace engineering, cryogenic engineering (liquefied natural gas tankers) etc, require either high dimensional stability with variation in temperature, or expansion characteristics matched with those of other materials such as glass, ceramics, or composite.In addition to the thermal expansion anomaly, Fe-rich fcc Fe-Ni alloys show many other anomalous properties, such as large negative pressure effects on the magnetization and on the Curie temperature , a large forced volume magnetostriction (the volume expansion induced by an applied magnetic field), and an anomalous temperature dependence of the elastic constants. any experiments and theoretical studies of INVAR alloys have been under taken in recent years using ultrasonic[5], neutron scattering high-energy heavy ion irradiation   [6] and other techniques to elucidate [7]   the microscopic origin of the effect but as yet no clear understanding has been reached.   Nevertheless in all models of the INVAR effect it is recognized that the relationship between magnetic and atomic volume or interatomic distance play a critical role.[8] In this work, we investigate the effect of nominal composition of Ni% on thermal expansion coefficient, transition temperature and structural properties of $Fe_{1-x}Ni_x$ alloys at(x=0.35, 0.50, 0.70).

II. MATERIALS AND METHODS

Alloy has been obtained from mixing different percentage of pure Fe (99.98%) and Ni (99.995%) powders using   planetary ball mill for 24 h and under argon atmosphere   using stainless steel balls and containers. The intensity of the milling expressed by the ball velocity hitting is 5 m/s, which is strong enough to obtain perfect alloys [9][10].A cylindrical samples of 10mm diameter and 30mm height have been made for each ratios and   uniaxally pressed at 1000(Kegs). Sintering is carried out at temperature of 1350°C for 2hrs of 10°C/min heating rate.The metallic ratios of the alloys are determined by atomic absorption spectroscopy and energy dispersive spectrometer techniques. The result of these measurements indicates no evidence of any impurities. The samples for XRD, DTA were obtained by crushing the alloys in ball mills down to 300 mesh grains. The structure of the $(Fe_{1-x}Ni_x)$ alloys (0.35,0.50,0.70) were initially investigated using XRD at room temperature with a (Philips diffractometer type PW 1877). To obtain lattice parameter more accurately, the Nelson-Riley extrapolation was used. [11]The specific heat measurements at high temperatures were obtained from the differential thermal analysis (DTA type Netzsch 409) where a 1100°C furnace is   used with a heating rate of 10°C/min. The weights of precipitated samples used were about 500mg. The specific heat determined from the peak area in DTA, which is depends on the mass (m) and the heat reaction or the enthalpy change (ΔH); then using the equation:

$$C_p = \left[\left(\Delta H \big/ dT\right)\right]\left(1\big/_m\right)$$

Where, dT is the difference in temperature between a sample (S) and an inter reference (R). A cylindrical specimens ~3cm in length is used for thermal expansion measurements which is connected to DTA equipment to measure the coefficient of linear expansion as a function of temperature in range 300 to 1300K.

III. RESULTS AND DISCUSSION

Alloys which exhibit INVAR behavior have a small thermal expansion coefficient below the Curie temperature.

'Figure 1', shows the thermal expansion coefficient versus temperature curves.
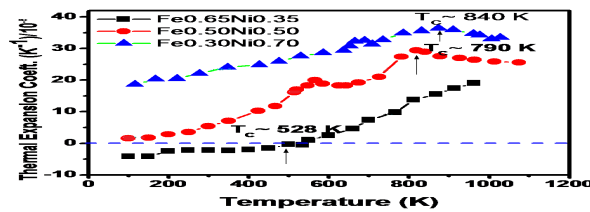

Figure 1: The relation between Coefficient of thermal expansion and temperature
(K) for different x values of $Fe_{1-x}Ni_x$ alloys

For alloys in the $(Fe_{1-x}Ni_x)$ system (x=0.35, 0.50, 0.70), the composition $(Fe_{0.65}Ni_{0.35})$ shows negative thermal expansion below Curie temperature (T_c) with more INVAR effect than in $(Fe_{0.50}Ni_{0.50})$. Both alloys have an (fcc, gamma phase) structure. The system $(Fe_{0.35}Ni_{0.70})$ has (bcc/ alpha phase) structure where there is no INVAR behavior.'Figure 2',shows XRD simulation for $(Fe_{0.65}Ni_{0.35})$and$(Fe_{0.50}Ni_{0.50})$alloys in which there is a significant shift in the peak diffraction due to changing in lattice parameter.
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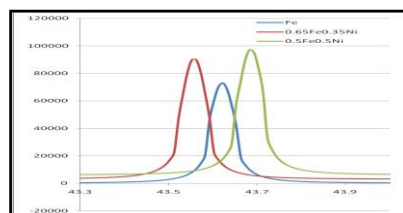Table 1' shows the results relevant properties of the three samples [12]

Figure 2: Simulation XRD for $Fe_{0.65}Ni_{0.35}$ and $Fe_{0.50}Ni_{0.50}$ alloys

Table (1) : Physical properties of the test Fe-Ni alloys at 296K[12]

| Properties	$Fe_{0.65}Ni_{0.35}$	$Fe_{0.50}Ni_{0.50}$	$Fe_{0.30}Ni_{0.70}$
Average atomic mass (amu)	56.90	57.28	56.31
Structure	FCC(alpha)	FCC(gamma)	BCC(alpha)
Lattice constant (A)	3.596	3.586	3.586
Melting temp(K)	1740	1710	1750
Curie temp(K)	528	790	840
Saturation magnetic moment (μ_B/atom)	1.85	1.68	1.25

The reason of negative thermal expansion of the system ($Fe_{0.65}N_{0.35}$) below Tc is due to the interaction between the lattice vibration and magnetic degrees of freedom. Increasing the temperature tends to produce expansion, but because of special strong magnetic attraction between atoms in this alloys, a small net expansion takes place. However, above Curie temperature the alloy expands normally. These results are identical with the spontaneous magnetization of the alloys which shows anomalies such as deviation of the magnetic moment curve Fe concentration [13]..The specific heat results for ($Fe_{1-x}Ni_x$) alloys are shown in Figure 3(a , b, c) which reveal an anomalous temperature dependence
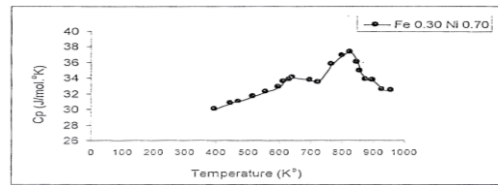
(a)
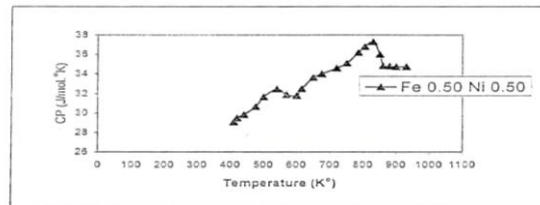
(b)
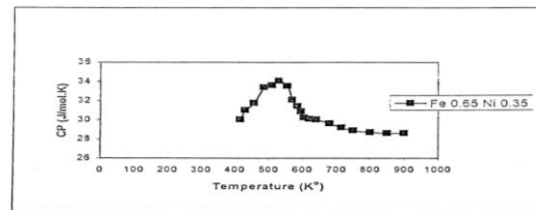
(c)

Figure 3(a ,b,c): The relation between specific heat (C_p) and temperature (K) for $Fe_{1-x}Ni_x$ alloys (x=o.70, 0.50, 0.35) respectively.
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At ($Fe_{0.50}Ni_{0.50}$), C_p-curve is characterized by two distinct maxima   one at about 790°K and the second reveals an anomalous maximum at about 540°K. The position of this anomalous maxima is only slightly concentration depended where their is a strong decrease of the curie temperature as well as of the magnetic heat of transformation occurs with decreasing nickel content so that the alloy with 0.35% Ni reveals only a single maximum composed of both curie peak and the maximum caused by the anomalous excess heat .Similar effects have been seen   in other INVAR alloys such as ($Fe_{0.50}(Ni_x$ $Mn_{1-x})_{0.50}$ of $Fe_{0.80-x}Ni_xCr_{20}$ system [14][15]   . To obtain a deeper understanding of the INVAR phenomena, it is helpful to collect microscopic information about the physical properties. More detailed discussion of these   phenomena will be given in future work.

IV. CONCLUTIONS

In This work, we investigate the effect of nominal composition on thermal and structural properties of INVAR $Fe_{1-x}Ni_x$ alloys with(x=0.35, 0.50,0.70.)It was found that:

1- $Fe_{0.65}Ni_{0.35}$ shows Negative thermal expansion below Curie Temperature T_c due to interaction between lattice vibration and magnetic degree of freedom

2- The specific heat result revel an anomalous temperature dependence with x-content with strong decrease of T_c and magnetic heat of transformation with decreasing Ni %

3 -$Fe_{0.65}Ni_{0.35}$ shows more INVAR effect than in $Fe_{0.50}Ni_{0.50}$. Both alloys have an fcc ,γ - phase structure while $Fe_{0.35}Ni_{0.70}$ has bcc α- phase structure where there is no INVAR behavior.

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