Room-Temperature Ferromagnetic $Sr_3YCo_4O_{10+\delta}$ and Carbon Black Reinforced Polyvinylidenefluoride Composites toward High-Performance Electromagnetic Interference Shielding

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Synthesis of $Sr_3YCo_4O_{10+\delta}$ (SYCO)

Oxygen deficient double perovskite $Sr_3YCo_4O_{10+\delta}$ (SYCO) was synthesized by solid state ceramic route. Stoichiometric amounts of $SrCO_3$, Y_2O_3 and Co_3O_4 were ballmilled in acetone medium for 24 hours. The uniform mixture was dried at 80 ℃ and then precalcined at 900 ^{0}C for 12 hours in order to decompose $SrCO_3$. The precalcined powder was pelletized and calcined at 1050 ^{0}C for 15 hours with preheating rate 1.16 ℃/min. and then ground well to obtain fine powder.

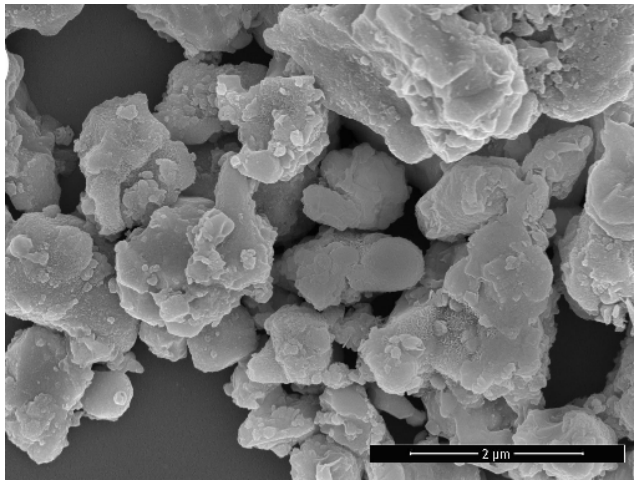

Figure S1. SEM image of SYCO

XPS Analysis of SYCO

The elemental components and various oxidation states of Co ions in SYCO were identified by X- ray photoelectron spectroscopy and the spectra are shown in Fig. S2. The survey spectrum in Fig. S2 (a)

shows respective elemental composition of the compound and the Co 2p scan (Fig. S2 (b)) indicates the presence of both Co^{2+} as well as Co^{3+}ions as surface states. Co $2p_{3/2}$ signal at 780.3 eV corresponds to Co^{3+} and peak at 781.4 eV along with shake-up satellites at binding energies (BE) 784.5 and 789.7 eV are due to Co^{2+} ions. In Co $2p_{1/2}$ signal, along with the Co^{3+} feature at 795.3 eV, peak at 796.7 eV and a shake-up satellite feature at 804.7 eV are attributed to Co^{2+} states were also present [1-4]. In this case the Co^{3+}/Co^{2+} molar ratio calculated from fitted area is 0.53 which indicates more concentration of Co^{2+} ions which in turn is an indication of more oxygen deficient sites in SYCO. The presence and distribution of Co ions having various oxidation states and spin states in the SYCO crystal lattice determines the type of magnetism in the compound.

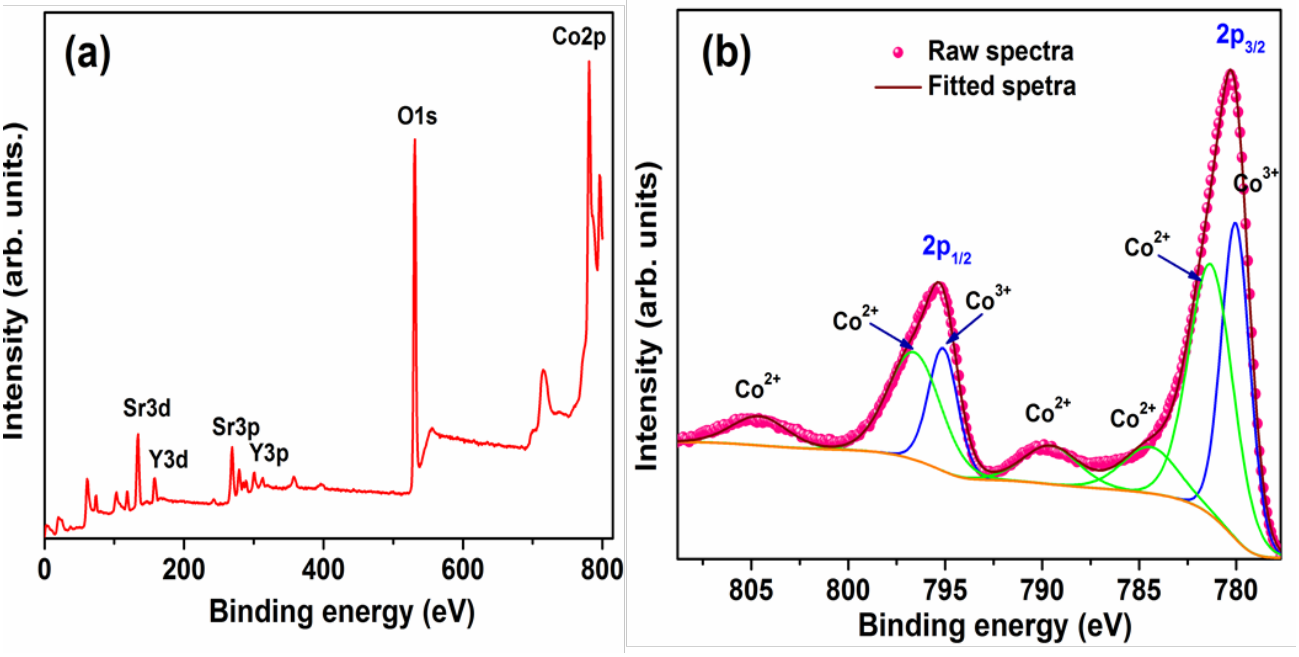

Figure S2.(a) XPS survey spectrum and (b) Co2p spectra of SYCO

Magnetic properties

Fig. S3 (a) shows Zero Field Cooled (ZFC) and Field Cooled (FC) temperature dependence of magnetization, measured for SYCO within the temperature range 50 – 400 K. In the field cooling process, the magnetic field applied is 100 Oe. The magnetization dependence shows a ferromagnetic nature of the compound having a clear transition temperature at 335 K with a saturation magnetization 0.30 emu/g. In the ZFC and FC curve, a significant divergence is observed, especially at lower temperatures, which is generally attributed either to magnetic anisotropy of the material or a competing interaction between ferromagnetic (FM) and antiferromagnetic (AFM) states [5, 6]. In 2005, Kobayashi *et al.* [7] studied the variation in magnetic properties of $Sr_{1-x}Y_xCoO_{3-\delta}$ (x = 0.1-0.4) and reported that the compound is a room temperature ferromagnet in a narrow compositional range $0.2 \leq x \leq 0.25$, and oxygen deficiency is

essential for this.They suggest a mechanism for the observed magnetism like that: at lower temperatures, the antiferromagnetic interaction along b axis due to super exchange interactions dominates in the compound and at temperatures above the transition temperature, thermally excited carriers induces double exchange interaction to align ferromagnetic chains along the a axis stabilize a ferromagnetic state. Here the observed magnetization is less than that of reported. This may be due to the difference in the oxygen stoichiometry of the synthesized compound from that of reported.

Fig. S3 (b) shows the variation of isothermal magnetization of SYCO and PCS-30 with applied magnetic field at room temperature (300 K). The hysteresis loop shows ferromagnetic behavior of the materials but no saturation is observed up to ±30 kOe that may due to insufficient applied magnetic field. However, the maximum value of magnetization observed in SYCO is 4.2 emu/g. The incorporation of SYCO in PC matrix results the reduction in magnetization value from that of SYCO to 1.8 emu/g.

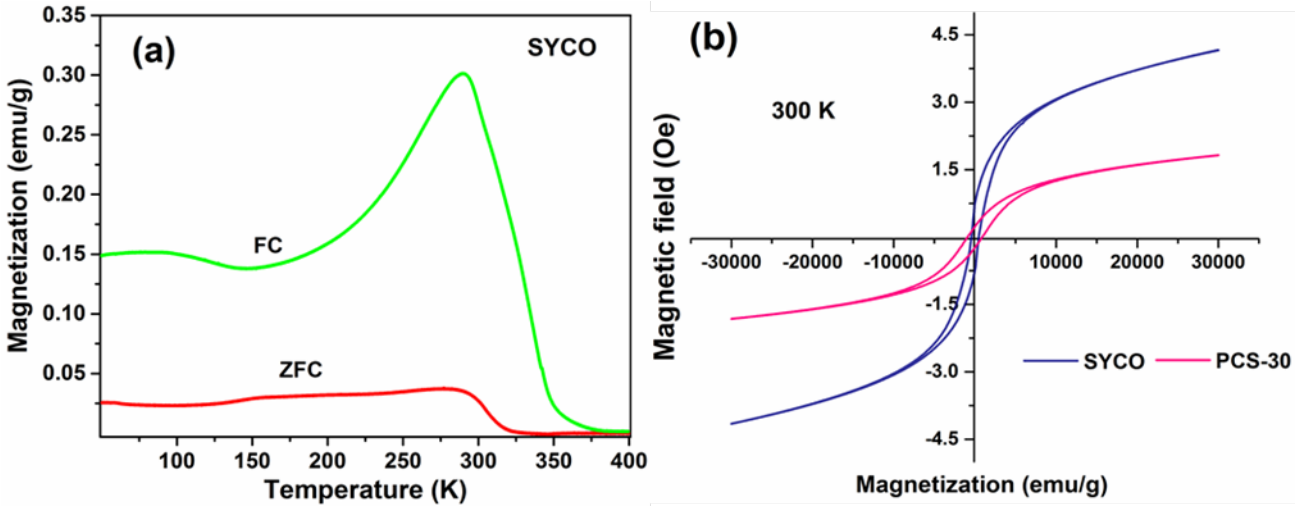

Figure S3. (a) Variation of magnetization as a function of temperature measured for SYCO and (b) Isothermal magnetization of SYCO and PCS-30 at 300 K

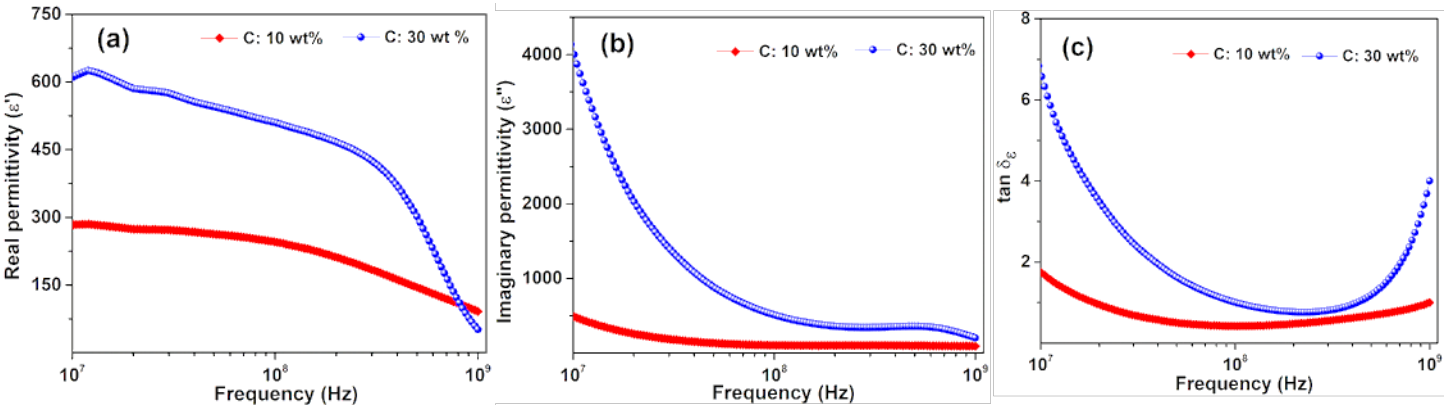

Figure S4. (a) The real part and (b) imaginary part of permittivity of PVDF/CB composites and (c) their loss tangent within 10 MHz to 1 GHz frequency range.

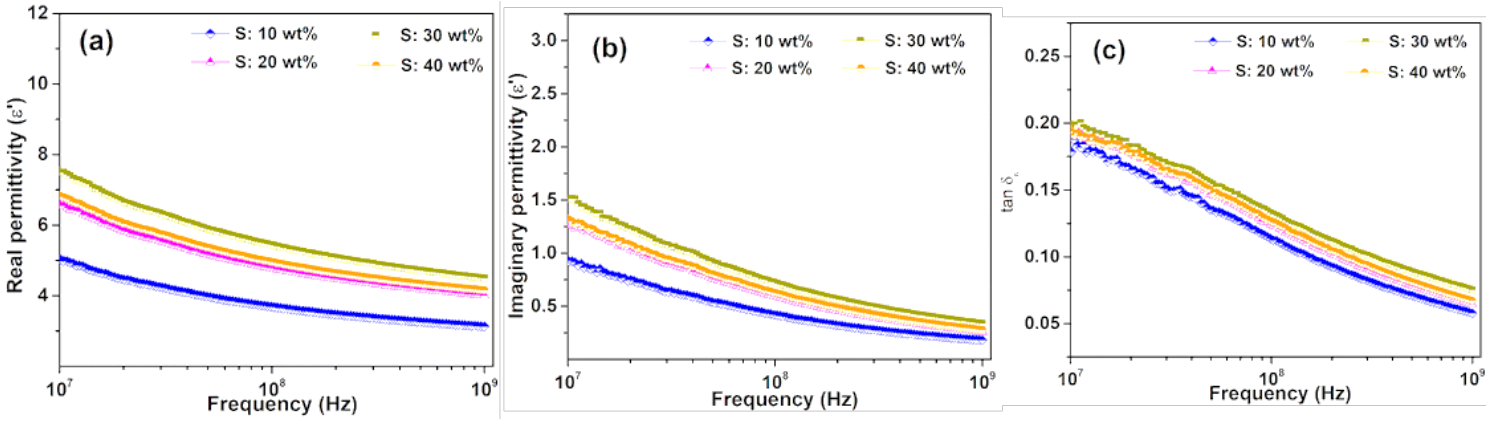

Figure S5. (a) The real part and (b) imaginary part of permittivity of PVDF/SYCO composites and (c) their loss tangent within 10 MHz to 1 GHz frequency range.

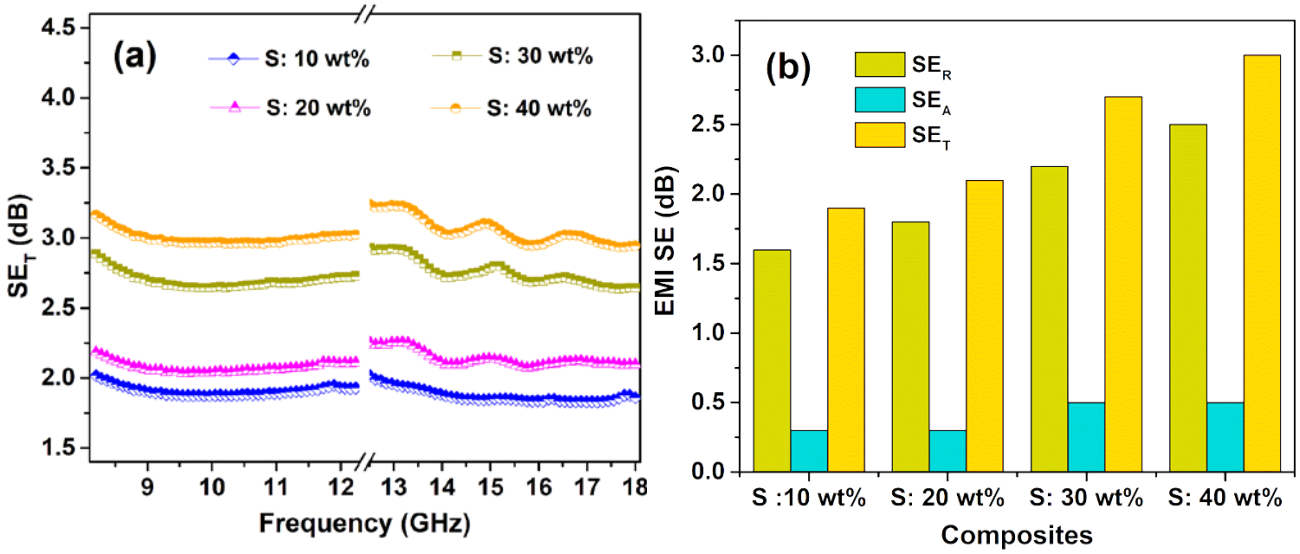

Figure S6. (a) Total EMI SE of PVDF/SYCO composites in the X and Ku band region (b) Average values of SE_A ,SE_R and SE_A of PCS composites.

Table S1 Average EMI SE values of PCS composites in the 8.2-18 GHz range

Composites	**EMI SE (dB)**			**Shielding efficiency (%)**	**SSE** $(\boldsymbol{dBcm^2g^{-1}})$
	SE_R	SE_A	SE_{Total}		
P	1.0	0.3	1.3	26	4
PC	9.0	20.0	29.0	99.9	82
PCS- 10	7.9	30.5	38.4	99.98	105
PCS- 20	8.5	37.0	45.5	99.997	112
PCS- 30	8.6	39.7	48.3	99.998	111
PCS- 40	9.0	41.2	50.2	99.9991	113

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