Synthesis Ni-Bi-Al Nanoferrite by sol-gel Method To investigate structural properties using a Signal Processing

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Abstract

Bismuth (Bi^{3+}) substituted in Ni- Al ferrite was synthesized as $NiBi_xAl_yFe_{2-(x+y)}O_4$ (where x= 0, 0.025, 0.050, 0.075, 0.1, 0.15) and (y=0.1, 0.2, 0.3, 0.4, 0.5) Nanoparticles by using the simple process to bring the greatest advantages as cost-effective technique, effectiveness which involving sol-gel auto combustion at low temperature. The sol-gel process is the high purity and uniform nanostructure formation.

Signal processing is carried out using Wavelet thresholding method of a noisy X-ray diffraction data In the present work the wavelet coefficients have been thresholded in order to eliminate their noisy part and that leads to study the structural properties of Bi^{3+} doped Nickel - Aluminium ferrite the synthesized samples. The results shows Wavelet thresholding methods show a very promising improvement in the de-noising of XRD data and the particle size estimated confirms spinel nanoferrite structure. Bi^{3+} substitution it shows that the particle size first increases with increasing B^{3+} and after in goes to decreasing all the samples were prepared under identical conditions.

Keywords: Signal processing, Wavelet thresholding, Ni-Bi-Al nanoferrite; XRD; .

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1. Introduction

The structural and particle size were obtain using X-ray diffraction (XRD) is a technique and atomic spacing. If the XRD data is noisy and It is very difficult to study structural properties of the materials. Then it is very necessary to remove the noise from the data.

The task of noise removal is generally concerned to as signal denoising or simply denoising. In the literature, there are many perspectives for the process of denoising. Denoising in the original signal domain and in the transform domain these are the two main categories. (Cohon, 2012).

The wavelet transforms unlike Fourier transform decomposes a time series into time and frequency domain simultaneously. Wavelets have been discovered to be a substantial way removing noise from several signals. They enable to examine the noise level independently at each wavelet scale and transform the denoising algorithm accordingly. In wavelet thresholding method the wavelet coefficients are thresholded in order to filter their noisy part (Donoho, 1995).

Effect of sintering temperature were investigated the various properties of Ni-Cafe2O4 spenal nanoferrite (H.F.Abosheiasha et al 2014). The structure of Ni-Cu nanoferrite particles were studied by synthesis solution combustion method (Composites B.J. et al 2014). The effect of Sm^{3+} substitution in Ni-Co spenal ferrite were synthesized by sol-gel method (M.K. Kokarea et al 2019). Prepared bismuth nano ferrite using sol-gel Method (A. Bismibanu et al 2018). Bismuth doped cobalt nanoferrites prepared by sol-gel combustion method were studied its structural properties (Naraavula Suresh Kumar and Katrapally Vijaya Kumar 2015)

Crystalline phases or Structural studies of Pb substituted $CoFe_2O_4$ were obtain by XRD pattern. The pattern was recorded using Cu-Kα radiation (λ = 1.54182 Å) in the 2θ range 20°-80° with step size 0.01°and time/step 2s. Using Debye-Scherrer's formula to calculate the particle size 't';

$$t = \frac{0.9\lambda}{\beta COS\theta}$$

'λ' is wavelength of the Cu-Kα radiation, β is the fullwidth at half maximum and θ is the Bragg's angle.

2. Method of Synthesis

In the present work, the samples was prepared by using Sol-gel method. this method is oxidizing metal salts and combustion agent (fuel) are essential for the combustion process. Metal nitrates and citric acid were used as oxidizing salts and combustion fuel for all the sample preparations. All chemicals were of high purity Analytical reagent and used without further purification. The Sol-gel auto combustion technique has been proved to be extremely facile, time-saving and energy-efficient route for the synthesis of ultrafine hexaferrite powders.

2.1 Samples preparation:

2.1.1 Materials:

Raw material are used in experiments are analytical grade with 99% purity in the form of nitrate i.e. Nickel nitrate ($Ni(NO_3)_2.6H_2O$), Aluminium nitrate ($Al(NO_3)_3.9H_2O$), Bismuth nitrate ($Bi(No_3)_3.5H_2O$),ferric nitrate ($Fe(NO_3)_3.9H_2O$) and citric acid ($C_6H_8O_7$) is used as fuel in the ratio 1:3 provided by Merck with ~99 % purity were used as a starting materials without further purification for the synthesis.

2.2.2 Chemical reaction**:**

$Ni(NO_3)_2).6H_2O+Al(NO_3)_3.9H_2O+Bi(No_3)_3.5H_2O+Fe(NO_3)_3.9H_2O+3C_6H_8O_7 \longrightarrow NiBiAlFe_2O_4$

100^0C for 4hrs

2.2.3 Preparation:

Using route chemical method the appropriate amount of raw materials by stoichometric ratio such as Nickel nitrate $Ni(No_3)_2.6H_2O$,Aluminium nitrate ($Al(NO_3)_3.9H_2O$), Bismuth nitrate($Bi(No_3)_3.5H_2O$) and ferric nitrate $Fe(NO_3)_3.9H_2O$ were dissolved in 100ml distilled water under magnetic stirring. Then citric acid ($C_6H_8O_7$) was mixed in the metal nitrate solution to chalet Ni^{2+}, Al^{3+}, $Bi^{3+}$and Fe^{3+} ions in the solution. The molar ratio of citric acid to total moles of nitrates was maintained at 1:3. A small amount of ammonia was added drop-wise into the solution to adjust pH value to about 7 and stabilize the nitrate-citrate solution. The solution was evaporated by intensive stirring and heating for 1 hour at 100˚C and kept at this temperature until the sol turned into a gel. The gel was then heated at 150˚C for auto-combustion to take place. The resulting powder is crushed in an agate mortar to obtain the nano ferrite particles and was sintered at 400˚C for 4 hours. Heat treatments was carried out to promote crystallization.

Bi^{3+}substituted $NiAl_yFe_2O_4$ ferrite with a chemical formula $NiBi_xAl_yFe_{2-x+y}O_4$(where, x =0, 0.025, 0.050, 0.075, 0.1, 0.15) and (y=0.1, 0.2, 0.3, 0.4, 0.5) have been synthesized via sol-gel method.

3. Characterization Technique

X-ray diffraction (XRD) is a technique which is used to find the structure of crystalline material and atomic spacing. The material should be fine powdered and homogenized. For determination of crystallinity, the XRD is the fastest and easy process. X-rays are the electromagnetic radiations and wavelengths from few angstroms to 0.1 angstroms are used for diffraction. X-ray diffraction is based on constructive interference of crystalline material and monochromatic x-rays. The electrons are produced by heating a filament in cathode ray tube (CRT) to produce the X-rays. Wavelength of x-rays should be comparable to size of atoms to find the structure. Powder diffraction is widely used in x-ray diffraction. The sample for characterization with powder diffraction method is in powder form. These x-rays are filtered to get monochromatic radiation and then directed towards the sample. To bombard target material with electrons, accelerate the electrons towards the target by applying voltage. X-ray spectra are produced when electrons have sufficient energy to eject electrons from inner shell of the target material. X-rays are used to measure size, shape of the sample. It is used to find the orientation of the sample. To find the crystallite size Debye-scherrer method is used.

X-ray diffraction (XRD) reveal that the confirmed the phase and purity of the synthesized $NiBi_xAl_yFe_{2-x+y}O_4$ samples ferrite nanoparticle is spinel cubic crystal structure and the hkl planes are (111 , 220, 311, 400, 422, 511, 440). The lattice parameter "a" calculated by using formula

$$a = d_{hkl}\sqrt{h^2 + k^2 + l^2}$$

The average crystalline size of all the synthesized samples were calculated by using Debye Scherer's formula

$$t = \frac{0.9\lambda}{\beta COS\theta}$$

Where 't' average crystalline size, λ – is the wavelength of incident X-rays, θ is the diffraction angle and β is full width at half maximum (FWHM).

4. Signal processing

In the present study, signal processing is carried out using Wavelet thresholding method. Figure 1 shows the Noisy signal (Blue line) and its denoised version (Brown line).

Wavelet Thresholding is very easy and non-linear method, which runs on one wavelet coefficient at a time. Each coefficient is thresholded by compare against threshold, if the coefficient is smaller than threshold, set to zero; otherwise it is kept or modified (Cohon, 2012). Replacing the all small noisy coefficients by zero and inverse wavelet transform on the result may assist to manage with the absolutely necessary signal characteristics and with less noise (Hedaoo and Godbole, 2011). The default mother wavelet is used in this study. Further details and demonstrations of the process are available in R software manual (Venables et al, 2019) Wavelet Comp 1.1: A guided tour through the R Package (Angi Rosch and Harald Schmidbauer, 2018) and Signal Denoising Using Wavelets (Cohon, 2012).

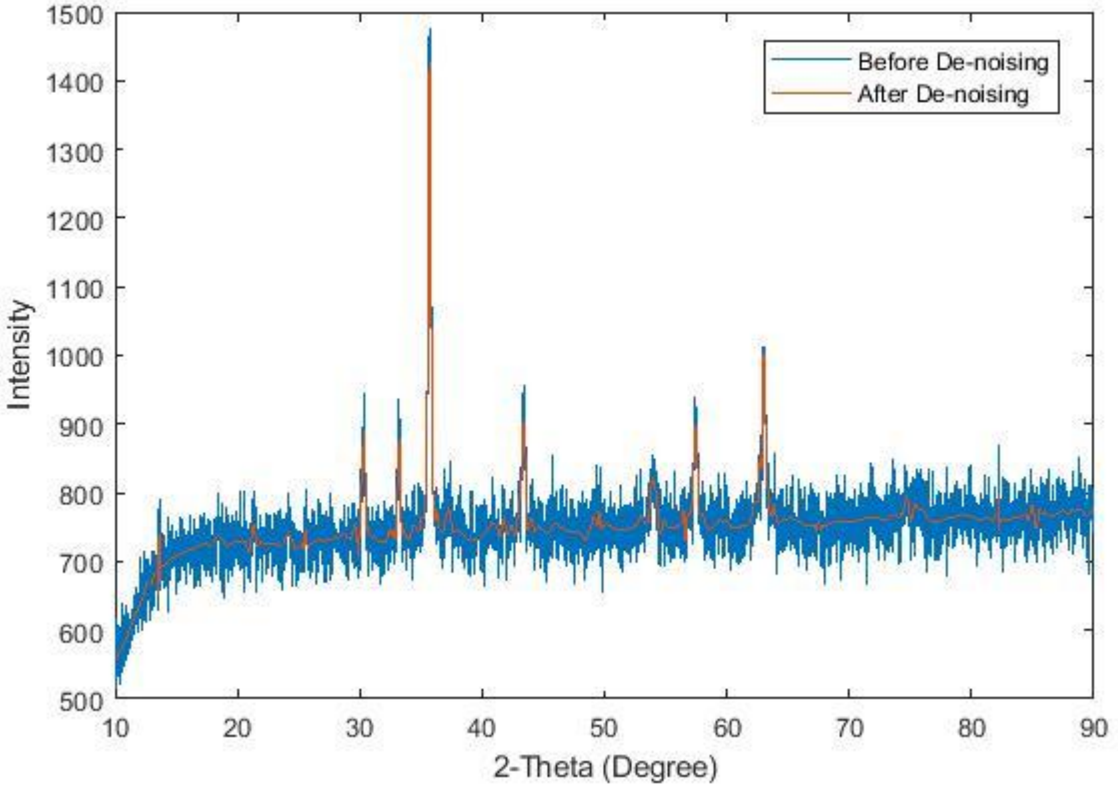

Figure 1. Noisy signal (Blue line) and its denoised version (Brown line)

5. Results

Figure 2 shows the XRD patterns for the Bi and Al samples with the Bi concentration of(x = 0,0.025, 0.050, 0.075, 0.100, 0.150) and Al concentration (y = 0, 0.1, 0.2, 0.3, 0.4, 0.5. It is clearly seen from the figure, the XRD data is noisy and it is very difficult to study structural properties of the materials using such a data.

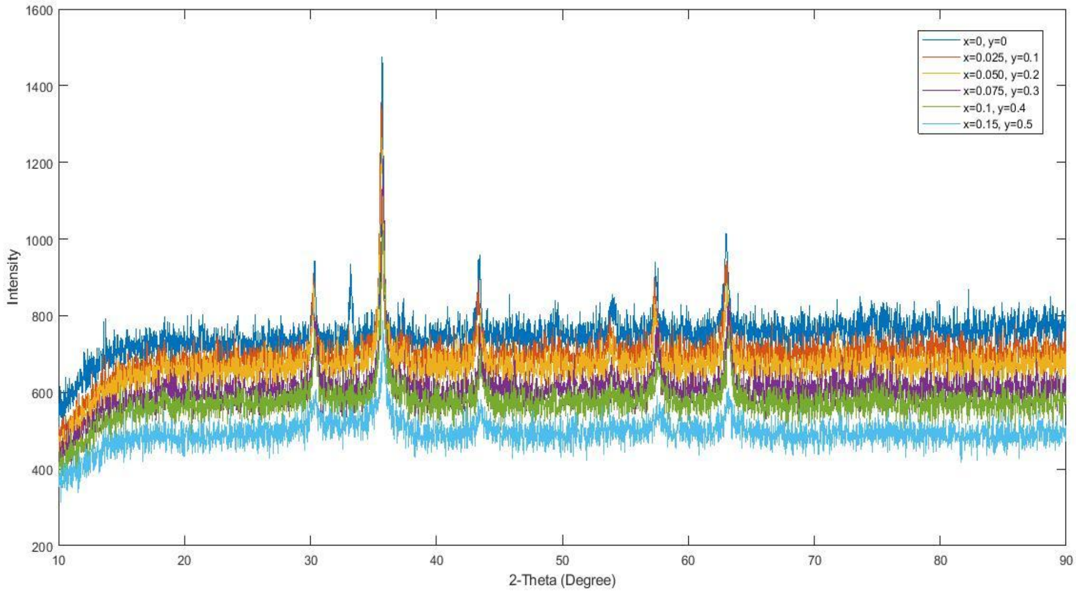

Figure 2: Stacked XRD patterns (Noisy data) for the Bi substituted $NiAl_yFe_2O_4$ samples with the Bi concentration of(x =0, 0.025, 0.050, 0.075, 0.100, 0.150) and (y = 0, 0.1, 0.2, 0.3, 0.4, 0.5).

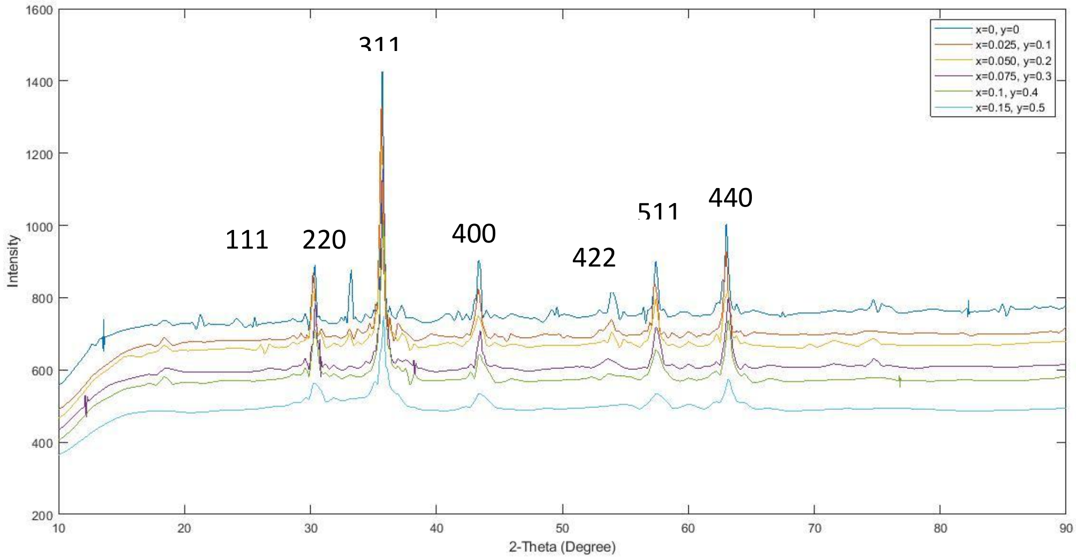

Figure 3: Stacked XRD patterns (De-noisy data) for the Bi substituted $NiAl_yFe_2O_4$ samples with the Bi concentration of(x =0, 0.025, 0.050, 0.075, 0.100, 0.150) and (y = 0, 0.1, 0.2, 0.3, 0.4, 0.5).

Figure 3 shows XRD patterns (De-noisy data) after noise removal of the data using Wavelet thresholding methods. It is cleary seen from Figure 3 the XRD patterns (De-noisy data) now in such a condition that can be studied. The average crystallite size (t) was calculated using the Debye-Scherrer'sformula. The average crystallite size was found to be in the range of 12-23nm.

Table 1 Values of Particle size of $NiBi_xAl_yFe_{2-x+y}O_4$

Sr. No.	**Composition**	**Average Grain Size (t) nm**	**Lattice Constant (a) Å**
1	$NiBi_0Al_0Fe_2O_4$	15.2364	8.3393
2	$NiBi_{0.025}Al_{0.1}Fe_{1.875}O_4$	15.8833	8.3527
3	$NiBi_{0.050}Al_{0.2}Fe_{1.75}O_4$	23.2033	8.3346
4	$NiBi_{0.075}Al_{0.3}Fe_{1.625}O_4$	15.2197	8.3256
5	$NiBi_{0.1}Al_{0.4}Fe1.5O_4$	12.6701	8.3210
6	$NiBi_{0.125}Al_{0.5}Fe_{1.375}O_4$	16.5668	8.3210

The particle size of all the samples has been calculated and is listed in Table 1. The particle size estimated shows decrease it's nature with increase in Bi^{3+} content though all the samples were prepared under identical conditions. Bismuth is ferromagnetic and ferroelectric materials it can use for microelectronic devises applications.

6. Conclusions

B^{3+} Bismuth substituted Nickel-Aluminium ferrite nanoparticles using chemical formula $NiBi_xAl_yFe_{2-x+y}O_4$ with (x = 0, 0.025, 0.050, 0.075, 0.100,0.150 and y = 0, 0.1, 0.2, 0.3, 0.4, 0.5) were prepared via sol-gel auto combustion route successfully. Wavelet thresholding methods shows a very promising improvement in the de-noising of XRD data.

The X-ray diffraction analysis reveal that confirm the formation of single phase structure nanoferrite and the hkl planes are (111 , 220, 311, 400, 422, 511, 440). It has been found that the average crystallite size is in the range of 12-23 nm and lattice constant lies between 8.32 to 8.35 A. The particle size estimated shows that first increases its nature with increase in Bi^{3+} content and afterthat it goes to decrease all the samples were prepared under identical conditions.

References

1. David L. Donoho. De-noising by soft-thresholding. IEEE Transactions on Information Theory, 41(3):613-627, (1995)
2. K. Raj, R. Moskowitz, R. Casciari, "Advance in ferrofluid technology" Journal of Magnetism and Magnetic Material 149 (1995) 177
3. X. Cao, L. Gu "Spindly cobalt ferrite nanocrystals: prepareation, characterization and magnetic properties", Nanotechnology 16 pp 180-185, (2005).
4. R. Cohen, "Signal Denoising Using Wavelets," 2012.
5. Angi Rosch and Harald Schmidbauer "Wavelet Comp 1.1: A guided tour through the R Package," 2018.
6. W. N. Venables, D. M. Smith and the R Core Team "An Introduction to R : A Programming Environment for Data Analysis and GraphicsVersion 3.6.1" 2019.
7. H.F.Abosheiasha S.T.Assar Effects of sintering process on the structural,magnetic and thermal properties of Ni0.92Ca0.08Fe2O4 nanoferrite, Journal of Magnetism and Magnetic Materials 370(2014) 54–61.
8. Composites B.J. Madhua,∗, S.T. Ashwinia, B. Shruthib, B.S. Divyashreea, A. Manjunatha, H.S. Jayanna, Structural, dielectric and electromagnetic shielding properties of Ni–Cu nanoferrite/PVP composites, Materials Science and Engineering B 186 (2014) 1–6.

9. M.K. Kokarea, Nitin A. Jadhavb, Vijay Singhc, S.M. Rathod, Effect of Sm3+ substitution on the structural and magnetic properties of Ni-Co nanoferrites, Optics and Laser Technology 112 (2019) 107–116.

10. Hedaoo, Pankaj, and Swati S. Godbole. "Wavelet thresholding approach for image denoising." International Journal of Network Security & Its Applications (IJNSA) 3.4 (2011): 16-21

11. .A. Bismibanu, M. Alagar, J.S. Mercy Jebaselvi, C. Gayathri," Preparation and Characterization of Bismuth Ferrite Nanoparticle Using Sol–Gel Meth" International Journal for Research in Applied Science & Engineering Technology (IJRASET**),** Volume 6 Issue IV, April 2018, **1767 – 1770.**

12. Naraavula Suresh Kumar, Katrapally Vijaya Kumar, "Synthesis and Structural Properties of Bismuth Doped Cobalt Nanoferrites Prepared by Sol-Gel Combustion Method" , World Journal of Nano Science and Engineering, 2015, 5, 140-151