



## EFFECT OF ALUMINUM SUBSTITUTION IN NICKEL FERRITE NANOPARTICLES ON THERMOELECTRIC PROPERTIES

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### Abstract

*Aluminum substituted nickel ferrite having generic formula  $NiAl_xFe_{2-x}O_4$  ( $0 \leq x \leq 1.0$ ) has been synthesized by sol-gel auto combustion technique using high purity nitrates. Electrical transport properties of aluminum substituted nickel ferrite been investigated from room temperature to well beyond the Curie temperature. Based on the Seebeck coefficient the ferrites under investigation have been classified as P-type semiconductors.*

**Keywords:** Nickel ferrite, thermoelectric properties, seebeck coefficient etc.

### Introduction

The rapid development of mobile communication and information technology, the electronic component with small size, high efficiency, and low cost are urgently demanded [X. Qi, et al, 2002.] Depending on their radius, volume fraction, and band offset, a nanoparticle doped sample can either suppress or enhance the electrical conductivity in comparison with the doped bulk sample, the advantage of incorporating nanoparticles inside thermoelectric materials is to reduce the lattice thermal conductivity and enhance the Seebeck coefficient due to electron energy filtering [Mona Zebarjadi, et al, 2009]. The formula unit of the spinel structure of ferrite is  $AB_2O_4$ , where oxygen ions form a cubic-close-packed cage and interstitial positions are occupied by metal ions in two non-equivalent lattice sites, known as tetrahedral (A) and octahedral (B) sites [I Panneer Muthuselvam and R N Bhowmik, 2010]. Ferrites have many applications as both low and high frequency devices and play a very much useful role in many technology and magnetic applications because of their high electrical resistivity and sufficiently low dielectric losses over a wide range of frequencies [Y. Purushotham et al, 1995]. In accordance of these facts, it is thought that systematic investigation of electrical properties such as electrical conductivity and thermoelectric properties would also very much essential. Study gives us the conduction mechanism are very much useful in these materials. The present investigation is designed to examine the

thermoelectric properties as a function of temperature and results and discussion of thermoelectric properties presented in this paper.

## **Experimental**

### ***Materials***

Ferric nitrate ( $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ), nickel nitrate ( $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ), citric acid ( $\text{C}_6\text{H}_8\text{O}_7 \cdot \text{H}_2\text{O}$ ), aluminium nitrate ( $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ), ammonium hydroxide solution ( $\text{NH}_4\text{OH}$ ) were supplied by Merk. All the chemicals were of analytical reagent (AR) grade and are used in its present form.

### ***Synthesis by Sol-gel auto-combustion method***

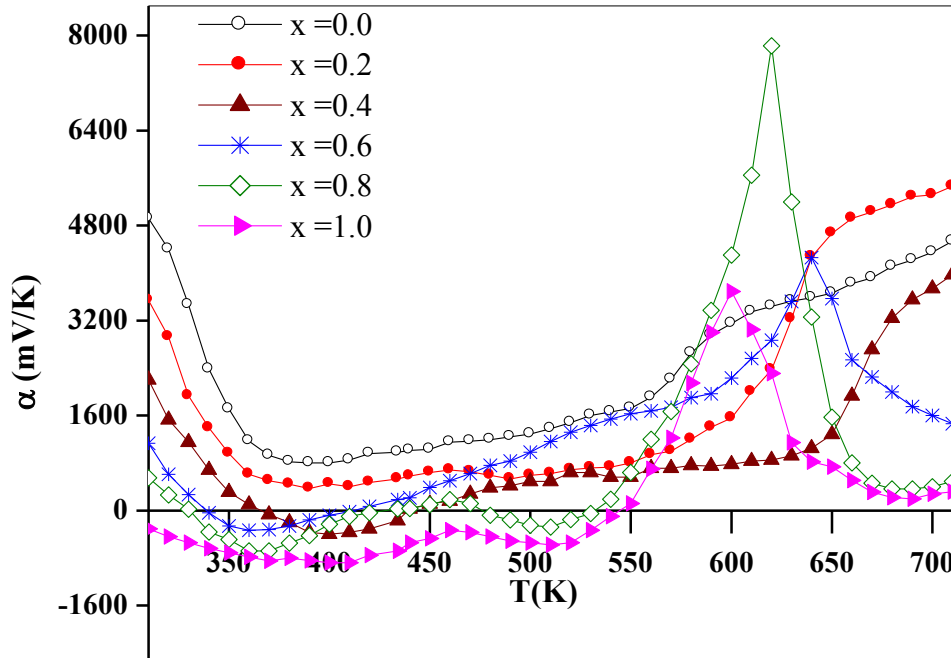
Aluminium substituted nickel ferrite ( $\text{NiAl}_x\text{Fe}_{2-x}\text{O}_4$ ) nano-particles were prepared by the sol-gel auto-combustion technique. The metal nitrate (i.e. nickel nitrate, ferric nitrate and aluminium nitrate) to citric acid ratio was taken as 1:3. The metal nitrates were dissolved together in a minimum amount of de-ionized water required to get a clear solution. Citric acid was then mixed with the metal solution and ammonia solution was slowly added to adjust the pH to 7. This solution was heated on a hot plate magnetic stirrer with continuous stirring at 90–100°C. During heating the solution became viscous and finally formed a very viscous green gel and began to bubble and the gel get auto combusted in few seconds. The dry and fluffy powder remains in the beaker. The as prepared powder was annealed at 550°C for 5 hrs to get single phase spinel product and used for further characterization.

### ***Thermoelectric properties***

Thermoelectric power ( $\alpha$ ) measurement studies were carried out over a temperature range 300 K- 710 K by the differential method. According to the relation  $\alpha = \frac{\nabla V(\mu\text{V})}{\Delta T(\text{K})}$  the thermo-emf ( $\nabla V$ ) was measured with the help of digital micro-voltmeter with an accuracy of  $\pm 3\%$ . The temperature difference between two ends of the sample was kept at 10 K throughout the temperature range studied.

**Results and Discussion**

*Thermoelectric Power Study*



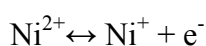
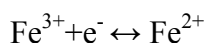
**Figure 1: Thermoelectric power versus hot junction of temperature of NiAl<sub>x</sub>Fe<sub>2-x</sub>O<sub>4</sub> ferrite nanoparticles**

The p- or n-type nature of the charge carriers below and above the transition temperature was determined from the sign of Seebeck coefficient as in Fig. 1. The presence of p and n-type charge carriers defines the two regions of temperature dependence of conductivity in which the sample is changed from p- type to n-type and vice versa[M. A. Hiti, 1968]. The Seebeck coefficient for all the samples is increases linearly by increasing temperature from 300 K – 700 K as in Fig 1, which means that dominant conduction process is due to impurities. The first observation suggests that for x = 0.0 to 0.6 ‘□’ is positive.

**Table 1: Seebeck coefficient ( $\alpha$ ) and Transition temperature ( $T_s$ ) for  $\text{NiAl}_x\text{Fe}_{2-x}\text{O}_4$  ferrite nanoparticles.**

x	Seebeck Coefficient ' $\alpha$ ' (mV/K)				$T_s$
	310K	400K	600 K	700 K	
0.0	4920	800	3150	4350	--
0.2	3540	450	1560	5320	--
0.4	2200	-400	780	3740	--
0.6	1130	-80	2230	1600	640
0.8	550	-230	4300	400	620
1.0	-310	-880	3690	278	600

This means that majority charge carriers are holes or p-type conduction mechanism is dominant. The Seebeck coefficient is independent of temperature at lower temperature region [M. A. Ahmed, A. A. I. Khalil *et.al*, 2007]. For  $\text{Al}^{3+}$  content 0.8 and 1.0, ' $\alpha$ ' is negative in low temperature range i.e. 300 to 540 K. This means that in this temperature range n-type conduction is dominant or majority charge carriers are electrons. Beyond 540 K both the compositions show p-type conduction. It means that negative and positive Seebeck coefficient suggesting that both p-type and n-type carriers are responsible for conduction mechanism [K.A. Mohammed, A. D. Al-Rawas *et.al*, 2012] in the present samples may as follows:



The second observation suggest that for all the samples the Seebeck coefficient ( $\alpha$ ) increases as increase in temperature up to a certain temperature, which is designated as  $T_s$  (K). The second hump was not observed because our thermoelectric power measurement was out of range. However beyond this temperature the Seebeck coefficient ( $\alpha$ ) starts to decrease with increase in temperature.

The values of Seebeck coefficient of the aluminium substituted nickel ferrite samples are depicted in the Table 1. From Table, Seebeck coefficient ( $\alpha$ ) decreases by increasing  $\text{Al}^{3+}$  content (310 K). It is clear from table that the variation in the charge carriers between p and n-type charge carriers is nearly independent on  $\text{Al}^{3+}$  type or size upto 0.8. The Seebeck coefficient increases after 400 K showing the hump which may corresponds to Curie temperature of the sample. The negative values of thermoelectric power found over the entire temperature range studied shows that the majority of charge carriers are holes. On the basis of

its positive sign, the above ferrites have been classified as p-type semiconductors. Thus, the conduction mechanism for p-type is predominant due to holes.

### **Conclusion**

The Seebeck coefficient for all the compositions under investigation is found to be positive showing majority charge carriers to be p-type. The substitution of aluminium ions reside at the B-site and causes decrease in the value of the Seebeck coefficient which is perhaps attributable to the decrease in the population of  $\text{Fe}^{3+}$  ions at the B-site.

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