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Synthetize and Magnetic Properties of Al Substituted Ni Spinel Ferrites Prepared by Conventional Method

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Abstract
The mixed polycrystalline Ni$Al_xFe_{2-x}O_4$ ferrites, where *x* is the percentage increments of *Al* content, were prepared using the standard double sintering by mixing pure metal oxides Al_2O_3, NiO and Fe_2O_3. The magnetization (*M*) was studied at room temperature as a function of applied magnetic field (*H*) over the range of (0-40) Oe in a constant magnetizing frequency (*f = 50 Hz*) and applied AC current (I_{AC}) over the range of (*0 – 5*) *A*. *M* showed increasing with increasing of *H*. *M* found to decrease while the concentration of *Al* increases in matrix. The decreasing of the magnetization with increasing of Al^{3+} ions is explained by Neel's two-sublattice model. The relation between *H* and relative permeability (μ_r) of *Al – Ni* ferrite samples shows that μ_r decrease for all samples as *Al* content increase. Substitution of the non-magnetic Al^{3+} ions in *Ni* spinel ferrite has a tremendous influence such the magnetic properties. So, the mixed *Al – Ni* spinel ferrite is considered a soft ferrite material, which is proved to be an interesting material for scientific and technological applications.

Keywords:
Magnetic Properties,
Spinel ferrite,
Relative permeability,
Neel's model.

1. Introduction:

Ni spinel ferrites have high electrical resistivity, low dielectric and eddy current losses, which are a major factor for microwave devices (Maghsoudi et al 2013). The substituting of *Al* content into spinel ferrites matrix increases resistivity, lowers the dielectric losses, and thereby lowers the saturation magnetization (Bhosale et al 2006; Mozaffari et al 2003). Moreover, the substituting of *Al* content improves the mechanical strength of ferrite, reduces the magnetic coercivity and leads the ferrite to become softer for high frequency applications. The magnetic properties of spinel ferrites was illustrated according to Wiess domains (Shaath 2012). The magnetizations (*M*) within these domains are defined as the intrinsic magnetization per unit mass (M_i/m) at a certain temperature (*T*) where its value at zero applied magnetic field (*H*) is the spontaneous magnetization (M_s). The value of M_s at zero absolute temperature (*T = 0 K*) is known as the saturation magnetization (M_{St}) (Dawoud et al 2016). For any unmagnetized sample the domains are randomly oriented at *H* = 0 as shown in Figure 1a. When *H* increases, the domains start to rotate in the direction of *H* until all of them are nearly aligned. At this state, the saturation condition corresponds to the state where all domains are in the same direction parallel to *H*, which results to magnetize the sample (Figure 1d) (Shaath 2012).

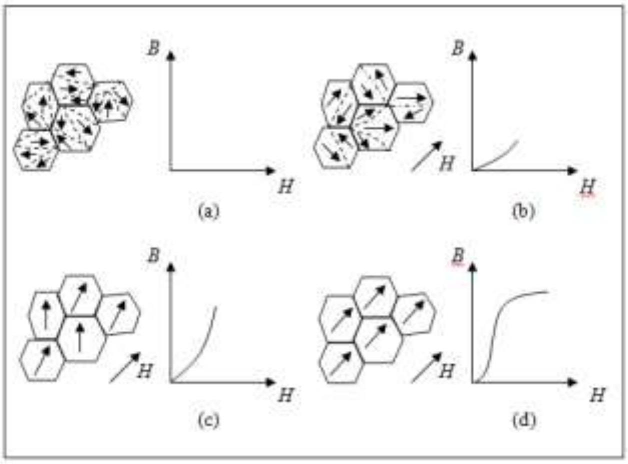

Figure 1 *Random orientation spins of an unmagnetized sample of the ferrimagnetic substance alignment of Wiess domains*.

Source: (Shaath 2012; Dawoud et al 2016).

The chemical formula of spinel ferrite is $D_1^{2+}T_2^{3+}O_4^{2-}$ (Shaat et al 2013; Noto et al 2016; Shaat et al 2014; Dawoud et al 2016; shaat et al 2014, Dawoud et al 2016) has a cubic structure with two sublattices i.e tetrahedral (T_d) and octahedral (O_h). The intensity of M_i in spinel ferrite depends on the spins distribution of magnetic ions into T_d and O_h sublattices (Dawoud 1997). The ions arrangement in the spinel lattice is likely to be most important as is shown in Figure 2 (Shaath 2012). Suppose that intra-sublattice (I_{TT}, I_{OO}) and inter-sublattice (I_{TO}) correspond to molecular-field constants of cations exchange interaction T_d-T_d, O_h-O_h and T_d-O_h, respectively (Chau et al 2008). Therefore, it is thought that the magnetic properties of spinel ferrites depend upon the relative strengths of these types of exchange interactions. When the cations of T_d and O_h are totally magnetic, the inter-exchange interactions I_{TO} are much stronger than I_{TT} and I_{OO} interactions i.e., $|I_{TO}| \gg |I_{OO}| \gg |I_{TT}|$ (Belov et al 1984) . So, spins have a collinear structure in which moments on the T_d are anti-parallel to the moments on the O_h. However, when one of the intra-sublattice interactions becomes comparable with the inter-sublattice interaction it leads to a non-collinear spin structure (Nath et al 2013). The conclusion is that the interactions between the sublattices are stronger than those within them. Further, these interactions between the ions within T_d are the weakest of all. This result thus; supports the assumption that the sublattices magnetizations are antiparallel.

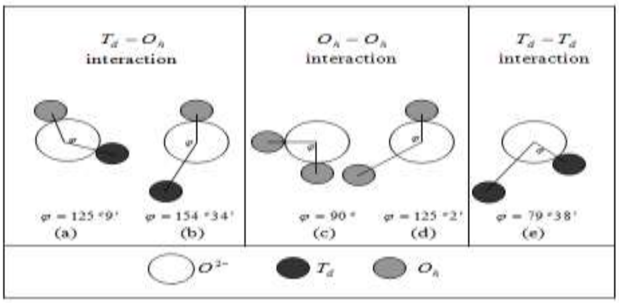

Figure 2 *Superexchange interactions of O^{2-}, T_d and O_h*

Source: (Shaath 2012; Dawoud et al 2016).

2. Experimental:

The mixed polycrystalline ferrites Ni$Al_xFe_{2-x}O_4$, where *x* is the percentage increments of *Al* content which have the value of (0.0, 0.1, 0.2, 0.3, 0.4 and 0.5), were prepared using the standard double sintering method. The pure metal oxides Al_2O_3, NiO and Fe_2O_3 were weighted using a sensitive electric balance (ADAM model PW124) with an accuracy $1\times10^{-4}\,gm$ to produce 25.0 grams. The weighted metal oxides were mixed and then grounded into a very fine powder for 5 hr's. The grounded powder was pre-sintered at 750^oC for 3 hr's soaking time using a laboratory Furnace (BIFATHERM model AC62). Then the prefired powder was regrounded for 3 hr's and then was pressed with a hydraulic press under constant pressure of 3×10^8 pa, by using a small quantity of butyl alcohol as a binding material. The samples were pressed into toroidial shape and were sintered at the 1200 ^oC for 5 hr's, then, were cooled down gradually to room temperature and the samples were polished to obtain uniform parallel surfaces to measure the magnetic properties. The magnetization was measured at room temperature as a function of *H* in the range (0-40) (*Oe)* at a constant magnetizing frequency (*f* = 50 *Hz*) and applied AC current (I_{AC}) over the range of (0–5) A.

3. Results and Discussion:

3.1 Magnetic Properties:

The relation between the magnetization [M (emu/g)] and the applied magnetic field intensity (H) were studied for the prepared ferrite samples of the Ni$Al_xFe_{2-x}O_4$ system at room temperature in the range of [0–40] (Oe). The variation of *M* and *H* for all samples was illustrated in Figure 3. Its noticed from the Figure that *M* is directly proportional to *H*. It is reported that the lower value of *M* indicates a higher degree of surface disorder and a lower densification stage (Richa et al 2016). Thus, the value of *M* increases with increasing of the crystallinity of ferrite materials.

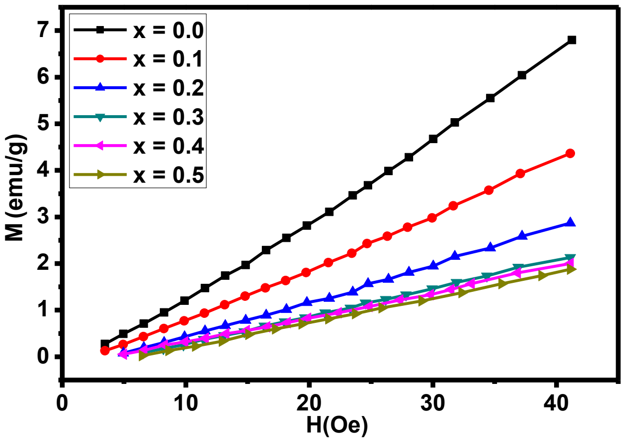

Figure 3 *Variation of M with H for the samples of x = 0.0, 0.1, 0.3, 0.4 and 0.5*.

According to the results in Figure 3, the variation of *M* on the ratio of *Al* content in matrix $NiAl_xFe_{2-x}O_4$ is represented in Figure 4 for different values of *H*. It can be noted from this Figure that *M* decreases for all samples while the concentration of *Al* content increases in matrix. The same behavior was seen earlier by many researchers (Maghsoudi et al 2013; Dawoud et al 2016;. Dawoud et al 2006) Depending on the particle sizes, which decreased with increasing *Al* concentrations (Maghsoudi et al 2013). This is because the ionic radius of $Al^{3+}(0.51\,A^o)$ is smaller than the ionic radii of $Fe^{3+}(0.67\,A^o)$ and $Ni^{2+}(0.72\,A^o)$ (Patange et al 2011).

The decreasing of the magnetization with increasing of Al^{3+} ions for all samples can be explained by Neel's two-sublattice model of the ferrimagnetic materials theory (Shaath 2012; Dawoud et al 2016). According to this model, the magnetic ordering in the simple spinel ferrites is based on T_d and O_h sublattices. The net magnetization are the difference between two of them provided that, they are collinear and anti-parallel to each other.

In mixed *Ni-Al* ferrite, Al^{3+} and Ni^{2+} ions concentrate preferentially in O_h sublattice in the cubic spinel lattice (Deraz et al 2012). As *Al* content increases, the exchange interactions are weakened and spins of O_h sublattice are no longer held rigidly parallel to the spins of T_d sublattice. Thus; the decrease of magnetization is attributed to that $T_d - O_h$ exchange interaction becomes weaker or comparable with $O_h - O_h$ exchange interactions. The decrease in a O_h sublattice moment, interpreted as a spin departure of co-linearity causes the effect known as canting, this effect also described in samples of *Li-Cu* ferrite (Mazen et al 1999).

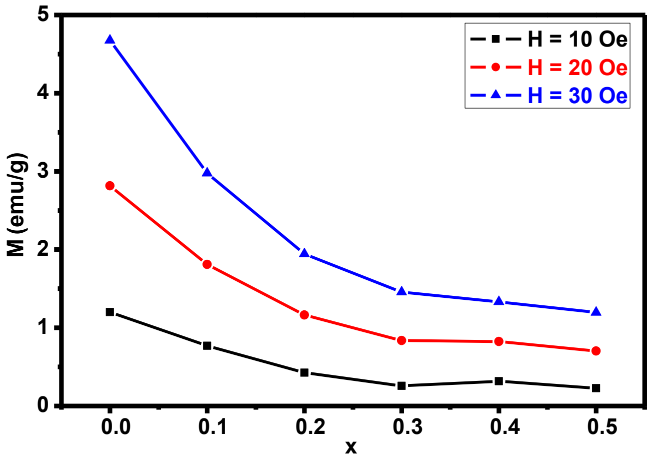

Figure 4 *Variation of M with the composition x for different values of H (Oe)*

3.2 Net Magnetization and Magnetic Moment:

The net magnetization (M_{net}) for the ferrimagnetic materials can be calculated from individual magnetization, M_T and M_O of T_d and O_h sublattices, respectively. Thus, according to Neel's theory M_{net} at $T = 0K$ has the form (Shaath 2012; Dawoud et al 2016)

$$M_{net} = |M_O - M_T| \tag{1}$$

where $M_{T,O}(0) = \sum_{i,j} N_{i,j} g \mu_B S_{mi,mj}$ with N_i and N_j are the numbers of the magnetic ions, S_{mi} and S_{mj} are spin quantum numbers of each ion in O_h and T_d sub-lattices, respectively, and g is the lande' splitting factor or spectroscopy splitting factor, which is approximately equal to 2.0003 for the free electron. M_O and M_T are calculated for the Al - Ni spinel ferrite according to the cations distribution formula $(Fe^{3+}_{1-t}Al^{3+}_t)\{Ni^{2+}Al^{3+}_{x-t}Fe^{3+}_{1-x+t}\}O^{2-}_4$, with $(0.0 \le t \le 0.1)$,

$$M_T = [0.1S_{Al} + (0.9)S_{Fe}]2\mu_B \quad (2)$$

and

$$M_O = [(S_{Ni} + (x-0.1)S_{Al} + (1.1-x)S_{Fe}]2\mu_B \quad (3)$$

Substituting equations (2) and (3) into the equation (1), which follows

$$M_{net} = [S_{Ni} + (0.2-x)S_{Fe}]2\mu_B \quad (4)$$

where $S_{Fe} = 5/2$ and $S_{Ni} = 1$ for Fe^{3+} and Ni^{2+} ions, respectively. The variation of M_O, M_T and M_{net} with the composition *x* is poltted in Figure 5. From this Figure, it is clear that, with increasing of composition *x*, both M_O and M_{net} decrease, while M_T is nearly constant. The variation of M_{net} with Al^{3+} ions could be explained by assuming that, by increasing of Al^{3+} ions in the samples the relative number of the Fe^{3+} ions will decrease in O_h sublattice, and will slightly decrease in T_d sublattice. This tends to decrease M_O but M_T is nearly not change. Therefore, M_{net} would dilute linearly with increasing of Al^{3+} ions causing M_{net} decreases due to the introduction of Fe^{3+} ions having $5/2\mu_B$ spin magnetic moment (Nath et al 2013). It can be concluded that, as Al^{3+} ions replaces sum of Fe^{3+} magnetic ions in T_d sublattice, and replaces more Fe^{3+} ions in O_h sublattice, as a result M_{net} decrease.

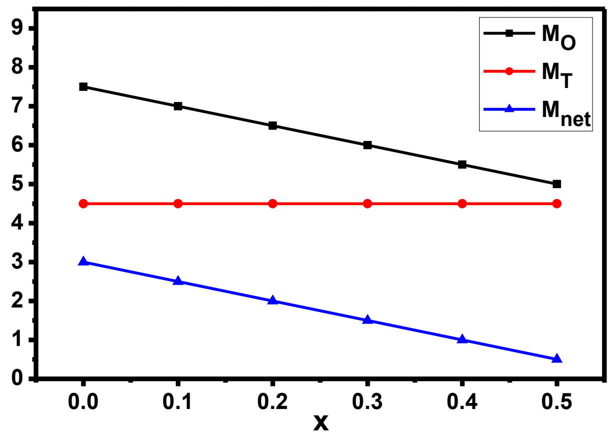

Figure 5 *Variation of M_O, M_T and M_{net} with Al ratio "x"*

3.3 Relative Permeability:

The relative permeability (μ_r) describes the magnetization behavior of the material during the change of the exposed *H*. μ_r is defined as the ratio between the induction field (*B*) and *H*, which is given by $\mu_r = B/\mu_o H$, where μ_o is the permeability of free space. The variation of *H* and μ_r for the present ferrite samples are shown in Figure 6, which are increased with an incearse of *H*. μ_r decrease with increasing of *Al* content (low spin ion) at different values of *H* as shown in Figure 7. The increment of μ_r could be related to the alignment effect of *H* on the ionic spins. In line with, the increase of *H* causes rapid increasing of *B*, which causes increasing of μ_r, the maximum value of μ_r is found in the sample of *x* = 0.0 with composition $Ni\ Fe_2O_4$. This means that the samples have less Al^{3+} ions have the highest spin ordering (highest intrinsic magnetization) compared with the other samples. Accordingly, the increase of *H* might cause a slight increase of *B* gives rise to a distinct increase in μ_r.

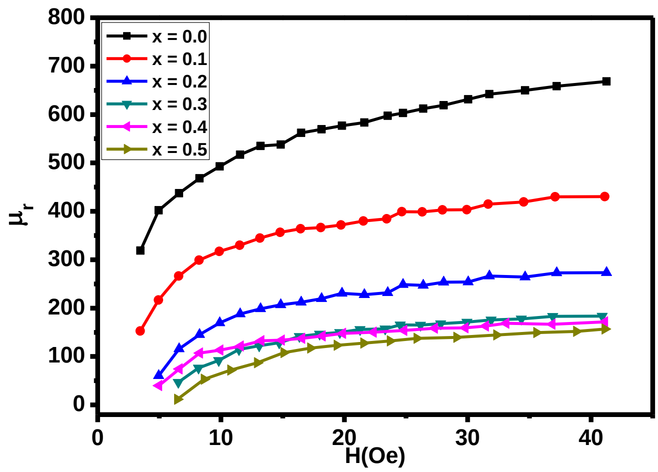

Figure 6 *Variation of μ_r with H for samples with composition x = 0.0, 0.2, 0.3, 0.4 and 0.5*

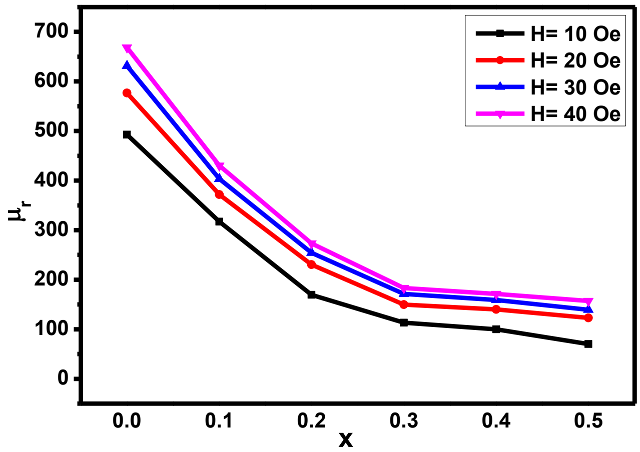

Figure 7 *Variation of μ_r with compostion x at different values of H*

4. Conclusion:

Substitution of the non-magnetic Al^{3+} ions in *Ni* spinel ferrite has a tremendous influence such the magnetic properties. From this study, we concluded from the obtained results that, the magnetization decreased with the increasing of the Al^{3+} ions. The decreasing of the magnetization was explained based on Neel's two sublattices model. The increase of *H* causes a pronounced increase of μ_r which decrease with increasing of *Al* content (low spin ion). Furthermore, *Al* content has significant influence on the magnetic properties for *Ni* ferrites, so, the mixed *Ni-Al* spinel ferrite is considered a soft ferrite material, which is proved an interesting material for technological and scientific applications

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دراسة تأثير إضافة عنصر الألومنيوم على الخواص المغناطيسية لنيكل اسبنل فيرايت المحضرة بالطريقة التقليدية

لقد استخدمت الطريقة السيراميكية التقليدية في تحضير مجموعة من عينات الأمونيوم نيكل فريت $NiAl_xFe_{2-x}O_4$ حيث أن x هي نسبة الزيادة في أيونات الألومنيوم وذلك من خلال خلط الأكاسيد عالية النقاء وهي أكسيد الألمونيوم وأكسيد النيكل وأكسيد الحديد.
لقد تم قياس الخواص المغناطيسية في درجة حرارة الغرفة كمتغير في المجال المغناطيسي في الفترة (0-40) أورستد عند تردد ثابت *50* هرتز وعند تيار متردد من (*0–5*) أمبير. لقد أوضحت النتائج أن تمغنط العينات تزيد مع زيادة المجال المغناطيسي. وكذلك تبين بأن قيمة التمغنط يقل مع زيادة أيونات الألومنيوم. وتم تفسير ذلك حسب نظرية (Neel's two-sublattice model). كما تم دراسة العلاقة بين المجال المغناطيسي والنفاذية المغناطيسية النسبية ووجد أنها تقل مع زيادة أيونات الألومنيوم. وأوضحت النتائج بأن هناك تأثير ملموس على الخواص المغناطيسية النيكل فريت بإضافة أيونات الألومنيوم مما يجعل هذه العينات من الأهمية بمكان بحيث تستخدم في الصناعات العلمية والتكنولوجية.

كلمات مفتاحية:
الخواص المغناطيسية،
اسبنل فيرايت،
النفاذية المغناطيسية النسبية،
نموذج نيل.