

## Effect of Time on the Structural and Magnetic Properties of Barium Ferrite substituted by Zn

Mohammed Abdul Malek Abdul Rahman

AlMaarif University College, Iraq

Department of Communication and Computer Engineering, Cihan University – Erbil, Kurdistan Region – Iraq.

[dr.mohamed.saadi@gmail.com](mailto:dr.mohamed.saadi@gmail.com)

### ABSTRACT:

Barium ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) substituted by  $\text{Zn}^{2+}$  ( $\text{BaZn}_x\text{Fe}_{12-x}\text{O}_{19}$ ) where ( $x=1.5, 2.5$ ) were prepared by solid state reaction (conventional ceramic techniques). The hexagonal Ferrite has been prepared in sintered temperature of  $1100^\circ\text{C}$  and  $1200^\circ\text{C}$  at period 10 hours. Conducted tests on the structural and magnetic properties for samples before and after the passage of ten years in preparation, where preserved at room temperature to determine the effect of time on those properties. The structural tests on the prepared samples to identify the crystalline phases for ferrite particles have been carried out by X-ray diffraction (XRD). The patterns of XRD obtained that the ferrite has a single phase and it has shown that there is a complete growth of the ferrite phases at  $1200^\circ\text{C}$ . To obtain detailed information about the morphology and to verify the size of the particles, a set of micrographs were taken by scanning electron microscopy (SEM). These micrographs demonstrate that the surface is homogenous and the grain size increases with temperature and with the extent of substitute matter ( $x$ ). Differential Thermal Analysis (DTA). Magnetic properties have also been measured by using a vibrating sample magnetometer (VSM). From hysteresis loop the magnetic properties  $H_c$ ,  $M_s$ ,  $M_r$  and  $\mu$  are calculated. The magnetic properties have been improved by adding Zn to the barium ferrite. It has been found that the ferrites will be changed from hard to soft by adding Zn ions (when  $x \geq 2.5$ ). The coercivity  $H_c$  has been decreased and saturation magnetization has been increased with increasing of sintering temperature and substitution ratio ( $x$ ).

**Keywords:** Barium Ferrite; Zinc; DTA; XRD; SEM; VSM.

### 1. INTRODUCTION

Ferrites are ferromagnetic materials containing predominantly oxides of iron along with other oxides of barium, strontium, manganese, nickel, zinc, lithium and cadmium. Barium hexaferrite powders have been investigated as a material for permanent magnets, microwave absorber devices and recording media. Barium hexaferrite is widely used due to its high stability, excellent high-frequency response, narrow switching field distribution and its temperature coefficient of coactivity in various applications. Barium ferrite with a hexagonal molecular structure has a fairly large magneto crystalline an isotropy, a high Curie temperature and a relatively large magnetization, as well as chemical and corrosion stability. The conventional ceramic methods, i.e., high- energy ball milling and chemical processes such as chemical co-precipitation, the hydrothermal process, the sol-gel process, etc., were employed to obtain high- quality barium ferrite [1,2,3]. Ferrites are ideally suited for making device like inductor cores, circulators, memory devices and also for various microwave application. A reduction in size and weight of power supplies can be achieved by using switched mode or resonant concepts. For the voltage conversion several circuit designs are in use with ferrites as transformer core materials. Transformer ferrites must show low energy losses at high induction levels at higher and

higher frequencies (Globus 1977; Snelling and Giler 1983; Goldman 1990; Stopples 1996; Yasuhara and Takagawa 2000). This requires development of new ferrite materials with constantly improving loss characteristics [4].

Although the saturation magnetization of ferrites is less than that of ferromagnetic alloys, they have advantages such as applicability at higher frequency, lower price and greater electrical resistance. Ferrites are classified into two groups, namely magnetically soft and hard. Furthermore, depending on their crystal symmetry, they may be cubic, hexagonal or orthorhombic. Barium ferrites belong to the class of hexagonal ferrite. They are normally prepared by using ceramic technique. Type-M hexaferrites of barium and strontium are very important components in permanent magnets, magnetic recording media, and microwave and high frequency devices. The attractive properties of these materials are high magnetization, remanence, coercivity and electrical resistivity. The materials also have great resistance to high temperature, humidity, chemicals and corrosion [5,6,7].

The ferrites are homogeneous ceramic materials dark gray or black color is made up of a blend of different types of oxides with iron oxide and have different crystalline structures [1]. It is also considered ionic compounds and its magnetic properties due to the magnetic ions they contain. Ferrite matters are materials with high permeability of the RF (radiofrequencies) and microwaves, therefore used to make microwave components of the circuits which are not mutually exclusive (nonreciprocal). The high permeability due to the distribution of the wall of the field controlled by the sintering density, and by increasing the size of particleboard [8].

The hexagonal ferrite  $\text{BaFe}_{12}\text{O}_{19}$  and its substituted derivatives have been considered as candidates with the most potential because of their chemical stability and suitable magnetic characteristics [8,9].

The Zn substitution has been previously studied and researchers [9,10] have agreed that this cationic combination yields materials with suitable characteristics for magnetic recording [11]. Where  $\text{Zn}^{2+}$  ions have been reported to have a positive influence on the saturation magnetization [1]. As pointed out by Rane et al. [12], the magnetic properties of substituted BaM directly depend on the electronic configuration of the substituting cations and on their preference to occupy the different Fe sub lattices of the hexagonal structure. It has also been highlighted that the magnetic properties of BaM are related to the occupancy of the  $\text{Fe}^{3+}$  ions at the different sub lattices of the magneto plumber structure [13]. In general, substitutions lead to a decrease of  $H_c$  through the reduction of the magneto crystalline anisotropy of the BaM. Moreover, the effects of substitutions, on crystallite size, aspect ratio (diameter/thickness), ordering temperature and magnetization are also observed. Some of these changes are closely related to the preparation procedure [14].

In this paper Zn substituted Barium Ferrite ( $\text{BaFe}_{12}\text{O}_{19}$ ) using conventional ceramic techniques and investigated by differential thermal analysis (DTA), X-ray diffraction (XRD), scanning electron microscopy (SEM) and magnetic measurements by vibrating sample magnetometer (VSM). The approach to saturation law was used to determine zero-field saturation magnetization  $M_s$ , anisotropy field  $H_a$ .

## 2. EXPERIMENTAL

Barium Ferrite  $BaFe_{12}O_{19}$  substituted by zinc ions as  $BaZn_xFe_{12-x}O_{19}$  where ( $x=1.5, 2.5$ ) was prepared by conventional ceramic techniques. Where raw materials of mixed oxides or salts of the elements such as chlorides or carbonate or nitrates after grinding the individual to equal their size, which facilitates the process of granule homogeneity between these compounds during the mixing. These materials included on the ferric oxide  $Fe_2O_3$ , barium oxide  $BaO$  and zinc oxide  $ZnO$  of purity greater than 98%. Ratios were calculated accurately by weighing of materials involved in the preparation of ferrite samples using an electronic balance its sensitive up to 0.0001 gram.

The materials mixed with de-ionized water and then dried at  $1100\text{ }^\circ\text{C}$  overnight. For the formation of  $BaFe_{12}O_{19}$  phase, the mixture were annealed at a rate of  $10\text{ }^\circ\text{C}/\text{min}$  in static air atmosphere up to different temperatures ( $1100$  and  $1200\text{ }^\circ\text{C}$ ) and then maintained at different periods 10 hours.

The crystallite phases present in the different samples were identified by X-ray diffraction (XRD) on a Brucker axis D8 diffract meter with crystallographic data software to pas 2 using Cu-Ka ( $1/41.5406$ ) radiation operating at  $40\text{kV}$  and  $30\text{mA}$  at a rate of  $2^\circ/\text{min}$ . The diffraction data were recorded for  $2\theta$  values between  $20^\circ$  and  $80^\circ$ . The ferrite particles morphology was observed using the scanning electron microscope (SEM) to study the surfaces of samples, to find out effect of substitution on the structural properties, to measure the particle size of ferrite .

The magnetic properties of the ferrites were measured at room temperature using a vibrating sample magnetometer (VSM; 9600-1 LDJ, USA ) in maximum applied field of  $15\text{ G}$  . From the obtained hysteresis loops, the saturation magnetization ( $M_s$ ), remanence magnetization ( $M_r$ ) and the coercive field ( $H_c$ ) were determined.

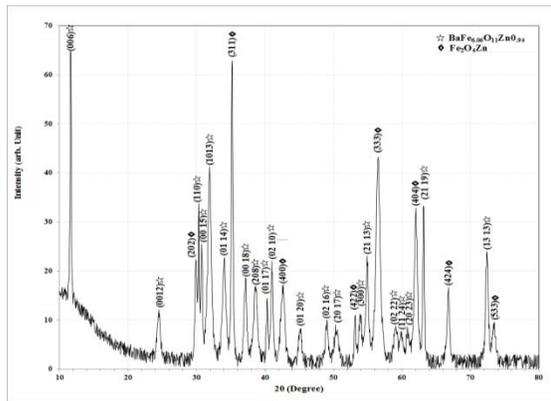
## 3. RESULTS AND DISCUSSION

### 3.1. Structure and Particle Morphology of the Sample

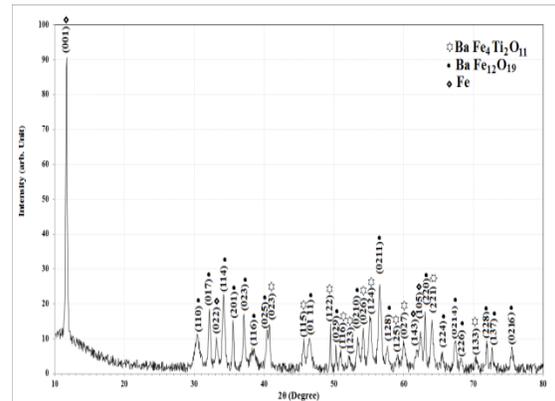
Fig(1) shows the XRD patterns of the prepared ferrite powders for  $BaZn_xFe_{12-x}O_{19}$  with  $x=1.5$  and  $2.5$  ,sintered at  $1100$  and  $1200\text{ }^\circ\text{C}$  for a period  $10\text{h}$  .Fig(1-a) at  $x=0$ , obtained a hexagonal ferrite of  $BaFe_{12}O_{19}$  with a survival small amount of  $Fe_2O_3$ . These were according with ASTM card and with published papers [2,14], so it is used as comparison.

Fig (1-b,c), shows the XRD pattern of barium ferrite substituted by Zn ions at  $x=1.5$  and  $2.5$  .A new phase shown in the figure not related to the hexagonal phase, so these phases may be related to the Y-Ferroxplana phase of the formula  $Ba_2Zn_2Fe_{12}O_{22}$  as well as hexagonal phase of barium ferrite.

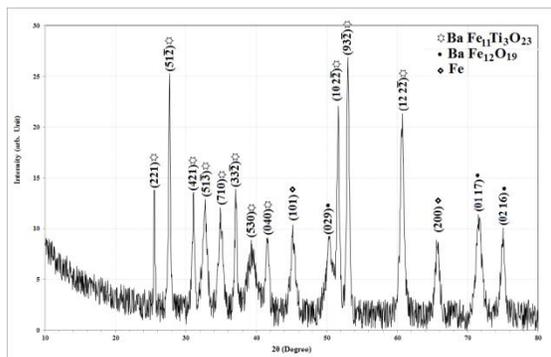
However, the XRD peaks became weaker and broader along with the shifts in peak positions alterations of relative peak intensities  $x$  was increased (fig(1)), the same results were observed by yue et al [2].



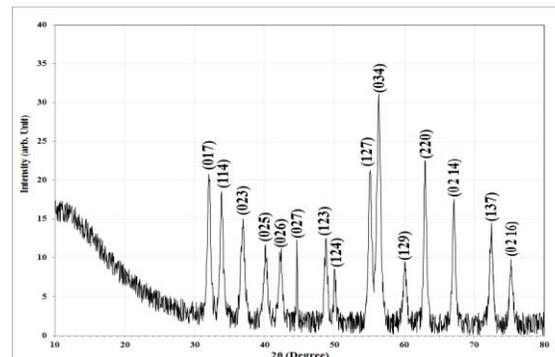
**Fig. 1-a X=1.5 T=1100 c**



**Fig. 1-b X=1.5 T=1200 c**

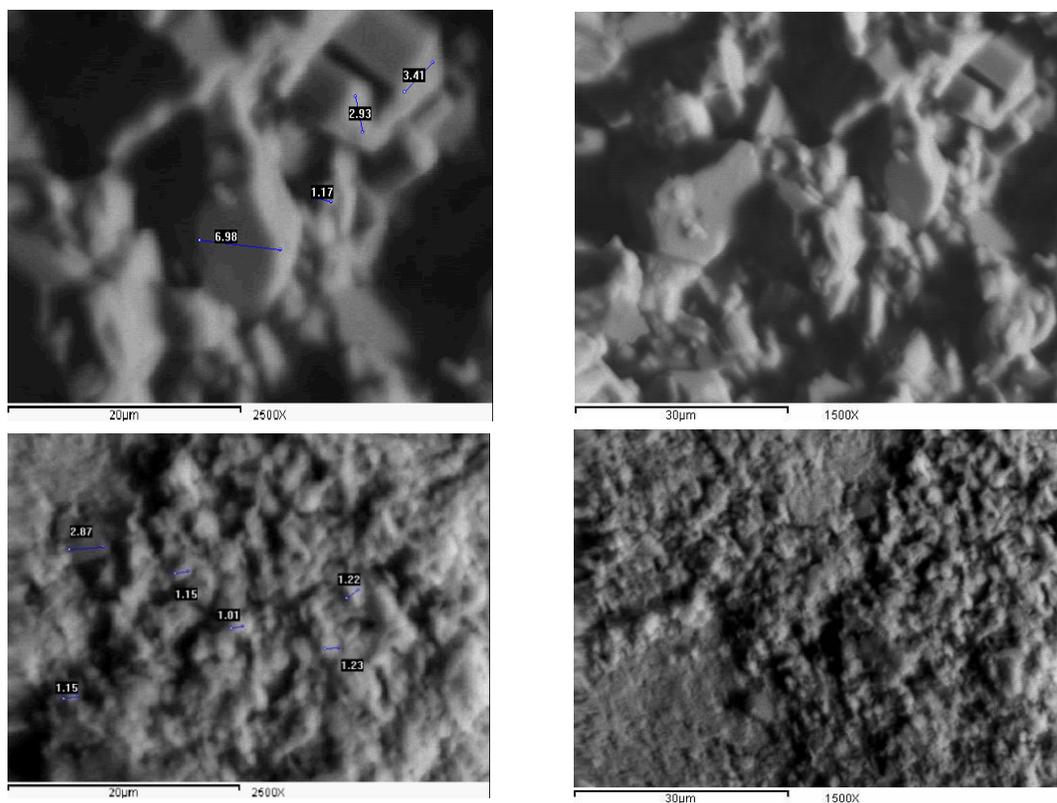


**Fig. 1-c X=2.5 T=1100 c**

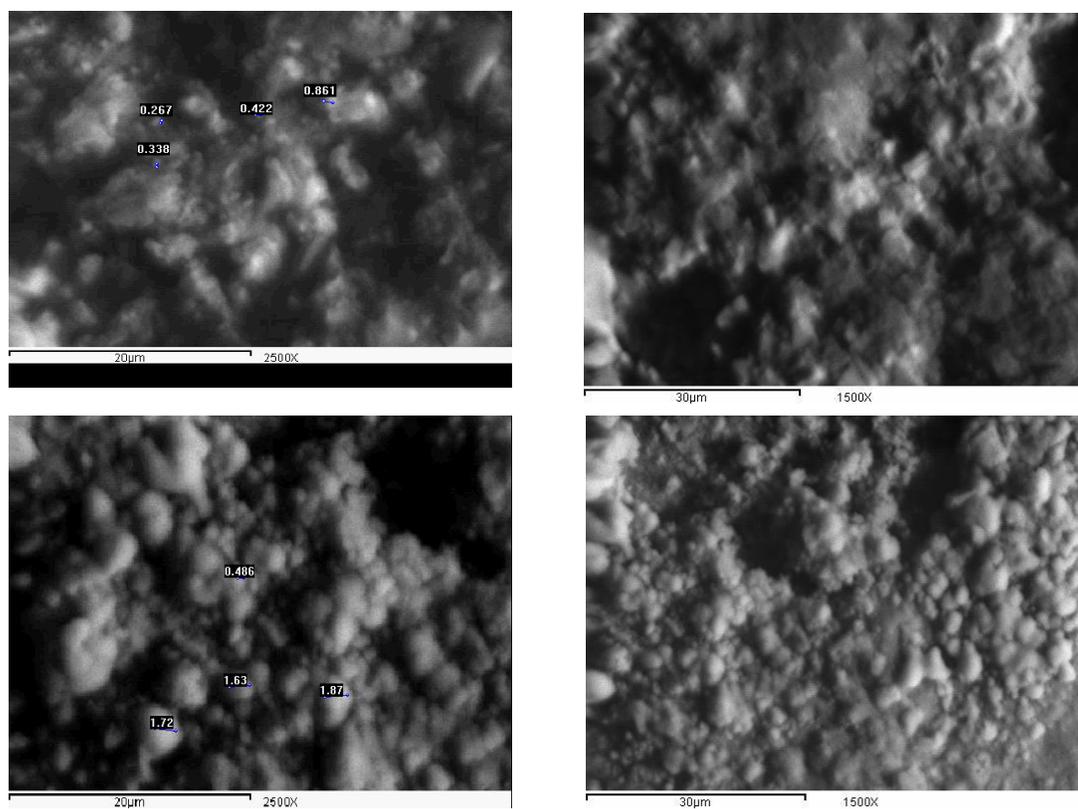


**Fig. 1-d X=2.5 T=1200 c**

Representative SEM images of the series are shown in Fig(2) and Fig(3). The phase were perfect crystalline. As shown in the figures the plane dimensions of the most grains were ranged between (6.98-1.17  $\mu\text{m}$ ) at x of 1.5 and T is 1100 c. while at the same x and at T of 1200 c, it was decreased to (2.87-1.01 $\mu\text{m}$ ) as shown in fig(2). For increasing doping concentration of larger values of x, the grain size was decreased as shown in fig(3). The grain size at x values of 2.5 at 1100 ranged between (1.87-0.4 $\mu\text{m}$ ). Also it decreased with increasing the sintering temperature to 1200 c, to range between (0.68-0.26 $\mu\text{m}$ ), these behavior was agree with which found by yue et al [2].



**Fig (2) SEM micrographs of samples (a) X=1.5 T= 1100 c (b) X=1.5 T= 1200 c**



**Fig (3) SEM micrographs of samples (a) X=2.5 T= 1100 c (b) X=2.5 T= 1200 c**

### 3.2. Magnetic properties.

Hysteresis loop of  $BaZn_xFe_{12-x}O_{19}$  are shown in fig (4). The hard ferrite became softer as more  $Fe^{3+}$  substituted by  $Zn^{2+}$  dopant. A magnetic parameters ( $H_c, M_r, M_s, \mu_i$ ) were calculated from the hysteresis loop as shown in table (1). Note that the magnetic parameter changed with  $x$  and  $T$ . the change in the grain size effect on the anisotropy, which changed the coercivity ( $H_c$ ) and other parameters,  $H_c$  and  $M_s$  were decreased with increased  $x$  from 0 to 1.5 at 1100 c, while  $M_r$  and  $\mu_i$  were increased in this interval with a hard ferrite. When  $T=1200$  c the magnetic parameters  $H_c, M_r$  and  $M_s$  were decreased but  $\mu_i$  was increased for hard ferrite compared with its value at  $x=0$ . When the doping concentration  $x$  increased to 2.5 the values of ( $H_c, M_r, M_s$ ) also decreased and  $\mu_i$  increased but with soft ferrite at sintering temperatures of 1100 c and 1200 c°. As shown in table (4). Our findings agree better with this reported in refs. [2,14].

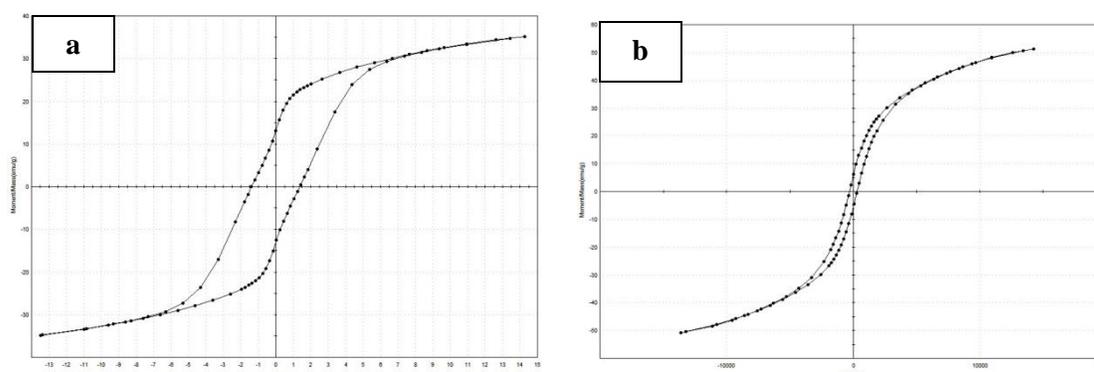


Fig (4) Hysteresis loops (a) X=1.5 T= 1100 c (b) X=1.5 T= 1200 c

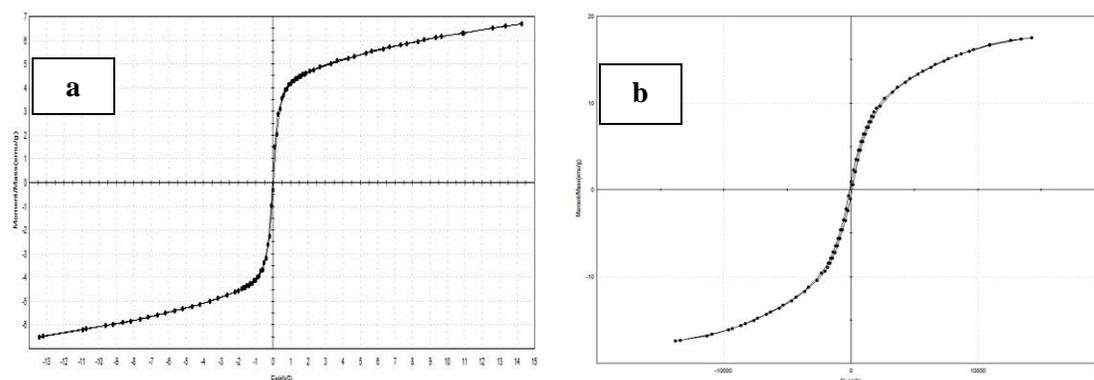


Fig (4) Hysteresis loops (a) X=2.5 T= 1100 c (b) X=2.5 T= 1200 c

Table (1) Magnetic Parameters

X	T (°C)	Hc (KOe)	Ms (emu/gm)	Mr (emu/gm)	$\mu_i$	Type
1.5	1100	1383.9	34.987	13.016	0.9	Hard
1.5	1200	288.48	51.030	5.4487	2.6	Soft
2.5	1100	29.110	6.5909	0.36338	1.3	Soft
2.5	1200	94.785	17.484	0.76371	3.1	Soft

#### 4. CONCLUSION

A  $\text{BaZn}_x\text{Fe}_{12-x}\text{O}_{19}$  ferrite can be prepared by a ceramic method with a hexagonal phase. The grain size of the prepared particles depends on the values of  $x$  and  $T$ . to give values ranged between  $(0.26-6.98\mu\text{m})$  as pointed by SEM technique. Hysteresis loop shows that the ferrite change from hard to soft with increasing  $x$  and  $T$ . Also the magnetic parameters changed with  $x$  and  $T$  to decrease a value of  $(H_c, M_r, M_s)$  and increase the value of  $\mu_i$  with increasing  $x$  and  $T$ .

#### REFERENCES

- [1] Charles Kittel "Introduction to solid state physics" 5<sup>th</sup>. Ed., John Wiley & Sons. New York, (1976).
- [2] Yue Liu a, Michael G.B. Drew b, Ying Liu a,n "Preparation and magnetic properties of barium ferrites substituted with manganese, cobalt, and tin" [Journal of Magnetism and Magnetic Materials 323 \(2011\) 945–953](#).
- [3] Aylin Gurbuz<sup>1</sup>, Nurhan Onar<sup>2</sup>, Ismail Ozdemir<sup>3</sup>, Abdullah Cahit Karaoglanli<sup>3</sup>, Erdal Celik<sup>1</sup> " STRUCTURAL, THERMAL AND MAGNETIC PROPERTIES OF BARIUM-FERRITE POWDERS SUBSTITUTED WITH Mn, Cu OR Co AND X (X = Sr AND Ni) PREPARED BY THE SOL-GEL METHOD" Professional article/Strokovni, MTAEC9, 46(3)305(2012).
- [4] S R MURTHY "Development of low-power loss Mn–Zn ferrites using microwave sintering method" Bull. Mater. Sci., Vol. 26, No. 5, August 2003, pp. 499–503. © Indian Academy of Sciences.
- [5] P. Kerschla\*, R. Gro. ssingerb, C. Kussbachb, R. Sato-Turtellib, K.H. Mu. llera, L. Schultz " Magnetic properties of nanocrystalline barium ferrite at high temperatures" Journal of Magnetism and Magnetic Materials 242–245 (2002) 1468–1470.
- [6] H. Ovalioglua, H. Sozerib , M. Kabaera and I. Kucuka," Magnetic Properties of Nano-Crystalline Barium Ferrite Synthesized by Different Synthesis Route" 14th Czech and Slovak Conference on Magnetism, Košice, Slovakia, July 6–9, 2010.
- [7] NURHIDAYATY MOKHTAR, MUSTAFFA HJ. ABDULLAH\* & SAHRIM HJ. AHMAD" Structural and Magnetic Properties of Type-M Barium Ferrite – Thermoplastic Natural Rubber Nano composites" Sains Malaysiana 41(9)(2012): 1125–1131.
- [8] Nibruchnnay, Umberto "Microwave ferrite and application" Electro. Mater., Chap. 17, P. 451,459,460, (1994) .
- [9] S. Yang An, S. Won Lee, In-Bo Shim, C. Sung Kim "Magnetic properties of water-based sol-gel derived  $\text{BaFe}_{12}\text{O}_{19}/\text{SiO}_2/\text{Si}(100)$  thin films" IEEE Trans. On Magn., Vol. 37, No. 4, (July 2001).
- [10] A.J. Baden Fuller "Ferrites of microwave frequencies" London ,United kingdom, P. 2.11,14-16, (1987).
- [11] M.B. Amin & J.R. Jams "Tech. for utilization of hexagonal in radar absorbers" part 1 , Elec. Eng., Vol. 51, No. 5, P. 209-218, (1981).
- [12] S. Sugimoto, K. Okayama, S. Kando. H. Kimura, Y. Yoshida, H. Nakamura, O. Beek. T. Kagotani, M. Homma "Barium M-type ferrite as an electromagnetic microwave absorber in the GHz range" Mater. Trans. JIM, Vol. 39, No. 10, P. 1080-1083. (1998).
- [13] R. Nowosielski a, R. Babilas a,\* , G. Dercz b, L. Paj<sup>1</sup>k b, J. Wrona c " Structure and properties of barium ferrite powders prepared by milling and annealing" International Scientific Journal Volume 28, Issue 12, December 2007, Pages 735-742.
- [14] G. Mendoza-Sua´rez a,\* , L.P. Rivas-Va´zquez a, A.F. Fuentes a, J.I. Escalante-Garc´ia a, O.E. Ayala-Valenzuela b, E. Valde´z c" Preparation and magnetic properties of Zn–Ti substituted Ba-ferrite powders" Materials Letters 57December (2002) 868 – 872.