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Synthesis and magnetic properties of Laves phase Fe_2Nb amorphous alloy

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Abstract

The formation process and magnetic properties of the Laves phase compound of nominal composition Fe_2Nb in its amorphous phase prepared by mechanical alloying have been investigated. The effect of milling time on the formation of amorphous phase has been studied using X-ray diffraction technique. Further characterizations were carried out by particle size measurement, dc magnetization, ac susceptibility and ferromagnetic resonance (FMR) studies. Magnetisation and susceptibility studies show soft ferromagnetic behaviour whereas ferromagnetic resonance studies show some sort of disorder/strain introduced during the mechanical alloying process.

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Keywords: A. Disordered system; Mechanical alloying; X-ray diffraction

1. Introduction

In recent years a great deal of interest has been generated in the synthesis of advanced engineering materials in their stable, meta-stable, crystalline, quasi-crystalline and amorphous phases, making their scope much wider. These advanced materials include metals, ceramics, intermetallic compounds, alloys and several other composites. Amorphous alloys play an important role both in basic research and application. It is interesting to note that these alloys are also used in most theft protection systems where the interplay between the magnetic and mechanical properties are employed. There are various methods for preparing amorphous alloys. Solid state amorphisation is one of the most important and has become an alternative route to rapid solidification process [1,2]. Mechanical alloying (MA) is a solid state amorphisation technique which has attracted world wide attention due to its simplicity, cost effectiveness and room temperature synthesis process [3,4]. It is a powder process usually carried out in an inert atmosphere in a high energy ball mill. The milling process eventually leads to an ultrafine composite, in which amorphisation by solid state reaction takes place to prepare unique materials in terms of composition and microstructure. In the past a large number of Al and transition metal (TM) based binary, ternary and quaternary alloys have been synthesized [5–12]. It is interesting to note that the Laves phase MFe_2 type intermetallic compounds with M = Y, Zr and Lanthanides crystallize in C 15 type cubic structure and show ferromagnetic behaviour whereas the same type of compounds with M = Sc, Ti, Nb, Hf, Ta and W crystallize in C 14 type hexagonal structure and show variable magnetic properties [13–15]. Experimental investigation shows that C 14 type hexagonal Fe_2Nb is a paramagnet [16] and later studies support the same compound to be a strongly enhanced pauliparamagnet [17]. Nb NMR studies show that Fe_2Nb is a near ferromagnet and subsequent studies by the same author revealed that the compound is a weak antiferromagnet with Neel temperature around 10 K which is suppressed by an external magnetic field of higher than 6 kOe [18,19]. From all these studies, it is observed that the magnetic properties of C 14 type hexagonal Fe_2Nb compound is fluctuating in nature. Not only this, almost all experimental and theoretical investigations were carried out only on crystalline compounds and practically no work has been carried out on the noncrystalline (amorphous) phase. In view of this the present investigations were carried out on the synthesis process and magnetic behaviour of the Laves phase Fe_2Nb amorphous alloy prepared by mechanical alloying.

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2. Experimental

The nominal composition Fe_2Nb was synthesized by mechanically alloying the mixture of high purity (>99.9%) metallic powder of iron and niobium (obtained from Johnson Mathew and Co., England) in a high energy Fritsch planetary ball mill with a ball to powder weight ratio of 20:1 and speed ranging from 450 to 650 rpm. In order to avoid oxidation during the alloying process, the vial was sealed with high purity argon gas. The ball diameter used in the experiment was 12 mm. The vial was opened after every 5–10 h and a small amount of powder was used for X-ray diffraction to investigate the formation of amorphous phase. Further characterizations were carried out by particle size, dc magnetization, ac susceptibility and ferromagnetic resonance (FMR) studies. The room temperature X-ray diffractograms were recorded on an Isodebyeflex X-ray diffractometer using CuK_α radiation and Ni filter in a wide scanning range of 2θ from 30° to 90° with a scanning rate of 2°/min. The particle size was measured with the help of Coulter counter model Z_B and B using NaCl as electrolyte. Each particle passing through the small aperture between the electrode displaces its own volume of electrolyte which is measured in terms of voltage pulse. The height of each pulse is proportional to the volume of the particle. The dc magnetization measurement was carried out on 150-A PAR vibrating sample magnetometer (VSM) as a function of field as well as a function of temperature from room temperature (RT) to 830 K/845 K at magnetic fields of 60 Oe and 8 kOe. The ac susceptibility measurement was carried out using an ac mutual inductance technique in the temperature range from 25 to 135 K. The ferromagnetic resonance (FMR) spectra were recorded at RT and at liquid nitrogen temperature (LNT) using Varian Associate 109 X band EPR spectrometer with 100 kHz field modulation. The g value was measured using the standard DPPH sample ($g = 2.0036$).

3. Results and discussion

The X-ray diffractograms recorded at different stages of the mechanically alloyed powder of nominal composition Fe_2Nb are shown in Fig. 1. In the early stages of the mechanical alloying process there is evidence of combination of pure metal and the desired Fe_2Nb phase. A completely amorphous phase was obtained after 17 h of mechanical alloying. Further alloying for 5 h has no effect on the structural behaviour. The formation of the amorphous phase depends upon the milling intensity i.e. change in energy during the alloying process. Apart from the milling intensity and kinetics of free enthalpy changes during the alloying process, the concentration of Fe presumably plays an important role in the formation of the amorphous phase. When Fe concentration changes slightly from Fe_{66} (in Fe_2Nb) to Fe_{60} in the case of the $Fe_{60}Nb_{40}$ alloy reported earlier [11]; the amorphisation could take place after 40 h of the alloying process. In the early stages of the alloying process, the powder particles become coated on the milling tool, showing the interdiffusion and reaction of the powder particles as observed earlier [9]. Further alloying removes the coated

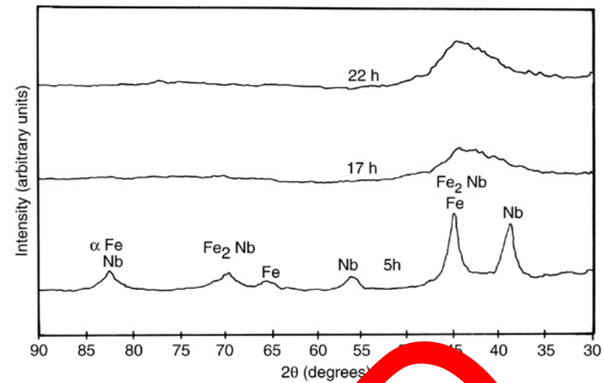

Fig. 1. X-ray diffraction patterns of mechanically alloyed Fe_2Nb after 5, 17 and 22 h.

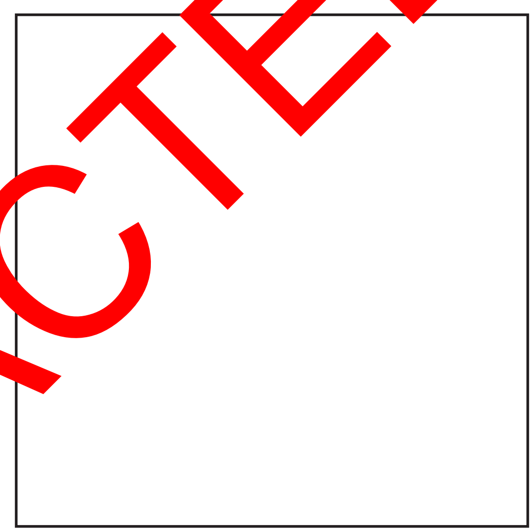

Fig. 2.

powder from the ball and gives a composite mass of uniform microstructure and random orientation of submicron size. The particle size as measured from the Coulter counter was of the order of 10–100 nm which is consistent with our earlier report [11,12].

Fig. 2 shows the magnetization versus temperature curve of amorphous Fe_2Nb alloy at a magnetic field of 60 Oe. The inset shows the corresponding M vs H plot. The M vs H plot, saturates at a field of 11 kOe but the hysteresis is not measurable. The saturation magnetization value of 13.4 emu/g at a field of 11 kOe and a remanence of 1.5 emu/g are noteworthy for a typical amorphous ferromagnet. The M vs T plot does not show any evidence of high temperature transition in the whole range of measurement. The estimation of Curie temperature from the M vs T plot is surprisingly high, not reported for any of the Fe based alloys. Further, the M vs T plot shows a tendency which is just on the border line of ferromagnetism upto 580 K. On further increasing the temperature, the moment formation increases rapidly upto 700 K, after that the enhancement of moment formation upto 830 K is not so fast, showing some sort of relaxation which is also confirmed by the step like behaviour of the M vs time

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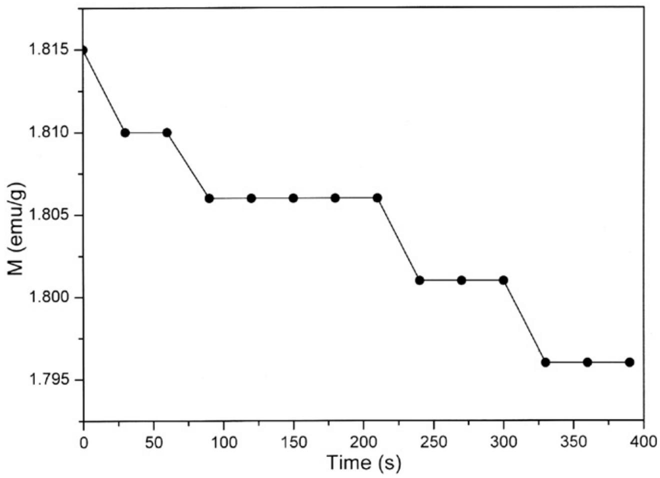

Fig. 3. M vs time plot of field cooled Fe_2Nb amorphous alloy.

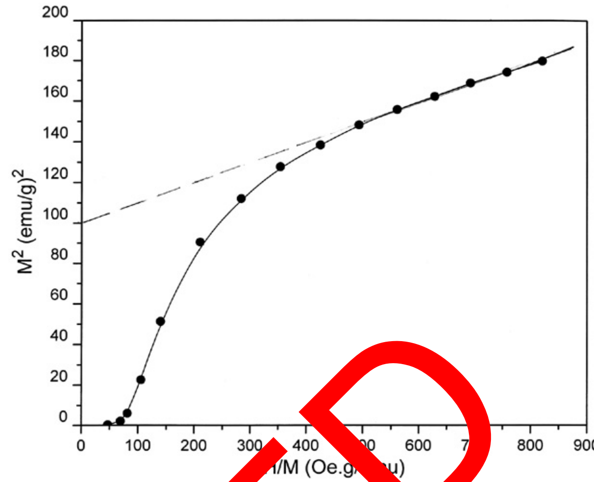

Fig. 5. Arrott plot (M^2 vs H/M) of Fe_2Nb amorphous alloy.

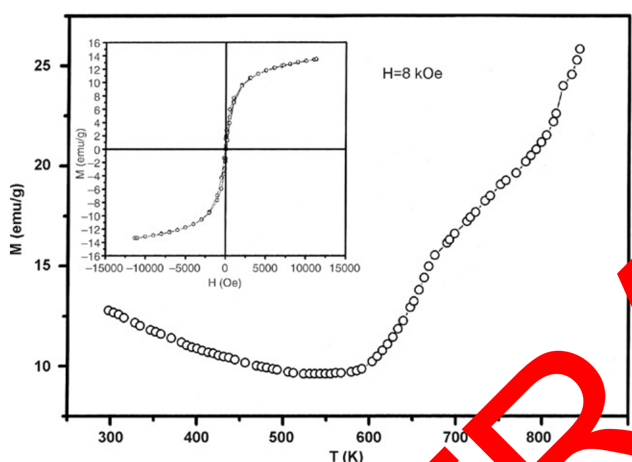

Fig. 4. M vs T plot of Fe_2Nb amorphous alloy.

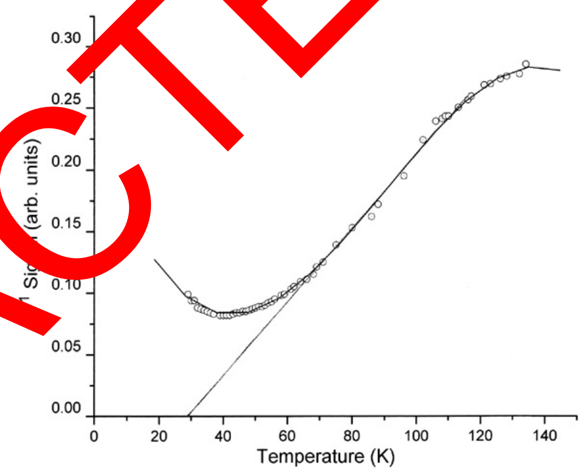

Fig. 6. χ^{-1} vs T plot of Fe_2Nb amorphous alloy.

plot shown in Fig. 3. The increase in the magnetic moment beyond 580 K may be due to crystallization process in the high temperature region. Again the measurement was carried out during the cooling cycle from 830 K to RT and a thermal hysteresis was observed with a maximum magnetization of 2.4 emu/g at RT. This increase in the magnetization value in the cooling cycle may be due to the crystallization of the amorphous phase and the subsequent enhancement of the ferromagnetic clusters and the exhibition of a weak ferromagnetic behaviour. The step like behaviour of the M vs Time curve also supports the relaxation behaviour of the ferromagnetic clusters during the measurement process. To obtain further insight into the magnetic behaviour, the M vs T measurement was repeated at a higher field of 8 kOe from RT to 850 K and is shown in Fig. 4. The inset shows the corresponding M vs H loop which is not measurable. The M vs T curve shows similar behaviour as observed at low field (60 Oe) with the only difference being that the magnitude of magnetic moment has been enhanced to around an order of magnitude. This also supports the weak ferromagnetic behaviour of the material. For further examination of the ferromagnetic behaviour, Arrott plot of magnetization (M^2 vs H/M plot) at RT is shown in Fig. 5. It is clear from the figure that the interception of the extrapolated value of M^2 to $H/M = 0$ with the vertical axis is positive which indicates that spontaneous magnetisation is present in Fe_2Nb and hence shows a weak ferromagnetic behaviour.

To obtain further insight into the magnetic behaviour at low temperature, ac susceptibility measurement has been carried out. It is interesting to note that susceptibility increases, goes to a maximum and then decreases again with the increase in temperature. Based on this observation it is likely that the maximum in the χ vs T curve in a low field is ascribed to neither antiferromagnetic ordering nor spin glass freezing. It is worth noting that this type of magnetic behaviour shows close resemblance to those observed in exchange enhanced metals, such as $TiBe_2$ [20]. As mentioned above the magnetic state of Fe_2Nb is just on the verge of onset of ferromagnetism. Hence the maximum in the χ vs T curve has the same origin as those observed in $TiBe_2$ which has been extensively discussed but not yet fully understood. To analyse the result further the temperature dependence of χ^{-1} vs T has been plotted and is shown in Fig. 6. From the figure it is clear that Curie–Weiss law does not hold. The deviation from the Curie–Weiss law may be due to the intrinsic property of the system. The extrapolated

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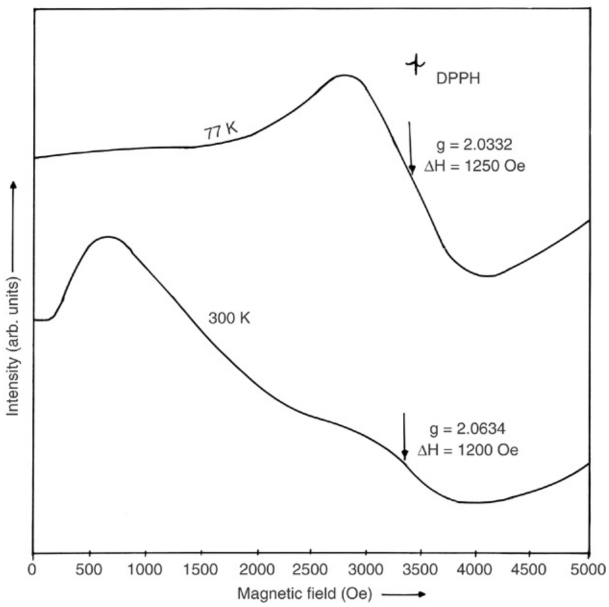

Fig. 7. FMR spectra of Fe_2Nb amorphous alloy.

line of the χ^{-1} vs T plot intercepts the temperature axis on the positive side, indicating the presence of spontaneous magnetization and hence ferromagnetic behaviour. The result obtained from the magnetic susceptibility also supports the result obtained from the Arrott plot shown in Fig. 5.

For further studies on magnetic behaviour as well as to get an idea about the role of magnetic interaction, the author has recorded the FMR spectra of the same amorphous alloy at 300 K and at 77 K and the results are shown in Fig. 7. At 77 K the resonance is broad and is on the higher side of the field whereas at 300 K the resonance is too broad and is on the same higher side of the field. At 77 K the line width and g values are 1250 Oe and 2.0332 whereas the same values at 300 K are 1200 Oe and 2.0634. The broadness of the resonance lines are consistent with the introduction of disorder and strain introduced during the mechanical alloying process as observed earlier by the author [21].

4. Conclusion

From all these studies it is concluded that the Laves phase Fe_2Nb alloy is magnetically soft indicating that its magnetic moment is weakly constrained by the local environment. Magnetisation and susceptibility measurements show weak ferromagnetic behaviour with some sort of relaxation whereas the ferromagnetic resonance spectra show some sort of disorder and strain which are introduced during the mechanical alloying process.

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