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# **ARTICLE TYPE**

# Development of a Novel 'One-Pot' Synthetic Method for Preparation of (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>x</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites and Study of their Microwave Absorption and Magnetic Properties

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Development of a simple but novel aqueous solution based 'one-pot' method has been reported for the preparation of soft nanocomposites, composed of ferrite (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>) and hard ferrite (BaFe<sub>12</sub>O<sub>19</sub>) phases. A physical mixing method has also been employed to prepare nanocomposites having same compositions. The effects of synthetic methodologies on the microstructures of the nanocomposites as well as their magnetic and microwave absorption properties have been evaluated. Crystal structures and microstructures of these composites have been investigated by using X-ray diffraction, transmission electron microscope and scanning electron microscope. In the nanocomposites, prepared by both methods, presence of nanocrystalline Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> phases were detected. However, nanocomposites, prepared by onepot method, possessed better homogeneous distribution of hard and soft ferrite phases than the nanocomposites, prepared by physical mixing method. Very good spring exchange coupling interaction between the hard and soft ferrite phases was observed for the nanocomposites, prepared by one-pot method and these composites exhibited magnetically single phase behaviour. The spring exchange coupling interaction enhanced the magnetic properties (high saturation magnetization and coercivity) and microwave absorption properties of the nanocomposites, prepared by one-pot method, in comparison with the nanocomposites prepared by physical mixing method as well as pure Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> nanoparticle. The minimum reflection loss of the composites was found to be  $\sim$  -25 dB (i.e. > 99%absorption) at 8.2 GHz with absorber thickness of 3.5 mm.

#### 1. Introduction

Microwave-absorbing materials have gained immense interest to the scientists and technologists due to their usage in military application and also commodity markets. Microwave-absorbing materials could be used in stealth defence system because they can effectively reduce the radar cross-section of targets. These materials also have the potential to address the issues related to environmental pollution caused by the Electromagnetic Interference (EMI) due to extensive use of electronic devices,

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computer networks, mobile phones etc.<sup>1-7</sup> Hence reduction of interference of electromagnetic radiation, mainly microwave radiation is highly desirable in current scenario. Therefore, there has been a great deal of interest to develop microwave radiation absorbing materials. The microwave absorbing materials which operate in the frequency range 8.2- 12.4 GHz (X-band) region is also known as radar absorbing material (RAM) and find wide range of applications including electronic devices, military equipment, etc. Researchers are showing their specific concern on development of efficient RAM from last few decades. Various types of materials such as dielectric, magnetic, conducting polymer or composites have been investigated by the scientists to achieve RAMs with desired properties.<sup>8-11</sup>

Ferrites are important class of magnetic materials which exhibit microwave absorption property.<sup>12-19</sup> Spinel ferrites are used in megahertz range due to their Snoek limitation and hexaferrites exhibit microwave absorption in gigahertz range though band width is narrow.<sup>20-22</sup> However, it is difficult for a single material to fulfil the requirements (such as large absorption peak, wide working frequency range and thin absorption layer) of an ideal radar absorber. Nanocomposites consist of hard and soft ferrite phase, can offer to construct unified systems whose properties are complimentary or even mutually exclusive.

Recently, there has been immense attention on the materials having good spring exchange coupling behaviour. According to the exchange spring theory proposed by Kneller and Hawig,23 nanocomposite magnets consist of soft and hard phases, having sufficiently exchanged coupling between both the phases are known as exchange spring magnet. Exchange spring magnets possess high saturation magnetization of soft phase and high coercivity of hard phase, which leads to produce large energy product value and are potential candidates for next generation permanent magnet.24 These materials also exhibit improved microwave absorbing property.<sup>1,2,25,26</sup> According to Maeda et al.<sup>27</sup> the exchange interaction between hard and soft ferrite phases can enhance the microwave absorption properties. The exchange spring behaviours of some metal alloy systems and multilayer systems (such as Pr<sub>2</sub>Fe<sub>12</sub>B,<sup>28</sup> Nd-Fe-B,<sup>29</sup> Sm-Co/Fe,<sup>30</sup> SmCo<sub>x</sub>-Co<sup>31</sup> Ba $Co_2Fe_{16}O_{27}$  or Ba $_2Co_2Fe_{12}O_{22}$  -FeCo alloy,<sup>32</sup>  $BaFe_{12}O_{19}/\alpha$ -Fe.<sup>33</sup> etc.) have been reported by researchers. Compare to metallic systems, nanocomposites composed of soft

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spinel ferrite and hard hexagonal ferrite, have shown their potential to be a promising candidate for superior permanent magnet, because of their low cost, excellent corrosion resistance behaviour and high electrical resistivity. However, best of our knowledge reports on soft- hard ferrite nanocomposites are very limited in the literature,<sup>1,2,24,34-38</sup> due to lack of availability of simple preparation techniques. Most of the reported methods are complex in nature and require high sintering temperatures. Roy et al. have reported preparation of NiZn ferrite-barium ferrite composite, where NiZn ferrite and barium ferrite were prepared separately by coprecipitation method and high temperature solid state method (sintered at 1200 °C) respectively. NiZn ferrite and barium ferrite powders were then mixed in appropriate weight ratios and heat treated at different temperatures ranging from 400 to 800 °C.35 They have also reported the preparation of BaCa<sub>2</sub>Fe<sub>16</sub>O<sub>27</sub>- Fe<sub>3</sub>O<sub>4</sub> composites, where BaCa<sub>2</sub>Fe<sub>16</sub>O<sub>27</sub> was prepared by citrate method and commercial grade Fe<sub>3</sub>O<sub>4</sub> was procured.<sup>34</sup> Chen et al.<sup>26</sup> have employed citrate based sol-gel technique to synthesize composite powders with different weight ratios of strontium hexaferrite to zinc ferrite and studied their microwave absorption properties. Song et al.<sup>24</sup> have reported a sol-gel citrate route for preparation of SrFe<sub>12</sub>O<sub>19</sub>/Ni<sub>0.5</sub>Zn<sub>0.5</sub>Fe<sub>2</sub>O<sub>4</sub> hollow microfiber at 900 °C. Synthesis and microwave absorbing properties of nanocomposites consist of BaFe<sub>12</sub>O<sub>19</sub> (BFO)/Ni<sub>0.5</sub>Zn<sub>0.5</sub>Fe<sub>2</sub>O<sub>4</sub> (NZFO) ferrite microfibers with different mass ratios was investigated by Shen et al.<sup>1</sup> Xie et al.<sup>38</sup> have investigated Sr-Zn ferrite composites which were synthesized by chemical coprecipitation with two step sintering process at 700 °C for 6h and then 1000 °C for 2h. Tyagi et al.<sup>2</sup> have reported composite consists of strontium ferrite/Ni-Zn ferrite powders prepared by co-precipitation method and they have also studied its microwave absorption properties in X-band.

High level of homogenous mixing of the hard and soft ferrite phases is required for the fabrication of exchange spring magnet. This is possible when both the phases can be grown together from the same reaction mixture. In this paper, we are reporting a simple 'one-pot' synthetic route for the preparation of nanocomposites, containing both  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  (soft ferrite)-  $BaFe_{12}O_{19}$  (hard ferrite) phases with wide range of compositions. Nanocomposites, having same compositions, were also prepared by employing 'physical mixing' method using pure  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  and  $BaFe_{12}O_{19}$  nanopowders.

types of composites were measured and compared.

#### 2. Experimental

#### 2.1. Materials

BaCO<sub>3</sub>, Mn(NO<sub>3</sub>)<sub>2</sub>.4H<sub>2</sub>O, Ni(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O, Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O, Zn dust, Nitric Acid and Ethylene diamine tetra acetic acid (EDTA) were purchased from Merck, India and used without further purification. Zn(NO<sub>3</sub>)<sub>2</sub> and Ba(NO<sub>3</sub>)<sub>2</sub> were prepared by dissolving Zn dust and BaCO<sub>3</sub> respectively in aqueous nitric acid.

## 2.2. Synthesis of $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$ nanocomposites by one-pot method (OP Method)

To prepare  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$ nanocomposites (with x= 0.85, 0.75, 0.5 and 0.25) using one-pot method stoichiometric amount of aqueous solutions of BaNO<sub>3</sub>,  $Mn(NO_3)_2.4H_2O$ ,  $Ni(NO_3)_2.6H_2O$ ,  $Zn(NO_3)_2$  Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O, and EDTA were mixed in a beaker (Table 1) and stirred for 2 h. This reaction mixture was then dried at ~110 °C for 2 h. Black colour floppy carbonaceous material was formed after drying, which referred as precursor powder. Then the precursor powders were calcined at 800 °C for 4 h in air atmosphere to obtain pure  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$  nanocomposites.

### 2.3. Synthesis of $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites by physical mixing method (PM Method)

We have also prepared a set of composite samples with various composition (with x = 0.85, 0.75, 0.5 and 0.25) by using 'physical mixing' method where pure Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> powders were mixed with appropriate weight ratio (Table. 1) using a mortar pestle. Pure BaFe<sub>12</sub>O<sub>19</sub> and Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> nanopowders were prepared separately by using the EDTAprecursor method which has been developed by us.<sup>39,40</sup> For synthesis of BaFe<sub>12</sub>O<sub>19</sub>, stoichiometric amounts of Ba(NO<sub>3</sub>)<sub>2</sub> and Fe(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O were dissolved in distilled water according to the molar ratio of 1:12, as shown in Table 1. An aqueous solution of EDTA was prepared by dissolving EDTA in hot water with drop wise addition of NH<sub>4</sub>OH. After complete dissolution of EDTA, the solution was boiled to remove the excess NH<sub>3</sub>. The pH of the solution was ~6. Aqueous solutions of metal nitrates and EDTA were mixed in a molar ratio of 1: 4 and stirred for 1 h at room re using a magnetic stirrer.

Ν	$Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ and $BaFe_{12}O_{19}$ nanopowders.	temperature using	g a magnetic sti
	Table 1: Starting materials required for preparation of (Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fc <sub>2</sub> )	O4)x-(BaFe12O19)1-x 1	nanocomposites.

Composition	*Mn <sup>II</sup> -	*Zn dust	*Ni <sup>II</sup> -	*Fe <sup>III</sup> -	*Ba-	*EDTA	**Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub>	**BaFe <sub>12</sub> O <sub>19</sub>
	nitrate (g)	) (g)	nitrate (g)	nitrate (g)	Carbonate(g)	(g)	(g)	(g)
Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> -pure	0.212	0.112	0.497	3.419		3.71		
(Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.85</sub> -	0.116	0.06	0.269	3.846	0.08	14.31	0.55	0.45
(BaFe <sub>12</sub> O <sub>19</sub> ) <sub>0.15</sub>								
(Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.75</sub> -	0.082	0.043	0.192	3.98	0.108	14.21	0.39	0.61
(BaFe <sub>12</sub> O <sub>19</sub> ) <sub>0.25</sub>								
(Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.5</sub> -	0.037	0.019	0.086	4.18	0.146	13.91	0.18	0.82
(BaFe <sub>12</sub> O <sub>19</sub> ) <sub>0.5</sub>								
(Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.25</sub> -	0.014	0.007	0.032	4.3	0.166	13.75	0.07	0.93
(BaFe <sub>12</sub> O <sub>19</sub> ) <sub>0.75</sub>								
BaFe <sub>12</sub> O <sub>19</sub> -pure				4.365	0.177	13.74		

\*For one-pot method

1 \*\*For physical mixing method

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formed when the mixture was evaporated to dryness on a hot plate at ~110 °C. The precursor powder was then calcined in air for 4 hr at 800 °C to obtain  $BaFe_{12}O_{19}$  nanopowder.

To prepare  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  nanopowder, we have used metal nitrates, such as  $Fe(NO_3)_3.9H_2O$ ,  $Ni(NO_3)_2.6H_2O$ ,  $Mn(NO_3)_2.4H_2O$ ,  $Zn(NO_3)_2$  as starting materials and water as solvent. Stoichiometric amounts of metal nitrates were dissolved in distilled water according to the molar compositions as shown in Table 1. The aqueous solutions of metal nitrates and EDTA were mixed in a molar ratio of 1: 1 and stirred for 1 h at room temperature using a magnetic stirrer. The pH of the reaction mixture was found to be ~2. Dark brown colored precursors were formed when the reaction mixtures were evaporated to dryness on a hot plate at 125 °C. Partial decomposition of the precursors was observed during drying. The precursor powders were then calcined in air for 2 h at 525 °C to obtain Mn-Ni-Zn ferrite nanopowders.

From now onwards, nanocomposites prepared by one-pot method and physical mixing method will be referred as 'composites-OP' and 'composites-PM' respectively.

#### 2.4. Characterization

Room temperature X-ray diffraction spectra of the precursors and the calcined powders were recorded by using a powder X-Ray diffractometer (Mini Flex II, Rigaku, Japan) with Cu  $K_{\alpha}$  ( $\lambda$  = 0.15405 nm) radiation. Thermogravimetric analysis (TGA) and differential scanning calorimetric (DSC) analysis were carried out on the precursor using a DTG-60 and a DSC-60 (Shimaduzu, Japan) respectively in air flow at a heating rate of 10 °C/ min between 30 to 550 °C. Aluminium and Platinum sample pans were used for DSC and TGA respectively. Transmission Electron Microscopic (HRTEM) (JEOL JEM 1400, Japan) images of samples was employed to analyze the shape and size of the synthesized nanocomposites. Morphology of the nanocomposites, prepared by one-pot method and physical method, was studied using Scanning Electron Microscope (SEM) (JSM-6360LV, JEOL, Japan) using an accelerating voltage of 15 kV. Elemental analysis of the composites were performed using Energy Dispersive X-Ray analysis (EDX) which was attached with SEM. Room temperature magnetization with respect to external magnetic field was measured for the synthesized composites by using a vibrating sample magnetometer (EV5, ADE Technology, USA).

For measurement of microwave absorption of the synthesized nanocomposites in X-band (8.2 - 12.4 GHz range), HP 8510 vector Network Analyzer (USA) was used and reflection loss (RL) was calculated using the measured values of complex permittivity and permeability. To prepare the samples for this purpose, nanocomposite powders were first mixed with aqueous solution of 10 wt. % polyvinyl alcohol (PVA) which acted as binder and the mixture was dried. This mixture was further ground to powders and then compressed under a pressure of 10 tons and shaped into rectangular pellets with size of 10.16 mm x 22.86 mm x 2 mm, so as to fit exactly into a rectangular waveguide of X-band.

#### 3. Results and Discussion

#### 3.1. Thermal analysis

TGA and DSC analysis were used to investigate the thermal decomposition behavior of precursors, prepared by OP method. A total weight loss of ~95 % was observed when the precursor powder was heated from 30 to 550 °C in air (Fig. 1). Initially, ~4 % weight loss occurred in the region of 40 to 100 °C due to loss of moisture from the sample. Then in the temperature range of 200 to 470 °C, ~91 % weight loss was observed. This might be due to the oxidative decomposition of precursor and evolution of  $CO_2$  and  $NO_x$  gases. This decomposition was also appeared as an exothermic peak at 429 °C in DSC thermogram. No weight loss was observed in TGA when the sample was heated beyond 470 °C. This confirmed the full decomposition of carbonaceous mass of the precursor occurred within 470 °C.



Fig 1: TGA- DSC thermogram of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.5}$ - $(BaFe_{12}O_{19})_{0.5}$  precursor.

#### 3.2. XRD analysis

XRD patterns of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$ nanocomposites, prepared by one-pot method as well as physical mixing exhibited the diffraction peaks corresponding to both spinel Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub><sup>40,41</sup> and hexagonal BaFe<sub>12</sub>O<sub>19</sub> [ICDD 84-0757] phase (Fig. 2 (a) and (b)) and indicated the coexistence of both the phases in the composite powders. Any impurity peak, such as NiO, MnO, ZnO, BaO, BaCO<sub>3</sub>, α-Fe<sub>2</sub>O<sub>3</sub> etc. within the resolution of the technique was not observed. However, variation in relative intensities of the diffraction peaks was observed for these two types of composites. This might be due to the variation of crystalline size and homogeneous distribution of the spinel and hexagonal phases in the nanocomposites with the method of preparation. The average crystallite sizes of Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> phases in  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites were calculated by X-ray peak-broadening method using Scherrer's equation<sup>42</sup> and listed on Table 2. For  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ , the diffraction peak at  $2\theta = 35.6^\circ$ , which corresponds to (311) plane and for BaFe<sub>12</sub>O<sub>19</sub>, diffraction peak at  $2\theta = 34.2^{\circ}$ , i.e. (114) plane were used.

The important feature was that, in  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ -

 $(BaFe_{12}O_{19})_{1-x}$  nanocomposites-OP method, the crystallite size of Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> phase was increased from 11 to 19 nm with increasing amount of Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> phase in the composite. The same trend was observed for BaFe<sub>12</sub>O<sub>19</sub> phase and its crystallite size was found to be increased from 22 to 43 nm. (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>x</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites -PM method, the crystallite sizes of both spinel and hexagonal phase not change much with varying amount did of Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> phases in the composites and were found to be ~18 nm for Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and ~43 nm for BaFe<sub>12</sub>O<sub>19</sub>. These values were almost same in comparison with the crystallite sizes of pure Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and BaFe<sub>12</sub>O<sub>19</sub> nanopowders and indicated that the pure phases retain their individual crystallite sizes in the composites.

Nanocomposites prepared by physical mixing and one-pot method showed two distinct types of microstructures when their morphology was investigated by HRTEM (Fig. 3). In case of the nanocomposites-PM (Fig. 3(d, f, h, j), clear segregation of hexagonal BaFe<sub>12</sub>O<sub>19</sub> nanoparticles (average particle size of ~60-70 nm, Fig. 3(a)) and spherical shaped agglomerated  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  nanoparticles (~20 nm average particle size, Fig. 3(b)) were observed. On the contrary, for the nanocomposites, prepared by one-pot method, almost uniform shaped nanoparticles (average particle size ~60-70 nm) were observed (Fig. 3(c, e, g, i)). SEM micrographs of the composites also revealed the intimate coexistence of  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ and BaFe<sub>12</sub>O<sub>19</sub> particles in the composites-OP (Fig. 4 (a)) and presence of large BaFe<sub>12</sub>O<sub>19</sub> and small  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ nanoparticles in the composites-PM (Fig. 4 (b)).



**Fig 2:** (A) XRD spectra of the (a) pure  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ ,  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>0.15</sub> nanocomposites prepared by (b) one-pot, (c) physical mixing methods and (d) BaFe<sub>12</sub>O<sub>19</sub> nanopowders, (B) XRD spectra of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites synthesized by one-pot method (#Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> and \*BaFe<sub>12</sub>O<sub>19</sub>).

Table 2: Average crystalline size of (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>x</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites prepared by one-pot and physical mixing method.

	Crystallite size (nm)					
Sample	One	e-Pot	Physica	1 mixing		
	*(114)plane	#(311)plane	*(114)plane	#(311)plane		
$Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$		19				
$(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2\mathrm{O}_4)_{0.85}(BaFe_{12}\mathrm{O}_{19})_{0.15}$	22	16	43	19		
$(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2 O_4)_{0.75}(BaFe_{12} O_{19})_{0.25}$	27	14	42	17		
$(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.5}(BaFe_{12}O_{19})_{0.5}$	34	12	43	16		
$(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2\mathrm{O}_4)_{0.25}(BaFe_{12}\mathrm{O}_{19})_{0.75}$	38	11	45	18		
$BaFe_{12}O_{19}$	43					

\*For BaFe<sub>12</sub>O<sub>19</sub>

**3.3. T** #For  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$ 

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Fig 3: TEM micrographs of (a) pure  $BaFe_{12}O_{19}$ , (b) pure  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  nanopowders and  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{x}$ - $(BaFe_{12}O_{19})_{1-x}$  nanocomposites synthesis by (c, e, g, i) one-pot and (d, f, h, j) physical mixing method.

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**Fig 4:** SEM micrographs of  $(Mn_0.2Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.5}$ - $(BaFe_{12}O_{19})_{0.5}$  nanocomposites prepared by (a) one-pot, (b) physical mixing method and (c) EDX spectra of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.5}$ - $(BaFe_{12}O_{19})_{0.5}$  nanocomposite prepared by one-pot method.

Electron microscopic analysis clearly shows that, the nanocomposites synthesized by one-pot method possess better homogeneous mixing of  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  and  $BaFe_{12}O_{19}$  phase than the composites prepared by physical mixing method. This difference in morphology of the samples may play important role in the magnetic and microwave absorption properties of the nanocomposites. EDX analysis (Fig. 4(c)) of final nanocomposite indicated the presence of all the elements (e.g. Mn, Ni, Zn, Fe, Ba and O).

Thermal, XRD, and microscopic analyses confirmed that, in one-pot synthesis route calcination of precursor powder leads to the formation of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$  nanocomposites. Here, precursor powders are prepared by mixing aqueous solution of metal nitrate with EDTA followed by drying of reaction mixture. EDTA, being a strong chelating agent, plays a significant role in the formation of nanocomposites. It prevents the segregation or intermittent precipitation of metal ions from solution during evaporation and also helps the formation of a fluffy, voluminous, porous carbon- rich precursor. During decomposition of precursor, nascent metal oxides form which are basically small atomic cluster with proper chemical homogeneity, imbedded into the precursor. These nascent metal oxides when calcined at 800 °C produce desired composite powders.<sup>39,43,44</sup>

#### 3.4. Magnetic measurements

VSM was used to measure the room temperature magnetization behaviours of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites prepared by two different methods (OP and PM method) with an applied field of 15000 Oe and shown in Fig. 5. The most important observation was that, composites-OP,

showed single hysteresis loop, signifying hard and soft phases were well exchanged coupled to each other. Whereas, composites-PM exhibited a typical two loop "bee waist" type hysteresis loop, indicating the absence of exchange coupling phase.1,24,34,35 and between hard soft Hence, (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>x</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites, prepared by one-pot method, though possessed crystallographycally two phase behaviour but demonstrated magnetically good single phase behaviour. Coercivity (H<sub>c</sub>) values of the nanocomposites prepared by both the methods increased with increasing hard ferrite phase (i.e.  $BaFe_{12}O_{19}$ ) content in the composite (Table. 3).

For the composites-OP, initial incorporation of BaFe<sub>12</sub>O<sub>19</sub> phase caused enhancement of saturation magnetization (M<sub>s</sub>) values which was due to the spring exchange coupling between hard and soft magnetic phases. However, subsequent increase of BaFe<sub>12</sub>O<sub>19</sub> content in the (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>x</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites (i.e., with increasing (1-x) values) did not affect much on M<sub>s</sub> value. Whereas, in case of composites-PM, M<sub>s</sub> values were increased with increasing amount of BaFe<sub>12</sub>O<sub>19</sub>. M<sub>s</sub> and H<sub>c</sub> values of all the composites prepared by one-pot method were higher than those of the composites prepared by physical mixing method. These facts indicate that hard and soft ferrite phases are sufficiently exchanged coupled to each other in the composites prepared by one-pot method. According to Moon et al.<sup>36</sup> M<sub>s</sub> of hard ferrite and soft ferrite nanocomposites without exchange coupling can be expressed as

$$M_{s} = M_{s,h} (1-f_{s}) + M_{s,s} f_{s}$$
(1)

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Where, M<sub>s,h</sub> and M<sub>s,s</sub> correspond to saturation magnetization of hard and soft ferrite respectively.  $f_s$  = weight fraction (percentage) of soft ferrite phases. Theoretically calculated values of M<sub>s</sub> of the composites (when hard and soft phases are not exchange coupling) were found to be almost matching with the experimentally obtained values of the composites prepared by physical mixing method (Table 3), which clearly indicates the absence of spin exchange coupling between hard and soft ferrite phases in the composites-PM. To the best of our knowledge, the values of the H<sub>c</sub> and Ms one-pot synthesized  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.15}$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>0.85</sub> are higher than the of available hard-soft ferrite reported values nanocomposites.<sup>1,2,3,4,34-38,45,46</sup>



**Fig 5:** Room temperature hysteresis loops for  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}$ - (BaFe<sub>12</sub>O<sub>19</sub>)<sub>0.15</sub> nanocomposite prepared by one-pot and physical mixing method.

#### 3.5. Microwave absorption study

The complex permittivity and permeability are usually used to analyze the dielectric and magnetic properties of absorber materials. Generally, the real parts ( $\epsilon'$  and  $\mu'$ ) signify the storage capability of dielectric and magnetic energy, whereas the imaginary parts ( $\epsilon''$  and  $\mu''$ ) stand for the loss of dielectric and magnetic energy.<sup>1,2,8</sup> The reflection loss (RL) was calculated from the complex relative permeability and permittivity at a given frequency and specimen thickness using a model of singlelayered plane wave absorber, proposed by Naito and Sutake.<sup>47</sup>

$$Z_{in} = Z_0 (\mu_r / \varepsilon_r)^{1/2} \tan h \left[ j \left( \frac{2\pi f d}{c} \right) (\mu_r \cdot \varepsilon_r)^{1/2} \right]$$
(2)

$$RL = 20 \log \left| \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \right|$$
(3)

Where,  $\mu_r = \mu' - j\mu''$  and  $\varepsilon_r = \varepsilon' - j\varepsilon''$  represents the relative complex permeability and permittivity of the absorber medium, respectively, *f* is the frequency of the electromagnetic wave, *d* is the absorber thickness, c is the velocity of light, Z<sub>0</sub> is the free space impedance, and Z<sub>in</sub> is the absorber impedance.

To determine the change of microwave absorption properties with thickness of the absorber, the reflection loss was calculated using equations (2) and (3) for different absorber thickness.

Microwave absorption behavior of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{x}$ -(BaFe<sub>12</sub>O<sub>19</sub>)<sub>1-x</sub> nanocomposites with different compositions (x= 0.85, 0.75, 0.5, 0.25) synthesized by one-pot method were investigated. Loss tangent vs. frequency were plotted to understand the particular loss mechanism for each composite and shown in Figure 6 (a) and (b). The dielectric and magnetic loss tangents can be expressed as  $\tan \delta_{\epsilon} = \epsilon''/\epsilon'$  and  $\tan \delta_{\mu} = \mu''/\mu'$ , respectively. Fig. 6 (b) shows that these composites are having only magnetic loss parameter whereas dielectric loss parameter is negligible (Fig. 6 (a)).

 $Table 3: M_s and H_c values of the (Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{x^-} (BaFe_{12}O_{19})_{1-x} nanocomposites prepared by one-pot and physical mixing method.$ 

Sample	One pot		Physica	Theoretical $M_s$	
	$H_{c}\left(\mathrm{O}e\right)$	$M_{\text{s}}(\text{emu/g})$	$H_{\rm c}({\rm O}e)$	$M_{\mathfrak{s}}(\text{emu/g})$	without exchange coupling
$Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4\text{-pure}$	61.1	39.1			
$\begin{array}{c} (Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}\text{-} \\ (BaFe_{12}O_{19})_{0.15} \end{array}$	923.7	59.3	459.1	47.1	46.9
$\begin{array}{c} (Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.75}\text{-} \\ (BaFe_{12}O_{19})_{0.25} \end{array}$	1577.7	57.6	1391.7	50.4	51.2
$(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.5}\text{-}\\(BaFe_{12}O_{19})_{0.5}$	3207.3	56.9	2466.8	53.9	53.4
$\begin{array}{c} (Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.25}\text{-} \\ (BaFe_{12}O_{19})_{0.75} \end{array}$	4450.9	55.8	2839.4	56.8	55.4
BaFe <sub>12</sub> O <sub>19</sub> -pure	4913.9	56.5			

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Fig 6 : (a) Loss Tangent of relative complex permittivity  $(tan\delta_{\epsilon})$ , (b) Loss Tangent of relative complex permeability  $(tan\delta_{\mu})$  and (c) Reflection loss vs. frequency plot  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$  nanocomposites synthesized by one-pot method.



Fig 7: (a) Real ( $\epsilon$ '), (b) imaginary ( $\epsilon$ ") parts of relative complex permittivity and (c) real ( $\mu$ '), (d) imaginary ( $\mu$ ") parts of relative complex permeability of pure BaFe<sub>12</sub>O<sub>19</sub>, Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub> nanopowders and (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>)<sub>0.85</sub>-(BaFe<sub>12</sub>O<sub>19</sub>)<sub>0.15</sub> nanocomposites prepared by one-pot and physical mixing method.

Reflection loss was calculated using equation (2 and 3) and plotted against frequency for different  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$  nanocomposites (x= 0.85, 0.75, 0.5, 0.25) synthesized by OP method for thickness 3 mm (Fig. 6 (c)). In RL vs frequency plot, the dip of the curves were designated for maximum absorption i.e. minimum RL. It was observed that, with decreasing value of x (i.e. increasing BaFe\_{12}O\_{19} content in the composite) maximum absorption was decreases. As the composite, having composition  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}$ - $(BaFe_{12}O_{19})_{0.15}$ , exhibited minimum reflection loss (~ -15dB) i.e. maximum absorption compare to other composites, so this

composition has been chosen for further studies and its microwave absorption properties were compared with the composite prepared by PM method as well as pure  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  and  $BaFe_{12}O_{19}.$ 

The real and imaginary permittivity (Fig. 7(a). and (b)) and permeability (Fig. 7 (c) and (d)) for the nanocomposites synthesized by both the methods, pure  $BaFe_{12}O_{19}$  and  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  nanopowders were plotted as a function of frequency in the X-band range (8.2- 12.4 GHz). It was observed that, over the entire frequency range,  $\varepsilon'$  values remained almost constant. Both nanocomposites  $\varepsilon'$  value was in between the values of pure  $BaFe_{12}O_{19}$  and  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  (Fig. 7(a)).

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The imaginary dielectric parameter ( $\epsilon$ ") values remained almost constant except a broad peak appeared in the range of 9.88 GHz to 12.4 GHz for the composite-OP (Fig. 7(b)). The intrinsic, electric dipole polarization and interfacial polarization are mainly responsible for this kind of behavior.<sup>4</sup> The real permeability ( $\mu$ ') values of composite and pure nanopowders remained almost constant throughout the entire frequency range (Figure 7 (c)). The imaginary permeability ( $\mu$ ") values (Fig. 7 (d)) showed a decreasing tendency from 8.2 GHz to 12.4 GHz for both the nanocomposites and pure ferrites, however, maximum imaginary permeability was observed for the composite synthesized by one-pot method.

Refection Loss for nanocomposites as well as pure ferrite nanopowders was calculated for absorber thickness 3 mm (Fig. 8 (a)). The estimated electromagnetic wave absorption values are listed in Table 4. In Fig. 8 (a), it was observed that, composite-OP method showed greater reflection loss ( $\sim -15$  dB at 9.88 GHz corresponds to  $\sim 96.84\%$  absorption) than composite-PM method ( $\sim -8.7$  dB at 8.62 GHz). Pure hard and soft ferrite nanopowders showed reflection loss lower than -10dB ( $\sim -7$  dB and  $\sim -6$  dB respectively). For composite-OP method showed >10 dB reflection loss (i.e. > 90% absorption) over the frequency range of 8.2 GHz to 10.6 GHz. Fig. 8 (b) illustrates the reflection loss vs frequency for one-pot synthesized nancomposites with different thickness of the absorber.

The reflection loss was found to be increased with increasing thickness of the sample till 3.5mm with the shifting of the frequency corresponds to maximum loss towards lower frequency.

Reflection loss of ~ -25 dB (i.e. > 99% absorption) was observed at 8.2 GHz for absorber thickness of 3.5 mm but frequency range corresponds to reflection loss > -10 dB (means > 90% absorption) becomes narrower than the specimen thickness of 3 mm. The RL value of -20 dB is equivalent to 99% absorption, which is considered as the satisfactory microwave absorption.

The improved microwave absorption property of hard- soft ferrite nanocomposites has already been reported by Shen et al.<sup>1</sup>, Tyagi et al.<sup>2</sup>, Chen et al.<sup>26</sup>, Xie et al.<sup>38</sup> and this enhancement of microwave absorption property was attributed to the exchange spin coupling interaction existing between hard and soft ferrite phases. Hence, in the present case the observed enhancement of microwave absorbing properties of the composites, prepared by one-pot method, was also considered due to the exchange coupling interaction between the hard (BaFe<sub>12</sub>O<sub>19</sub>) and soft (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>) ferrite phase.



**Fig 8:** Reflection loss vs. frequency plot for (a) pure  $BaFe_{12}O_{19}$ ,  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  nanopowders and  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}$ - $(BaFe_{12}O_{19})_{0.15}$  nanocomposites prepared by one-pot and physical mixing method and (b) Reflection loss vs. frequency plot for  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{0.85}$ - $(BaFe_{12}O_{19})_{0.15}$  nanocomposites prepared by one-pot method at different specimen thickness.

Fable 4 : Microwave absorption characteristics of pure BaFe <sub>12</sub> O <sub>19</sub> , Mn <sub>0.2</sub> Ni <sub>0.2</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> nanopowders and (Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.85</sub> -(BaFe <sub>12</sub> O <sub>19)0.</sub>
nanocomposites prepared by one-pot and physical mixing method.

Sample	Minimum RL (dB)	Frequency (GHz)
BaFe <sub>12</sub> O <sub>19</sub> -pure	-5.8	8.62
$(\mathbf{Mn}_{0.2}\mathbf{Ni}_{0.4}\mathbf{Zn}_{0.4}\mathbf{Fe}_{2}\mathbf{O}_{4})_{0.85}\textbf{-}(\mathbf{B}\mathbf{aF}\mathbf{e}_{12}\mathbf{O}_{19})_{0.15}$	-15	9.88
(one p ot method) (Mn <sub>0.2</sub> Ni <sub>0.4</sub> Zn <sub>0.4</sub> Fe <sub>2</sub> O <sub>4</sub> ) <sub>0.85</sub> -(B aFe <sub>12</sub> O <sub>19</sub> ) <sub>0.15</sub> (physical mixing method)	-8.7	8.62
$\mathbf{Mn}_{0.2}\mathbf{Ni}_{0.65}\mathbf{Zn}_{0.35}\mathbf{Fe}_{2}\mathbf{O}_{4}\text{-pure}$	-7.6	10.3

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In the composites (OP method), hard ferrite phase and soft ferrite phase are coupled to each other by an exchange through interfacial interaction which influences the relative complex permeability of the materials.<sup>2</sup> Interfacial interaction between the two phases is one of the important factors for the microwave absorption in the GHz frequency range.<sup>1</sup>

The interfacial multipoles in nanocomposite causes the surface spin of ferrite nanoparticles disordered, which leads to high magnetic loss. Therefore, the microwave absorption improves.<sup>1</sup> There will be stronger exchange coupling interaction at the interface if the grain size is smaller.<sup>2,26</sup> In addition, the nanocomposites, prepared by OP method, are made of nanoparticles and both  $Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4$  and  $BaFe_{12}O_{19}$  nanoparticles are homogeneously mixed, which would have a small size effect and enhanced spring exchange coupling interaction. In a hard-soft ferrite system, three types of magnetic interactions are present. The most important one is the exchange-coupling interaction between soft and hard phase, the remaining are dipolar interactions between the hard-hard and soft-soft ferrite phases.<sup>1,48</sup>

The hard ferrite possesses a high magnetocrystalline anisotropic energy compare to soft ferrite. If the hard ferrite grains are sufficiently exchange-coupled with the neighbouring soft ferrite grains, then the exchange-coupled interaction will not only help to align the magnetization in the soft ferrite phases but also helps to arrange magnetic moments of the hard and soft ferrite phases parallel to each other in hard soft ferrite nanocomposites. This leads to higher energy product and enhances the Reflection Loss in microwave absorption.<sup>1,27</sup>

In one-pot synthesis method, as both the phases are grown together from a single reaction mixture, intimate co-existence of nanosized hard and soft ferrite phases were observed in these nanocomposites (Fig. 3 (c, e, g, i)). This fact leads to sufficient exchange coupling between hard and soft ferrite phases, and supply marvellous opportunities for absorbing microwave and dissipating energy.

#### 4. Conclusion

A novel 'one-pot' synthetic methodology has been reported for the synthesis of  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_x$ - $(BaFe_{12}O_{19})_{1-x}$ nanocomposites. In this method, as hard ferrite phase (BaFe<sub>12</sub> $O_{19}$ ) and soft ferrite phase (Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>) both have grown together from a single reaction mixture, their intimate mixing in the final composite was observed. In the nanocomposites, prepared by one-pot method, homogeneous distribution of the hard and soft phase was better than in the nanocomposites, prepared by physical mixing method. Hard-soft ferrite nanocomposites thus formed (one-pot method) possessed excellent exchange spring coupling behaviour with high saturation magnetization as well as high coercivity. These composites also exhibited very good microwave absorption property in the X- band region. Nanocomposites, prepared by one-pot method, demonstrated superior magnetic as well as microwave absorption properties in comparison with the nanocomposites, prepared by physical mixing method, and pure BaFe<sub>12</sub>O<sub>19</sub> and Mn<sub>0.2</sub>Ni<sub>0.4</sub>Zn<sub>0.4</sub>Fe<sub>2</sub>O<sub>4</sub>nanoparticles.

This one-pot method, offers several advantages such as (i)

preparation method is simple, (ii) water is used as solvent, (iii) cheap metal nitrates are used as starting materials, (iv) unlike hydrothermal or sol-gel method, this method can be operated in open atmosphere and complicated reaction set up is not required, (v) composition of nanocomposites can simply be manipulated by mixing appropriate amount of metal nitrate solutions. The novelty of this method lies in its simplicity, requirement of relatively low calcination temperature, cost-effectiveness. Simple method of preparation, superior magnetic and microwave absorption properties make these nanocomposites potential candidates for application in next generation permanent magnets as well as radar absorbing materials.

The one-pot method has also shown its capability of producing nanocomposites with variety of compositions. Employing this one-pot method several other hard- soft ferrites nanocomposites, such as  $Ni_xZn_{(1-x)}Fe_2O_4$ -BaFe<sub>12</sub>O<sub>19</sub>,  $Ni_xZn_{(1-x)}Fe_2O_4$ -SrFe<sub>12</sub>O<sub>19</sub>,  $NiFe_2O_4$ -SrFe<sub>12</sub>O<sub>19</sub>,  $Ni_xZn_{(1-x)}Fe_2O_4$ -SrFe<sub>12</sub>O<sub>19</sub> etc., have also been prepared and results will be communicated shortly.

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#### **Notes and References**

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Electronic Supplementary Information (ESI) available: Room temperature M-H curve for  $(Mn_{0.2}Ni_{0.4}Zn_{0.4}Fe_2O_4)_{x^*}(BaFe_{12}O_{19})_{1.x}$ nanocomposites (X= 0.85, 0.75, 0.5, 0.25) prepared by one-pot and physical mixing method. See DOI: 10.1039/b000000x

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