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Review Paper

A Comprehensive review on Fe, Co and Ni based Ferromagnetic Alloys

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Abstract

Ferromagnetic alloys have been exhibited to be promising candidate due to their high magnetic moment value at room temperature and feasible for device application. Among all the ferromagnetic materials, Fe, Co and Ni have exhibited the strong ferromagnetic character due to their d-electron's contribution. However, these elements have shown the tremendous enhancement in their magnetic moment when substituted in several alloys like Heusler Alloys. Here, we report a review on several Fe, Co and Ni based Full-Heusler alloys which have revealed the strong ferromagnetic behavior even above the room temperature. These alloys have been considered to be a suitable material for spintronic devices in future.

Keywords: Ferromagnetic Materials, Magnetic Moments, Heusler Alloys, Spintronic Devices, Device Application etc.

Introduction

In recent decades, magnetic materials have been revealed as a future material for spintronic devices. Among other, several materials such as Heusler alloy have been exhibited as a significant materials due to their high magnetization value feasible even above the room temperature. As we know, Ni, Fe and Co elements have revealed the ferromagnetic properties in their ground state while Cr, Mn are identified as antiferromagnetic materials. Although, several elements from lanthanide (f-block) elements have also shown the magnetic properties. Basically, Heusler alloys have been made of these combining these ferromagnetic and antiferromagnetic (d-block) elements including few p-block elements. In 1903, Friedrich Heusler had firstly prepared the material with the combination of Cu, Mn and Al in the ratio of 2:1:1 respectively. Generally, Cu, Mn and Al are non-magnetic in nature. After preparing the materials, Cu_2MnAl was formed in face centered cubic (fcc) structure[1].

From his name, materials are now known as full-Heusler alloy. Cu_2MnAl full-Heusler alloy have exhibited the very high magnetic moment around $5\mu_B$. After this discovery, large number of Heusler alloys have been explored till now. Basically, Heusler alloys are categorized in four types depending upon the site occupancies and number of elements used to fabricate them i.e. half-Heusler, full-Heusler and Quaternary Heusler alloys. Half-Heusler alloys contain three elements (ABC) in which A and B are transition metals and C lies in p-group. Site occupancies of A, B and C respectively while on vacant site exists in the fcc unit cell.

Vacant site in half-Heusler could be filled by A to form A_2BC which is known as full-Heusler alloy. One can fill different transition metal elements (say A´) also in vacant site of half-Heusler to form AA´ BC which is known as Quaternary Heusler alloy. These alloys makes four interpenetrated fcc lattices separated by one fourth of their body diagonal along the body diagonal direction. Effective number of each element are four occupied by octahedral and tetrahedral site.

In order to increase and achieve the theoretical value of magnetic moments, large number of Heusler alloys based on Co, Mn, Fe, Ni have been widely explored. Here, we have focused on the famous full-Heusler alloys such as Co_2MnSi, Co_2FeSi, Ni_2MnSb, Ni_2MnSb, Ni_2MnGe, Mn_2NiSb, Mn_2NiSn, Mn_2NiGe, Mn_2NiIn, Fe_2VAl etc. Manuscript is divided in four sections depending upon the nature of materials along with their compositional elements i.e. Mn_2, Ni_2, Co_2 and Fe_2 based full-Heusler materials.

First half-Heusler half-metallic NiMnSb alloy was studied by de Groot el. though theoretical calculation[2]. Excess Ni substitution had implemented at vacant site in NiMnSb and prepared full-Heusler Ni_2MnSb alloy by several groups[3,4].

Interestingly, both NiMnSb and Ni_2MnSb have exhibited the ferromagnetic ground state having magnetic moment $4\mu_B$ excepted half-metallic nature of NiMnSb as shown in Figure-1. Further, several other materials such as Ni_2MnGe have prepared and magnetic properties have been measured.

Synthesis and Characterizations of Heusler Alloys

All the elements in their powder form are mixed about an hour through mortar and pestle. Mixed powder is then placed in an ampoule made by quartz and maintained the high vacuum in the quartz tube in order to avoid the oxidation of elements through the environment. These evacuated tubes are then subjected in the muffle furnace for further reaction of the samples for 9-10 days at around 1000℃ temperature. After completing the reaction, quartz been taken out from the furnace and dip in the cold water which is known as quenching. All the samples are again crushed using mortar and pestle about an hour. These samples are ready for the XRD measurement, Scanning Electron Microscopy (SEM), Tunneling Electron Microscopy (TEM) and magnetic measurements through Vibrating Sample Magnetometer (VSM) also. After finding the XRD data, Rietveld refinement is performed for matching the experimental XRD data with standard one. Surface morphology and internal crystal structures are obtained from SEM and TEM. Magnetization with magnetic field and with temperature are measured form VSM. Schematic chart is given in the below for the preparation of Heusler alloys through solid state reaction route. This is lengthy but easy route for alloy preparation. Although some other methods have also been used for the preparation such as arc melting, melt spinning, ball milling etc. Schematic chart of the procedure of fabrication and characterization of the magnetic sample have been represented in the Figure-2.

Figure-1: Schematic unit cell of NiMnSb half-Heusler (left one) and Ni_2MnSb full-Heusler (right one).

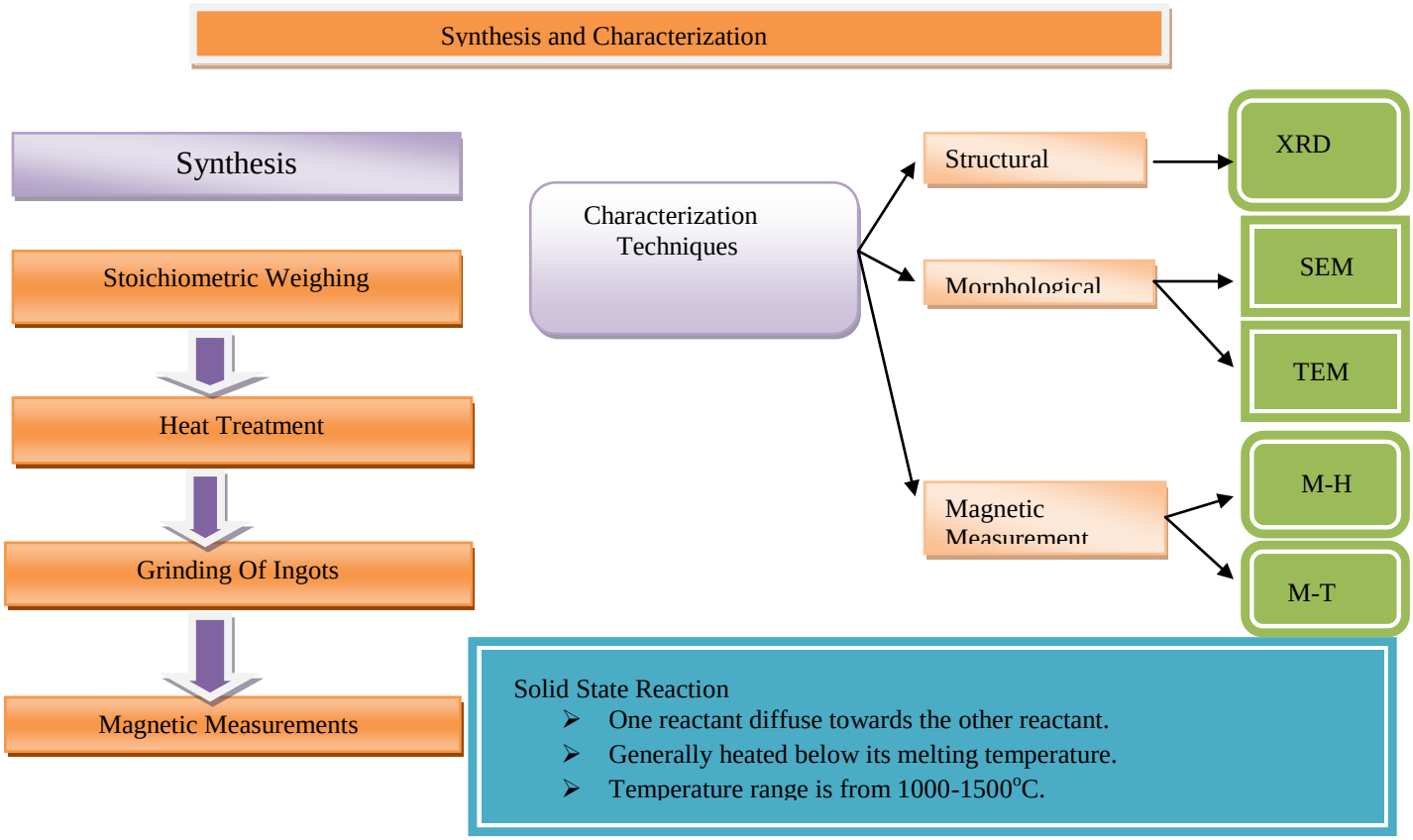

Figure-2: Schematic chart of experimental procedure for the preparation of Heusler alloys.

Ni and Mn_2-based full-Heusler alloy

Mn_2NiSb have been basically found to be stabilized in two different prototype structure i.e. Cu_2MnAl and Hg_2CuTi prototype structure. Particular structure depends upon the type of preparation. Schematic diagram of unit cell of both prototypes have shown in Figure-3. Mn_2NiSb have also found in ferromagnetic state in both the structure[5,6]. Although, Mn has antiferromagnetic character in their fcc structure. In case of Heusler, Mn shows the ferromagnetic nature due to the distance between the Mn atoms. Spin of Mn atom with its nearest neighbor align himself parallel to each other if the distance between them maintained larger than 2.9Å.

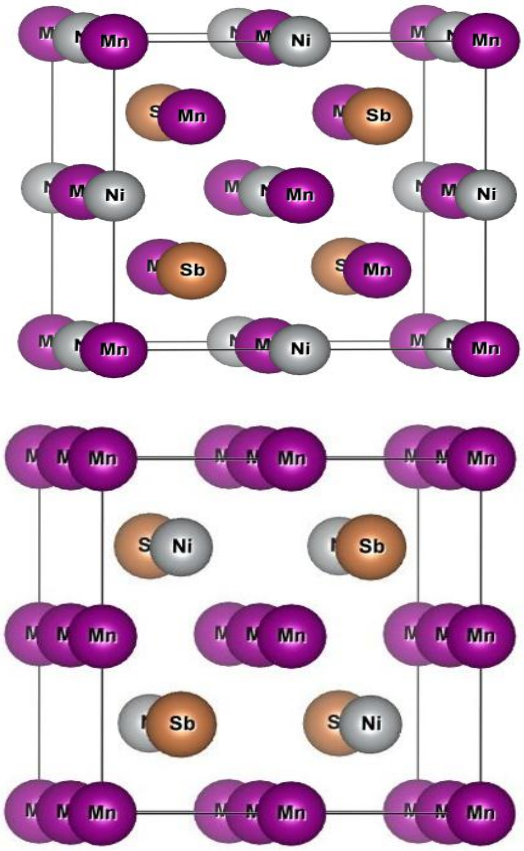

Figure-3: Hg_2CuTi Prototype (Mn_2NiSb) Cu_2MnAl Prototype (Mn_2NiSb).

Moreover, Scaling of magnetic moments along with the Curie temperature is given by Slater Pauling rule which is as follows-
$M_t = N_v$- 18, for half Heusler
$M_t = N_v$- 24, for full-Heusler
$M_t = N_v$- 28 for full-Heusler

Where M_t is total magnetization and N_t is the total valence electron count of the unit cell. From this rule, we have calculated magnetic moments of several Mn_2-based alloys.According to this rule, we have calculated the magnetic moment of Mn_2NiSb. Valence electron count (VEC) of Mn, Ni and Sb are 7, 10 and 5 respectively so the total magnetic moment is given by-
Mn_2NiSb- $N_V = 7X2+10+5 = 29$
$Mt = N_v$- 24 = 29-25 = $5\mu_B$.

We have already mentioned that XRD data are sensitive to the elements having large difference in size and electronegativity. However, superlattice reflection peaks are very sensitive to the swapping of atoms. Hence, we have calculated the ratio of super-reflection peaks (111/200) and found the difference of 23% in their ratio as shown in inset of Figure-4. This difference suggests that rigorous analysis of XRD data especially super reflection peaks is very important factor for determining the type of structures in which samples are stabilized.

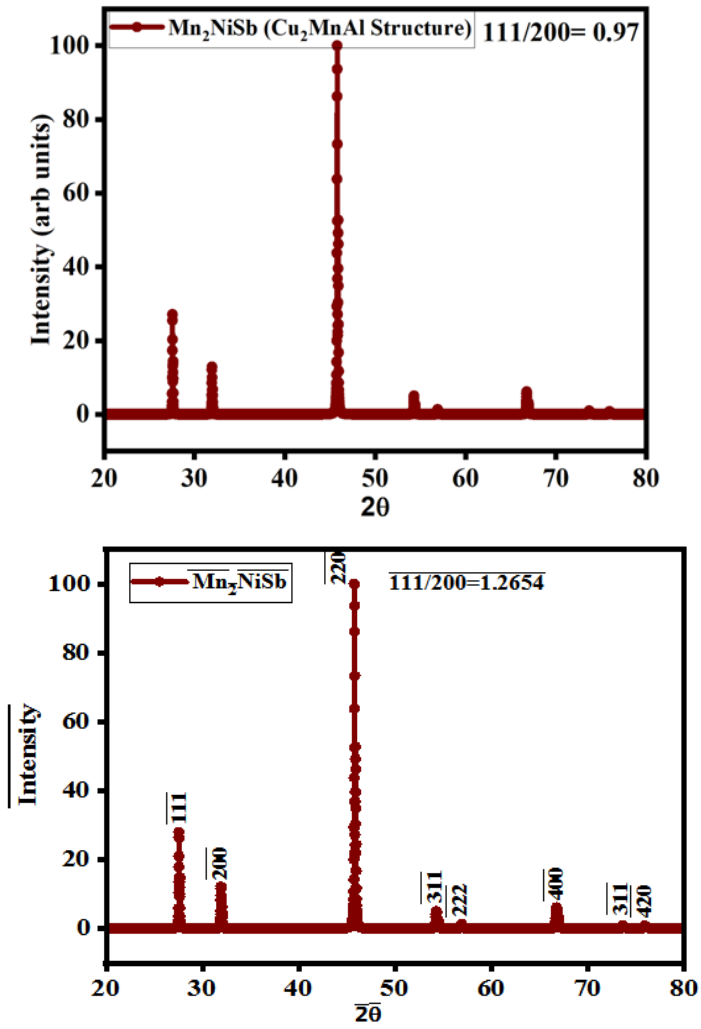

Figure-4: Simulated XRD pattern of Mn2NiSb in Hg_2CuTi and Cu_2MnAl prototype structure.

Moreover, we have listed the elements which might be useful for the fabrication of half and full-Heusler alloys as at X, Y and Z position in FCC lattices as shown in Table-1.

Table-1: Selection of elements X, Y and Z for X_2YZ full-Heusler alloys.

Elements of Half and Full-Heusler Alloys															
X	Mn	Fe	Co	Ni	Cu	Ru	Rh	Pd	Ag	Cd	Ir	Pt	Au	Li	Mg
Y	Li	Be	Sc	Ti	V	Cr	Fe	Co	Ni	Zn	Y	Zr	Nb	Mo	Hf
Y	W	La	Ce	Pr	Nd	Sm	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	….
Z	Mg	Zn	B	Al	Si	Ga	Ge	As	In	Sn	Sb	Pb	Bi	…	….

Now, we have plotted the magnetization value for several Mn_2NiX (Al, Si, P, Ga, Ge, AS, In, Sn, Sb, Ti, Pb Bi) based alloys based on the Slater Pauling rule which is shown in Figure-5.

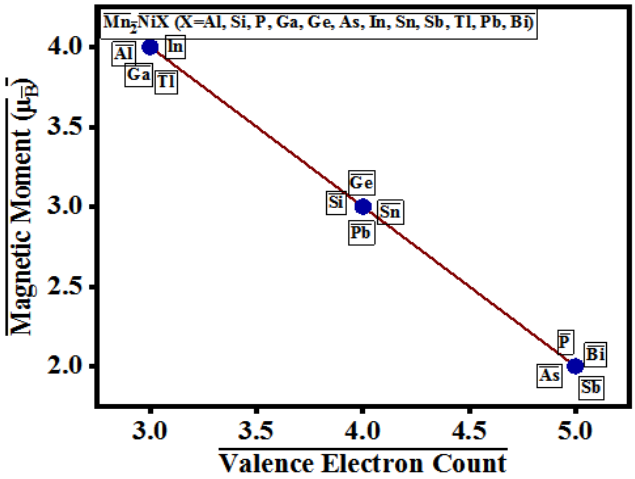

Figure-5: Valence electron count versus magnetization calculated using Slater Pauling rule.

In several reports, it has been shown that magnetic moment resides mainly on Mn site rather than Ni and Sb. Now, several other studies have been performed on Mn_2 based Heusler alloys such as Mn_2NiGe, Mn_2NiSn etc. Mn_2NiSn has been found to be stabilized in Hg_2CuTi prototype structure with ferromagnetic state having magnetic moment of $4\mu_B$. However, Mn_2NiGe has exhibited the ferrimagnetic ground state. Mn_2NiGe has also revealed the half-metallic nature reported by N. Asli et. al. and they have found the more stability of Mn_2NiGe in Hg_2CuTi type of structure while Ni_2MnGe is half-metallic in both Hg_2CuTi and Cu_2MnAl prototype[7].

On the other hand, Mn_2FeAl, Mn_2NiAl, Mn_2CoAl have also been theoretically studied in β −Mn type crystal structure and found the antiferromagnetic ordering of Mn atoms[8]. Further, Mn_2NiSi and Ni_2MnSi have also been studied and found in the austenite and martensite phase with energy difference of 0.3meV/cell[9]. Among the others, Mn_2PtSn have revealed as multifunctional materials such as giant topological Hall effect in their tetragonal structure, antiferromagnetic characters, compensated antiferromagnetic nature etc[10-14].

Co_2-based full-Heusler alloys

Co_2 based full-Heusler alloys have been most studied materials among the family of Heusler alloys due to their half-metallic, high magnetic moments and high Curie temperature. Co_2MnSi and Co_2FeSi is the most famous full-Heusler alloy in this group[15-18]. In a study, it has been shown that Co_2FeSi is more suitable for spintronic devices in comparison to Co_2MnSi when formed at low temperature using molecular beam epitaxy grown on MgO (001), $SiTrO_3$(001) and $MgAl_2O_4$ (001)[19]. Pressure and disorder effect have also been studied and found that minority band gap has increased with volume compression and half-metallicity has been found to be preserved under pressure. Due to the low difference of size and electronegativity between the Co and Fe, swapping between them is established which leads to reduce the half-metallic nature and destroy the spin polarization. However, swapping between the Fe and Si do not have any role to reduce the spin polarization. Magnetism has shown to be decreased with Fe-Co disorder but there is no any effect on magnetism due to the Fe-Si type disorder[20]. Epitaxial layer of Co_2FeSi has also been grown in B_2 and $L2_1$ ordering and found anisotropic magnetoresistance which is attributed to the transition from non half metallic to half metallic and small value of Gilbert constant[21, 22].

Fe_2-based full-Heusler alloys

Fe_2VAl is the most famous materials in iron based full-Heusler alloys. It has generally shown the semiconducting like behavior in their ground state[23]. Thermoelectric properties are found to be very staring in Fe_2VAl full-Heusler alloy[24]. Transition from ordered to disordered has also been theoretically investigated through the Rietveld refinement of the Neutron diffraction data taken during the heating of the sample[25]. Nuclear magnetic measurement has also performed for Fe_2VAl full-Heusler alloy which also showed the semiconducting behavior[26].

Conclusion

We have briefly reviewed the Co, Ni, Fe and Mn based full-Heusler alloys. We have also simulated the XRD pattern using Vesta software for two prototype structure and calculated the ratio of super reflection peaks which revealed the difference between these structures.

We have also discussed the preparation technique using solid state reaction route. We have also given the magnetization curve with VEC using Slater Pauling rule. We have also included the study Co and Fe based full-Heusler alloy in brief.

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