A facile approach for obtaining NiFe2O4/C nanocomposites and their magnetic properties assessment

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Abstract
NiFe$_2$O$_4$/C nanocomposites were prepared using the two-step process, including reverse microemulsion followed by thermochemical vapour deposition (TCVD). Micelles made in the first step worked well and NiFe$_2$O$_4$ nanoparticles owing sizes smaller than 10 nanometer with narrow size distribution were obtained. Acetylene gas was used as a carbon source for deposition on the surface of NiFe$_2$O$_4$ nanoparticles. Coating applied for 1 and 2 h at 700°C and resulted in an enhancement of NiFe$_2$O$_4$ nanoparticle size to a level of 30 nanometer. Powder X-ray diffraction (PXRD) patterns revealed the spie formation for all samples; however, the crystallinity of them has been increased as much as temperature rose to a level of coating. Fourier transform infrared spectroscopy (FT-IR) verified the metal-oxygen bonding linked to the octahedral and tetrahedral vibrations. Micro-Raman spectra for the coated samples were recorded as well, and the results confirmed the existence of ordered and disordered carbon-based materials in nanocomposites. It is noteworthy to mention that, in the sample with 2 h coating time, the defect densities were lower than the sample with an hour coating time. The vibration sample magnetometer (VSM) was also used to investigate the magnetic properties. The as-prepared sample behaves like a superparamagnetic matter; however, the coated ones behave like ferromagnetic materials. The saturation magnetization and coercivity of coated samples were lower than their bulk counterparts which was due to their smaller size.

1. Introduction
A mixture of magnetic and dielectric compounds is currently a hot research topic due to its practical application ranging from catalysts [1], sensors [2], wave absorbers [3], lithium-ion batteries [4] and especially biomedicine [5]. As a magnetic composition, spinels are attractive compounds including binary and ternary spinel ferrites. Ni$^{2+}$ and Fe$^{3+}$ are considered as two most essential cations to be placed in spinel unit cell to form that. Nominal chemical composition is NiFe$_2$O$_4$ in which Ni and Fe cations supposed to occupy 8 octahedral and 16 tetrahedral sites [6, 7]. Their distribution between the octahedral and tetrahedral sites can affect the magnetic properties, and the way how they distribute between the octahedral and tetrahedral sites can significantly affect the performance of them. Chemical and thermal stability of these compounds is also another advantage of them to be
considered among researchers [8-10].

On the other hand, carbon-based compounds well-thought-out as well for their non-magnetic highly dielectric materials. Graphite, graphene, carbon nanotubes, and etc. are the well-known derivatives of carbon-based structures [11, 12].

A complex of these two compounds can make them particular for unique application, as mentioned above. However, the quality of this composite is significantly affected by the synthesis and production approach. NiFe$_2$O$_4$ nanoparticles were variously synthesized through hydrothermal method [2], sol-gel combustion [13], solid-state reaction [14], and etc.; nevertheless, micromulsion method because of providing nano-reactors can enhance the phase purity, nano-scale particle size, and more importantly narrow size distribution [15, 16]. Additionally, making a NiFe$_2$O$_4$/C composite has been widely done through arc discharge [17], solvothermal [18], magnetron and ion-beam co-sputtering [19], and spraying methods [20]. Assessment of the reports was shown that there are, however, some drawbacks including the incomplete coating of carbon, poor efficiency in the formation of graphite, and incapability in controlling the particle size.

In the current study, reverse microemulsion and thermo chemical vapor deposition (TCVD) as the two-step process was employed for composite preparation. In this way, the size of magnetic nanoparticles will be controlled and also a complete coating owing high quality will be made. Although there are enough number of reports on NiFe$_2$O$_4$/C nanocomposites, no work on NiFe$_2$O$_4$@C core–shell nanoparticles, to our knowledge, has been reported. Overall, in this work, not only new core–shell nanoparticles were synthesized but also two techniques to achieve ultra-small particles with a narrow size distribution were considered.

2. Experimental Procedure
Iron (III) chloride hexahydrate (FeCl$_3$·6H$_2$O (+%99)) and Nickel (II) chloride hexahydrate (NiCl$_2$·6H$_2$O (+%99)) were purchased from MP Biomedical. Isooctane, 1-butanol, sodium hydroxide (NaOH (%99)), and cetyl trimethyl ammonium bromide (CTAB) purchased from MERCK chemicals, and were used without further purification. These precursors were used according to the NiFe$_2$O$_4$ chemical formula
and basis of the quaternary phase diagram of water/CTAB,1-butanol/isoctane applying reverse microemulsion method and its particular circumstances, as described elsewhere [21]. 2.7 was kept as the NaOH to metal salts ratio. There were two solutions which one of them was added to another one drop by drop. During addition, pH changed and the color of the solution has changed as well. The final solution has been stirred for an hour and the pH level was preserved at 11. Finally, black precipitates of NiFe$_2$O$_4$ after this step.

The reaction is described as below:

$$\text{FeCl}_3.6\text{H}_2\text{O} + 3\text{NaOH} = \text{Fe(OH)}_3 + 3\text{NaCl} + 6\text{H}_2\text{O} \quad (1)$$

$$\text{NiCl}_2.6\text{H}_2\text{O} + 2\text{NaOH} = \text{Ni(OH)}_2 + 2\text{NaCl} + 6\text{H}_2\text{O} \quad (2)$$

$$4\text{Fe(OH)}_2 + \text{O}_2 + 2\text{H}_2\text{O} = 4\text{Fe(OH)}_3 \quad (3)$$

$$\text{Ni(OH)}_2 = \text{NiO} + \text{H}_2\text{O} \quad (4)$$

$$2\text{Fe(OH)}_3 = \text{Fe}_2\text{O}_3 + 3\text{H}_2\text{O} \quad (5)$$

$$\text{NiO} + \text{Fe}_2\text{O}_3 = \text{NiFe}_2\text{O}_4 \quad (6)$$

The as-prepared precipitates were washed several times with ethanol and distilled water several times and then were dried in an oven for 24 h at 90°C and tagged as A1. A1 sample was heat-treated at 550°C for 30 minutes and then will be referred as A2 in the next sections. A2 sample was placed in an alumina boat and transferred into the tube furnace. NiFe$_2$O$_4$ nanoparticles were coated via TCVD process with purging Ar gas up to 700°C, and then acetylene gas was purged into the tube for 1 and 2 h separately to make B1 and B2 samples. Ar and acetylene gases were purged respectively with 200 and 20 sccm flow rate. The system was cooled down to reach room temperature under Ar flow. XRD patterns were collected at ambient temperature in the 10–90° range on a PANalytical X’Pert Cu-Kα1 powder X-ray diffractometer (PXRD). The samples were ground and deposited on a silicon zero background holder. TG-DSC curves were recorded from room temperature till 900 °C with a 10 °C/min heating rate under air on a STA-1500 Rheometric Scientific instrument. Fourier transform infrared spectroscopy (FT-IR) of the sample was recorded using the FTLA ABB 2000 from 400 to 4000 cm$^{-1}$.
Transmission electron microscopy (TEM), a Zeiss EM10C instrument with an acceleration voltage of 80 kV and high-resolution transmission electron microscopy (HRTEM) (JEOL JEM-A 2100 operating at 200 KV), Energy-dispersive X-ray spectroscopy (EDS) attached to Phillips ESEM-XL30 were used to obtain images from the nanoparticles and nanocomposites. Vibration sample magnetometer (VSM) at 25 °C has been done via Meghnatesh Daghigh Kavir instrument and the micro-Raman spectroscopy has been done by Bruker-Senterra (2009) with laser wavenumber equal to 785 nm.

3. Results And Discussion

3.1. Structural and microstructural analysis

FTIR spectrum of A1 was obtained to confirm the spinel formation. There are two interstitial sites in spinel structure named as octahedral and tetrahedral which the number of them is respectively 8 and 16. Two signs of oct. and tetr. in figure 1 are presenting the metal-oxide bonding in oct. and tetr. positions. Metal-oxide bonding is generally in the range of 400 to 700 cm\(^{-1}\). In this study, octahedral and tetrahedral vibrations have been respectively detected at around 471 cm\(^{-1}\) and 677 cm\(^{-1}\). The difference in frequencies of the octahedral and tetrahedral is originated from the bond length of metal-oxide in the mentioned positions [22,23]. The rest of the peaks are originating from the precursors.

Changes in weight of the as-prepared sample was found out via the STA analysis. It also supports information about the temperatures at which each reaction and/or transformation are going to occur. Figure 4 demonstrates the STA analysis for A1 in which various weight loss is happening.

As it is evident in figure 4, multiple weight loss has been observed corresponding to sharp peaks on DSC curve. The first one was an endothermic peak in the range of 50 to 120°C which was due to water removal. The next phenomenon was around 210 to 300°C originating from organic compounds burning as a sharp exothermic peak on DSC curve. A peak around 400°C is related to the burning of the rest of the organic compounds. Besides, there is a small peak at around 590°C attributed to the crystallization completion of NiFe\(_2\)O\(_4\) from the residual Fe\(_2\)O\(_3\) and NiO [24]. There might be some carbonates absorbed on the surface of nanoparticles which were removed at approximately 780°C.
Figure 1 shows the XRD patterns of A1 and A2. As it can be seen, the ferrite peaks are reinforced after heat treatment. On the other hand, it helped to phase detection has been accomplished more appropriately.

NiFe$_2$O$_4$ with the $Fd\overline{3}m$ space group and ICDS number: 00-003-0875 [25] has been detected. Moreover, the sharpest peak is known to be (3 1 1) and located at $2\Theta = 35.7^\circ$ which was considered as the preferential plane in spinel unit cell and applied for crystallite size calculation.

Figure 2-a displays the FE-SEM image from the A1 illustrates obtaining spherical particles with some more impurities which were originated from the other precursors consisting CTAB. EDS analysis was done to find out about the deviation of chemical composition from the nominal formula. The weight percentage of each element are brought in the inset table in figure 2-b. Calculation respect to the atomic weight of Ni and Fe was shown that the Ni:Fe was approximately 0.45 based on the EDS analysis which was known as a rough approach.

As the EDS analysis was done on as-prepared sample (A1); hence, the high amount of carbon seems to be logical. It is, actually, originated from the CTAB.

After coating nanoparticles with carbon-contained compound, XRD patterns were recorded again. The difference among these patterns and the first group is that spinel related peaks are much sharper which is attributed to an enhancement of the crystallinity of spinel phase. Another evidence which must be noted is the presence of graphite in sample. It is, indeed, originated from the carbon shell created around the NiFe$_2$O$_4$ nanoparticles. The sharpest peak for NiFe$_2$O$_4$ and graphite was found out respectively at $2\Theta = 35.38^\circ$ and $25.98^\circ$ corresponding to (3 1 1) and (0 0 2) planes. Changes of the diffraction angle for NiFe$_2$O$_4$ originates from this fact that the placement of Fe$^{3+}$ and Ni$^{2+}$ cations in spinel structure has been modified after coating at higher temperatures.

Scherrer’s equation [26] was employed to find out the crystallite size of the synthesized nanoparticles.

$$D = \frac{0.9\lambda}{\beta \cos\theta}$$  \hspace{1cm} (1)
In which $D$, $\lambda$, $\beta$, and $\theta$ are respectively crystallite size, wavelength of X-ray (1.5406 Å), full width at half maximum, and the diffraction angle. The calculation was done based on the (3 1 1) plane in XRD pattern of NiFe$_2$O$_4$. Furthermore, the lattice parameter was calculated according to

$$a = d\sqrt{h^2 + k^2 + l^2},$$

in which $a$, $d$, and $(h k l)$ are respectively lattice parameter, space between plane, and the plane specification.

Figure 6 shows the TEM images of A1 and A2. Nanoparticles size for A1 is smaller than 10 nm; however, for A2 this value is higher than 10 nm. It is noticeable to mention that there might be some particle smaller or bigger than the specified values. Totally, microemulsion method is well-known as a procedure in which particle size distribution is narrow.

As it can be seen in figure 5-b, heat treatment caused changes in morphologies and particle size. Since there are some organic compounds before heat treatment, burning and removal of those compounds, as confirmed during STA analysis, can change to somehow the shape of nanoparticles. Moreover, joining smaller particles together and making bigger ones can be provided as a size enhancement. Another fact which must be considered is the effect of temperature on the preferential direction for the growth of nanoparticles and heterogeneous incorporation of nanoparticles together. Actually, heat treatment strengthens the Ostwald ripening [27].

Micro-Raman spectroscopy was used to study the quality of prepared carbon on the surface of NiFe$_2$O$_4$ nanoparticles. Reports on carbon compound offered two prominent peaks in the Raman spectra of carbon compounds [28]. These two are called G and D bands are respectively known for ordered and disordered carbon and are assigned in figure 7. They are apparent in both B1 and B2 samples. As important parameters which must be considered are the intensities of G and D bands and their ration as well. The $I_D/I_G$ ratio for both samples are brought on top of each spectrum in figure 7. There are some peaks recognized which were attributed to the spinel [29] and are in the range of 400–700 cm$^{-1}$.

The $I_D/I_G$ ratio was decreased as the coating time was increased from an hour to two hours. It means that by increasing time, the order of deposited carbon on nanoparticles was enhanced. The G-band in
The micro-Raman spectrum originates from the sp\(^2\) vibration of carbon atoms with E\(_{2g}\) symmetry within the planes. This is while D-band represents the disordered band of sp\(^3\) hybrid [30,31]. As it is presented, the I\(_D\)/I\(_G\) ration shows the ordering. The lower the I\(_D\)/I\(_G\) is, the order of created carbonic structure is higher [32]. Another important note is the FWHM of each spectrum. The reduction in FWHM signifies the formation of graphite and graphite rings in the crystal structure of carbon. This also was obvious in XRD patterns for graphite peak. In this section, in addition, the FWHM of B2 is smaller than B1, showing the effect of time on the quality of graphite. Indeed, Acetylene gas had enough time to decompose, regulated, and finally deposited on the surface of NiFe\(_2\)O\(_4\) nanoparticles. Carbon-carbon bond angle is also affecting the narrowing of D- and G-bands [33]. It is found out that the bond angles of carbon atoms in the amorphous structure have a lot of distortion. With an enhancement in crystallinity, opening of bond angles occurs at an angle linkage, so that the bond angle in sp\(^2\) approaches to its steady-state value of 120°. The defect concentration can be understood from the I\(_D\)/I\(_G\) as well. As this ration decreases, the defect concentration decreases and hexagonal rings will be enhanced. It means that the graphite-like structure owing the sp\(^2\) increases and ordering will be obtained.

TEM and HRTEM instruments were employed to see the prepared nanocomposites and the obtained images are illustrated figure 8.

The carbon shell encompassed the NiFe\(_2\)O\(_4\) nanoparticles. B1 image displays a mixture of shell and separate carbon all around the NiFe\(_2\)O\(_4\) nanoparticles; however, conditions are different for B2 and the well-coated carbon was observed around the NiFe\(_2\)O\(_4\) nanoparticles. The atomic layers of this ordered carbon around the nanoparticles is visible in figure8-c screening and verifying the results of the micro-Raman study.

The small size of NiFe\(_2\)O\(_4\) approved the applicability of nano-reactors formed in microemulsion method retarded the growth of nanoparticles during the process.

### 3.2. Magnetic properties
To consider the magnetic properties such as $M_s$ and $H_c$ for all four samples, a field equal to 1T has been separately applied on them. Figure 9 demonstrates the B-H hysteresis loops for A1 and A2. They behaved like a superparamagnetic matter in which there is no saturation; however, $M_r$ and $H_c$ are not zero [13]. It originates from this fact that the time for magnetic measurement is higher than the relaxation time for the dipole. Actually, it is the ambient temperature that can affect the dipole to turn from an easy direction to another one. Furthermore, it shows that both samples have nanoparticles owing single domains. It is noteworthy to mention that according to the inset image in figure 9, $H_c$ for A2 is higher than A1 which is related to the bigger crystallites for A2.

Magnetic assessment of carbon compounds revealed that they behave like a diamagnetic [34]. Therefore it is supposed to affect the magnetic properties of nanocomposites. Due to the existence of disordered carbon in samples, there is a high density of defects in their structures; hence their resistivity will increase and the diamagnetism characteristic of it will be weaker than the bulk carbon. Therefore, the effect of carbon shell on magnetic properties of NiFe$_2$O$_4$ will be negligible.

Figure 10 demonstrates the hysteresis loops for B1 and B2. It is found out that the saturation for this samples is smaller than the bulk samples counterparts. The smaller size of the current samples is the reason for this. Indeed, in samples with lower crystallites or particles, the ration of surface to volume increases than the bulk samples and the preferential sites for cations will change. It means that the antisite defects degree enhances and then the magnetization declines [35].

As the B2 was kept an hour more than B1 at 700°C, it is logical to have higher coercivity due to bigger crystallites. As it is provided in table 4, $M_s$ value for B1 and B2 are respectively 46.52 and 27.62 emu/g. additionally, the coercivity for these two samples was recorded as 19.7 and 62.77 Oe, respectively.

There is defined a critical value for crystallite size till that as the crystallite size increases, the coercivity rises. Grain growth in B2 rather than B1 results in an enhancement in $H_c$. The NiFe$_2$O$_4$/C nanocomposites due to existence of diamagnetic carbon have different saturation magnetization and coercivity respect to the other results.
4. Conclusion
In this study, NiFe$_2$O$_4$/C nanocomposites were successfully prepared through reverse microemulsion and TCVD. Their structural magnetic properties have been obtained to find out their specification. TEM images were revealed that the as-prepared nanoparticles are spherical and are smaller than 10 nm which were affected then by heat treatment and changed their morphology. TEM images were also verified the reverse microemulsion approach is practical for synthesizing nanoparticles with small size and narrow size distribution. Hysteresis loops for the as-prepared and heat-treated samples were shown that they behaves like a superparamagnetic and owing a single domain of dipoles. On the other hand, coated samples due to undergoing higher temperatures perform like a ferromagnetic. HRTEM images showed that the generated carbon shell on the surface of nanoparticles is ordered; however, the rest of it is a disorder. Micro-Raman studies have also approved this suggestion, and the density of defects in B2 was lower than B1.

Declarations
Conflict of interests
There are no conflicts to declare.

References
[1] M. Bahgat, A.A. Farghali, W.M.A. El Rouby, M.H. Khedr, Efficiency, kinetics and thermodynamics of toluidine blue dye removal from aqueous solution using MWCNTs decorated with NiFe$_2$O$_4$, Fullerenes, Nanotub. Carbon Nanostructures. 22 (2014) 454–470.

[2] S. Ahmadian-Fard-Fini, D. Ghanbari, M. Salavati-Niasari, Photoluminescence carbon dot as a sensor for detecting of Pseudomonas aeruginosa bacteria: Hydrothermal synthesis of magnetic hollow NiFe$_2$O$_4$-carbon dots nanocomposite material, Compos. Part B Eng. 161 (2019) 564–577.

[3] S.A. Soomro, I.H. Gul, M.Z. Khan, H. Naseer, A.N. Khan, Dielectric properties evaluation of NiFe$_2$O$_4$/MWCNTs nanohybrid for microwave applications prepared via novel one step synthesis, Ceram. Int. 43 (2017) 4090–4095.

[4] R. Jin, H. Jiang, Y. Sun, Y. Ma, H. Li, G. Chen, Fabrication of NiFe$_2$O$_4$/C hollow spheres constructed by mesoporous nanospheres for high-performance lithium-ion batteries, Chem. Eng. J. 303 (2016) 501–510.
[5] Z. Chen, Y. Gao, D. Mu, H. Shi, D. Lou, S. Liu, Recyclable magnetic NiFe$_2$O$_4$/C yolk-shell nanospheres with excellent visible-light-Fenton degradation performance of tetracycline hydrochloride, Dalt. Trans. 48 (2019) 3038–3044.

[6] Z. Zhang, Y. Liu, G. Yao, G. Zu, Y. Hao, Synthesis and Characterization of NiFe$_2$O$_4$ Nanoparticles via Solid-State Reaction, Int. J. Appl. Ceram. Technol. 10 (2013) 142–149.

[7] H. Kavas, N. Kasapoğlu, A. Baykal, Y. Köseoğlu, Characterization of NiFe$_2$O$_4$ nanoparticles synthesized by various methods, Chem. Pap. 63 (2009) 450–455.

[8] M.R. Phadatare, A.B. Salunkhe, V.M. Khot, C.I. Sathish, D.S. Dhawale, S.H. Pawar, Thermodynamic, structural and magnetic studies of NiFe$_2$O$_4$ nanoparticles prepared by combustion method: Effect of fuel, J. Alloys Compd. 546 (2013) 314–319.

[9] L. Sun, R. Zhang, Z. Wang, L. Ju, E. Cao, Y. Zhang, Structural, dielectric and magnetic properties of NiFe$_2$O$_4$ prepared via sol-gel auto-combustion method, J. Magn. Magn. Mater. 421 (2017) 65–70.

[10] A.A. Ensafi, N. Zandi-Atashbar, M. Gorgabi-Khorzoughi, B. Rezaei, Nickel-Ferrite Oxide Decorated on Reduced Graphene Oxide, An Efficient and Selective Electrochemical Sensor for Detection of Furazolidone, IEEE Sens. J. (2019).

[11] L.L. Zhang, X.S. Zhao, Carbon-based materials as supercapacitor electrodes, Chem. Soc. Rev. 38 (2009) 2520–2531.

[12] O.C. Compton, S.T. Nguyen, Graphene oxide, highly reduced graphene oxide, and graphene: versatile building blocks for carbon-based materials, Small. 6 (2010) 711–723.

[13] A. Ahlawat, V.G. Sathe, V.R. Reddy, A. Gupta, Mossbauer, Raman and X-ray diffraction studies of superparamagnetic NiFe$_2$O$_4$ nanoparticles prepared by sol-gel auto-combustion method, J. Magn. Magn. Mater. 323 (2011) 2049–2054.

[14] Z. Zhang, Y. Liu, G. Yao, G. Zu, X. Zhang, J. Ma, Solid-state reaction synthesis of NiFe$_2$O$_4$ nanoparticles by optimizing the synthetic conditions, Phys. E Low-Dimensional Syst. Nanostructures. 45 (2012) 122–129.
[15] F. Foroughi, S.A. Hassanzadeh-Tabrizi, J. Amighian, Microemulsion synthesis and magnetic properties of hydroxyapatite-encapsulated nano CoFe$_2$O$_4$, J. Magn. Magn. Mater. 382 (2015) 182–187.

[16] P. Sivakumar, R. Ramesh, A. Ramanand, S. Ponnusamy, C. Muthamizhchelvan, Synthesis, studies and growth mechanism of ferromagnetic NiFe$_2$O$_4$ nanosheet, Appl. Surf. Sci. 258 (2012) 6648–6652.

[17] Z. Xie, D. Geng, X. Liu, S. Ma, Z. Zhang, Magnetic and microwave-absorption properties of graphite-coated (Fe, Ni) nanocapsules, J. Mater. Sci. Technol. 27 (2011) 607–614.

[18] J. Zheng, Z.Q. Liu, X.S. Zhao, M. Liu, X. Liu, W. Chu, One-step solvothermal synthesis of Fe$_3$O$_4$@C core–shell nanoparticles with tunable sizes, Nanotechnology. 23 (2012) 165601.

[19] J.J. Cuomo, J.P. Doyle, J. Bruley, J.C. Liu, Sputter deposition of dense diamond-like carbon films at low temperature, Appl. Phys. Lett. 58 (1991) 466–468.

[20] J.N. Wang, L.I. Zhang, F. Yu, Z.M. Sheng, Synthesis of carbon encapsulated magnetic nanoparticles with giant coercivity by a spray pyrolysis approach, J. Phys. Chem. B. 111 (2007) 2119–2124.

[21] C.-C. Wang, D.-H. Chen, T.-C. Huang, Synthesis of palladium nanoparticles in water-in-oil microemulsions, Colloids Surfaces A Physicochem. Eng. Asp. 189 (2001) 145–154.

[22] S. Ayyappan, J. Philip, B. Raj, A facile method to control the size and magnetic properties of CoFe$_2$O$_4$ nanoparticles, Mater. Chem. Phys. 115 (2009) 712–717.

[23] J.G.S. Duque, E.A. Souza, C.T. Meneses, L. Kubota, Magnetic properties of NiFe$_2$O$_4$ nanoparticles produced by a new chemical method, Phys. B Condens. Matter. 398 (2007) 287–290.

[24] Z.K. Karakaş, R. Boncukcuoğlu, İ.H. Karakaş, M. Ertuğrul, The effects of heat treatment on the synthesis of nickel ferrite (NiFe$_2$O$_4$) nanoparticles using the microwave assisted combustion method, J. Magn. Magn. Mater. 374 (2015) 298–306.

[25] M. Kooti, A.N. Sedeh, Synthesis and characterization of NiFe$_2$O$_4$ magnetic nanoparticles by combustion method, J. Mater. Sci. Technol. 29 (2013) 34–38.

[26] A.L. Patterson, The Scherrer formula for X-ray particle size determination, Phys. Rev. 56 (1939)
Table 1: Crystallite size and the lattice parameter for NiFe$_2$O$_4$ nanoparticles in A1, A2, B1, and B2.

| Sample No. | D (nm) | a (Å)     |
|------------|--------|-----------|
| A1         | 5      | 8.393(1)  |
| A2         | 11     | 8.364(2)  |
| B1         | 16     | 8.369(2)  |
| B2         | 18     | 8.370(3)  |

Table 2, mentioned on page 11, was omitted by the authors in this version of the paper.

Table 3: obtained magnetic parameters for A1, A2, B1, and B2 from their hysteresis loops.
| Sample No. | $H_c$(Oe) | $M_s$(emu/g) | $M_r$(emu) |
|------------|------------|--------------|------------|
| A1         | 0.6        | 0.92         | 0.009      |
| A2         | 0.64       | 0.68         | 0.007      |
| B1         | 19.7       | 46.52        | 1.61       |
| B2         | 62.77      | 27.62        | 1.36       |

Table 4, mentioned on page 11, was omitted by the authors in this version of the paper.

Figures

![FTIR spectrum of A1](image)

Figure 1

FTIR spectrum of A1
Figure 2

TG/DSC analysis of A1.
Figure 3

XRD results of A1 and A2 samples showing the spinel formation.
Figure 4

(a) FE-SEM image of A1, and (b) EDS analysis results of the specified area.
Figure 5

XRD patterns of B1 and B2.

Figure 6

TEM images of synthesized nanoparticles for (a) A1 and (b) A2
Figure 7

Micro-Raman spectra for B1 and B2 from 0 to 3500 cm$^{-1}$.
Figure 8

TEM images for (a) B1, (b) B2 and (c) HRTEM image for B2.

Figure 9

Hysteresis loops for A1 and A2 based on the magnetic measurement under 1T.
Figure 10

Hysteresis loops for B1 and B2.