Electronic Supplementary Information for **"Ferromagnetic Resonance of $NiCoFe_2O_4$ Nanoparticles and Microwave Absorption Properties of Flexible $NiCoFe_2O_4$-Carbon Black/Poly(vinyl alcohol) Composites"**

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1. The synthesis procedure

The schematic of synthesis procedure of ap-NCF NPs and ap-NCF ferrite-CB hybrids discussed in manuscript is given below.

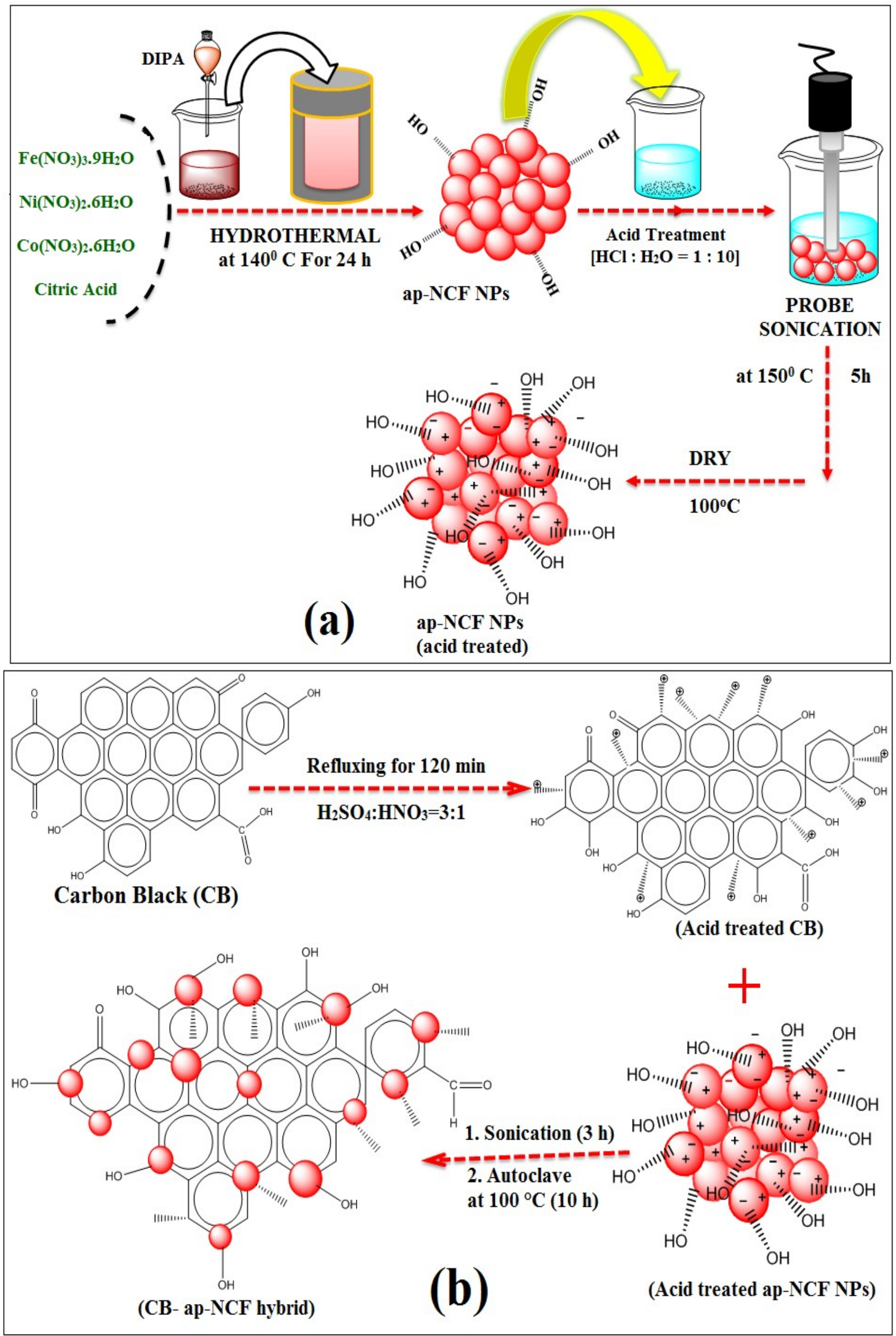


Figure-S1: The synthesis procedure for (a) ap-NCF NPs and (b) ap-NCF ferrite-CB hybrid

2. Magnetic Properties of Nanoparticles: The magnetization curves for both as-prepared and annealed NCF NPs at room temperature, 300 K were recorded by a VSM with applied field up 2

Tesla. The M-H loops of both the NPs along with their initial magnetization curve are shown in Figure-S2. The initial magnetization curves fitted with the law of approach to saturation in which the saturation magnetization (M_S) and magnetic anisotropy constant (K) can be estimated by equation:[1]

$$M = M_S\left(1 - \frac{a}{H} - \frac{b}{H^2}\right) + cH \qquad (S1)$$

Where, H = applied magnetic field, $= \frac{8}{105}\frac{K^2}{M_S^2}$, 'a' is a constant and 'c' corresponds to the high-field magnetic susceptibility. The parameters so obtained are presented in the Table 1.

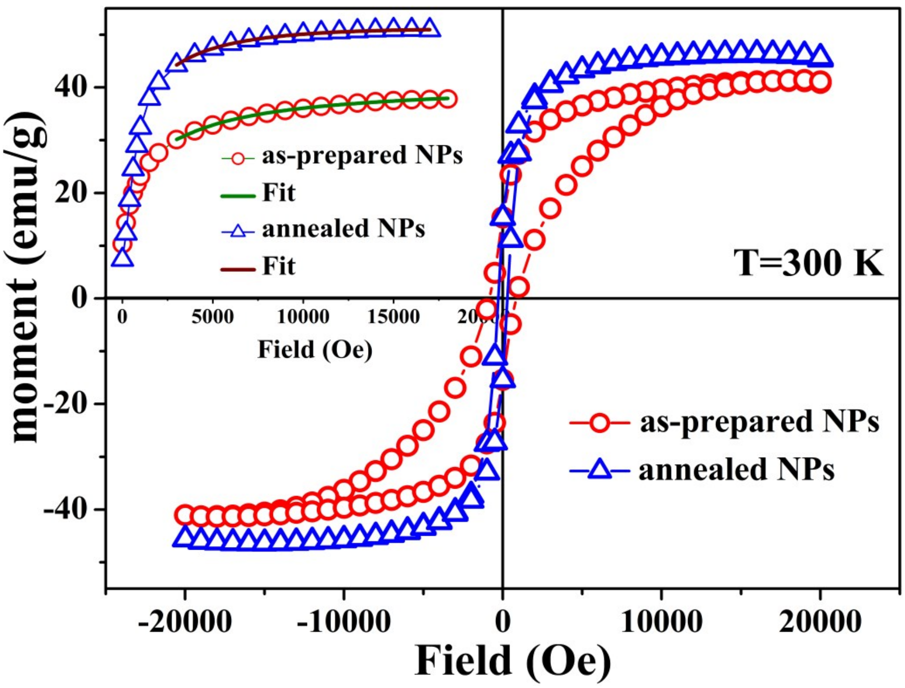


Figure S2: Magnetization curve with initial magnetization (insert of LHS) for both the samples.

Table-S1: Magnetic parameters of both the samples at 300 K

Samples Name	M_s (emu/gm)	H_C (Oe)	K (erg/cm^3)
as-prepared NCF NPs	41	775	$1.5x10^5$
Annealed NCF NPs	55.24	225	$0.88x10^5$

The annealed (as-prepared) NPs exhibit higher (lower) saturation magnetization and lower (higher) magnetocrystalline anisotropy and coercivity. The magnetocrystalline anisotropy and coercivity is at least 200% more in as-prepared as compared to annealed NPs (see Table-S1). This large value of anisotropy in the as-prepared NPs can be mainly attributed to non-stoichiometric divalent cation distribution. It is clear from cation distribution of NPs that, in case of as-prepared NPs the considerable amount of Ni^{2+} ions resides at A-site, while on the other hand in annealed NPs, most of the Ni^{2+} ions resides at B-site. It has been reported[2] that the Co^{2+} ions at the B- site and Ni^{2+} ions at the A-site induce the high magnetocrystalline anisotropy due to incomplete quenching of orbital momentum. Since Co^{2+} and Fe^{3+} cations occupancies does not change much after annealing, which indicate that non-equilibrium occupation of Ni^{2+} ions at A-site in as-prepared NPs results in large magnetocrystalline anisotropy and hence coercivity.

3. AC conductivity of NCF-CB/PVA composite:

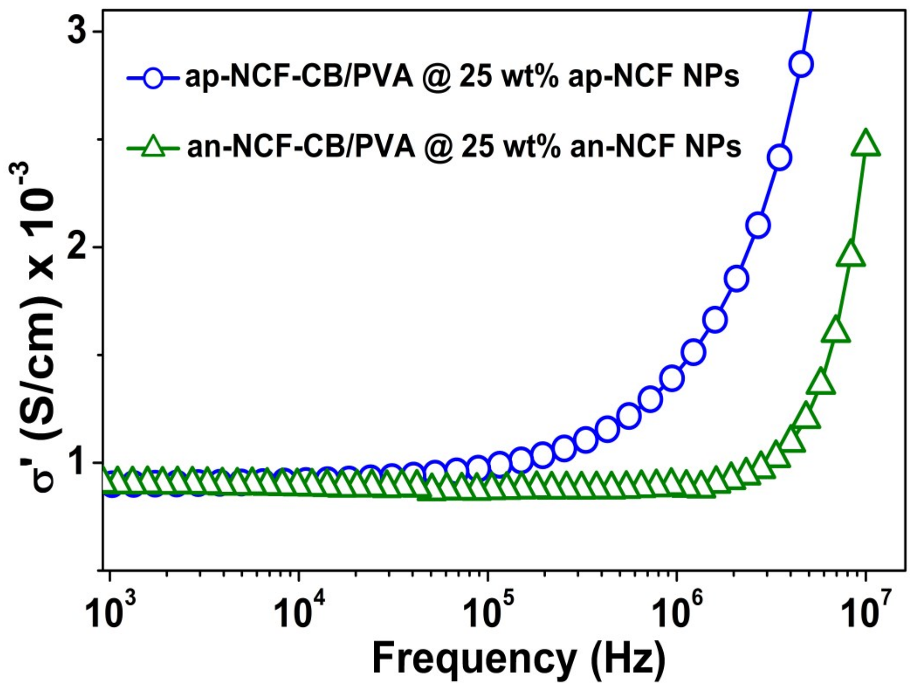
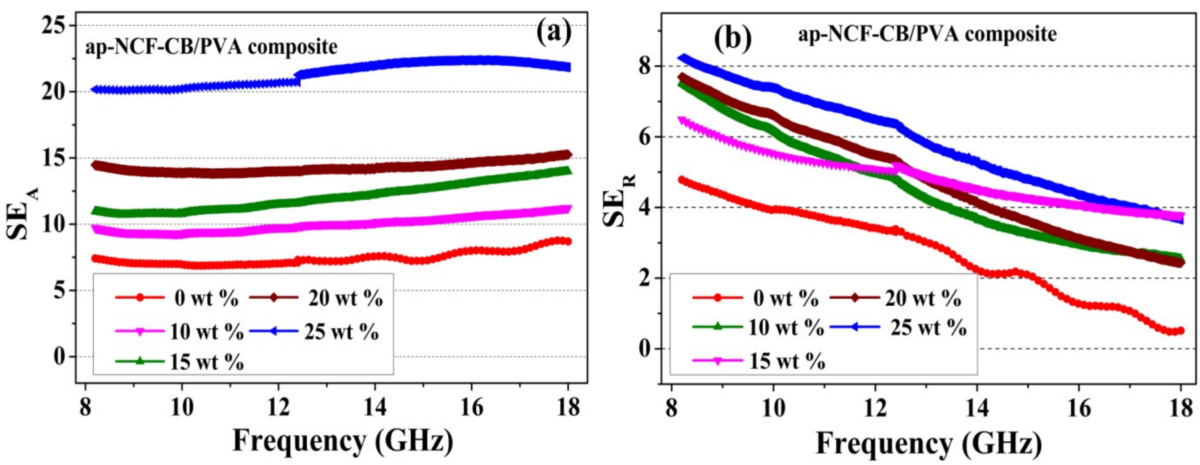


Figure-S3: The comparison of the real part of AC conductivity of the 25 wt% ap-NCF-CB/PVA composite (in blue color) and 25 wt% an-NCF-CB/PVA (in olive color).

4. Effect of varying wt% of ap-NCF NPs in EM wave absorption properties: Figure-S4 illustrates the total shielding effectiveness SE, of the ap-NCF based composite films; in these ap-NCF-CB/PVA composite based films, the wt% of ap-NCF NPs is varied from 0 wt% to 25 wt%; while the CB wt% was fixed at 15 wt% throughout the experiments. It can be observed from the figure-S3 that on increasing the wt% of ap-NCF NPs in ap-NCF-CB/PVA composite from 0 wt% to 25 wt%, the total shielding effectiveness SE is found to increase from 12 dB to 27 dB. This increase in SE of ap-NCF-CB/PVA composite on increasing ferrite NPs wt% can be attributed to: (a) enhancement in interfacial dielectric polarization as discussed in dielectric section as on increasing the wt% of ferrite NPs number of interfaces in ap-NCF-CB/PVA composite increases; and (b) enhancement in magnetic losses due to the magnetic nature of an-NCF NPs through various mechanism. It can be noted from the figure-S3 that above 20 wt% of ap-NCF NPs in composite films, SE achieves the value above 20 dB (99.5 % attenuation of EM wave), necessary for commercial level applications[31], therefore it can be concluded that, above 20 wt% of ap-NCF loading in composite, these composite films can be used as a efficient microwave absorber. It is also important to note that the ap-NCF-CB/PVA composite with 25 wt% of ap-NCF shows the maximum value for shielding due to reflection SE_R, which is in between 8 to 5 dB (figure-S3 (a, b), in supplementary sheet).



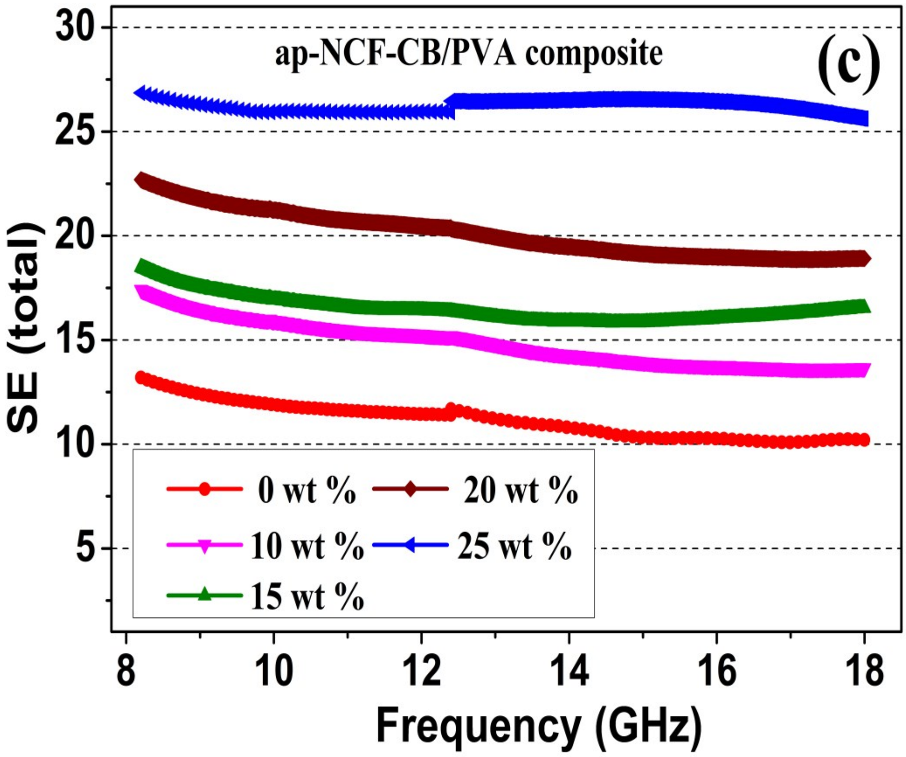


Figure-S4: The shielding effectiveness (a) due to absorption (SE_A), (b) due to reflection (SE_R) and (c) total shielding effectiveness (SE) of all ap-NCF-CB/PVA composite with different wt% of ap-NCF NPs (0 wt%, 10 wt%, 15 wt%, 20 wt% and 25 wt%).

5. The eddy current losses in NCF-CB/PVA composite: The eddy current losses of a system can be evaluated by the equation: $\mu^{''} \approx 2\pi\mu_0(\mu^{'})^2\sigma d^2 f/3$, where μ_0 is the permeability of free space, d is the thickness of the sample and σ is the electrical conductivity[3, 4]. If eddy current losses are the main contribution to the magnetic loss in the system, then the value of the factor $\mu^{''}(\mu^{'})^2 f^{-1}$ must be a constant over entire frequency rage. However it can be seen from the figure-S5, that the $\mu^{''}(\mu^{'})^{-2} f^{-1}$ value is not independent of frequency; which implies that the eddy current losses are not only contribution to the magnetic losses but natural resonance and FMR loss also contributes to the magnetic losses in these composite.

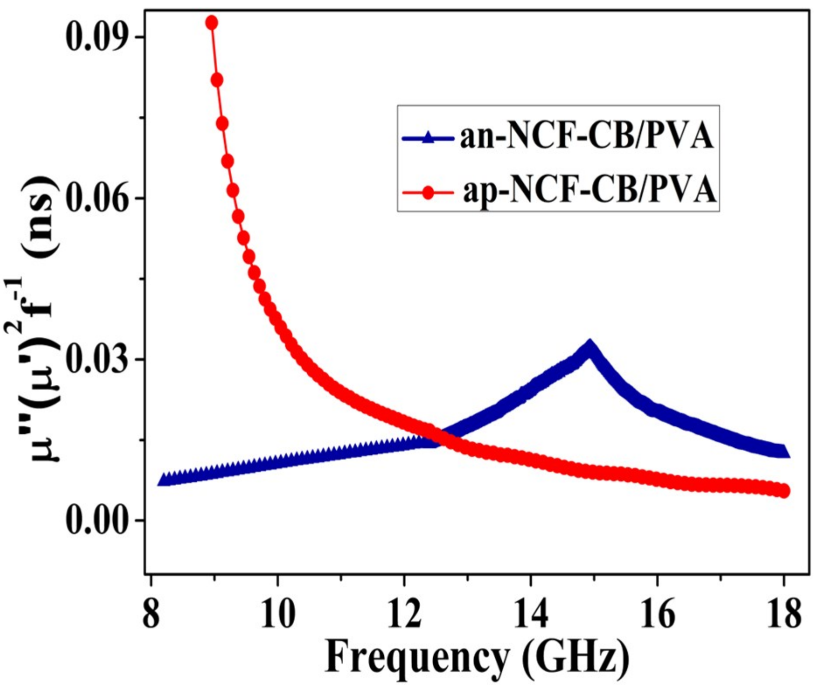


Figure-S5: Plot of $\mu^{''}(\mu^{'})^{-2}f^{-1}$ versus frequency for ap-NCF-CB/PVA composite ap-NCF-CB/PVA composite (for 25 wt % of NCF NPs).

6. The impedance matching:

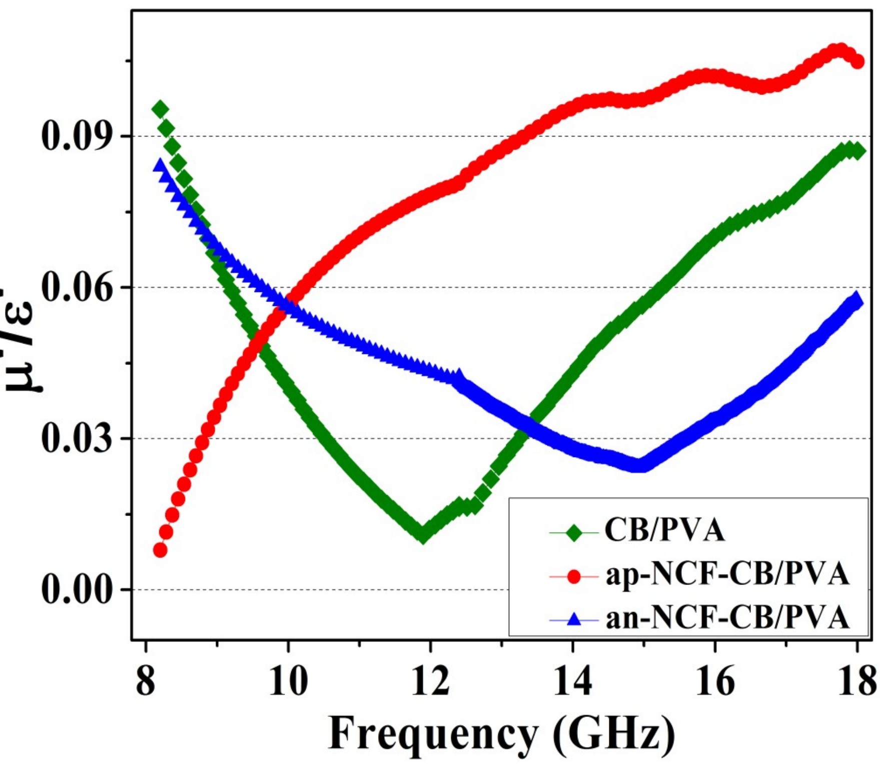


Figure-S6: The ratio of $\varepsilon^{'} and\ \mu^{'}$ i.e. $\left(\frac{\mu^{'}}{\varepsilon^{'}}\right)$ for three different composites namely CB/PVA, ap-NCF-CB/PVA and an-NCF-CB/PVA.

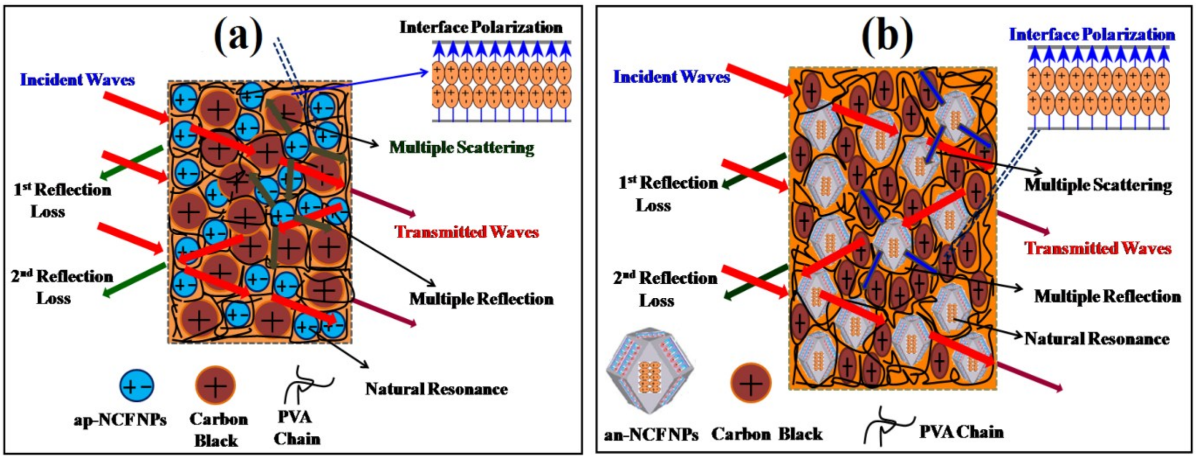


Figure-S7: The schematic representation of the proposed microwave shielding mechanism,(a) in ap-NCF-CB/PVA composite film and (b) in an-NCF-CB/PVA composite film.

References:

1. Morrish, Allan H. "The physical principles of magnetism." ISBN 0-7803-6029; Wiley-VCH, **2001**.
2. Desai, Mrugesh, Shiva Prasad, N. Venkataramani, Indradev Samajdar, A. K. Nigam, N. Keller, R. Krishnan, E. M. Baggio-Saitovitch, B. R. Pujada, and A. Rossi. "Anomalous variation of coercivity with annealing in nanocrystalline NiZn-ferrite films." *Journal of applied physics*, **2002**, 91, 7592-7594.
3. Tong, Guoxiu, Yun Liu, Tingting Cui, Yana Li, Yanting Zhao, and Jianguo Guan. "Tunable dielectric properties and excellent microwave absorbing properties of elliptical Fe_3O_4 nanorings." *Appl. Phys. Lett.,* **2016**, 108, 072905.
4. Arief, Injamamul, Sourav Biswas, and Suryasarathi Bose. "Tuning the Shape Anisotropy and Electromagnetic Screening Ability of Ultrahigh Magnetic Polymer and Surfactant-Capped

FeCo Nanorods and Nanocubes in Soft Conducting Composites." *ACS Appl. Mater. Interfaces,* **2016**, 8, 26285-26297