

Supporting Information

Frequency and Temperature Dependent Dielectric Properties of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$: Polarization and Conduction Mechanisms Pertained to Crystallographic Symmetry and Electronic Transitions

**Arun Raj R S¹, Aruna Joseph¹, Shamima Hussain², Mohd Fahad³, Tuhin Maity⁴, P M
Sarun³ and Lija K Joy^{1, *}**

¹ Centre for Advanced Functional Materials (CAFM), Department of Physics, Bishop Moore College, Mavelikara, Alappuzha, Kerala, 690110, India

² UGC-DAE Consortium for Scientific Research, Kalpakkam Node, Kokilamedu-603104, India

³ Functional Ceramics Laboratory, Department of Physics, Indian Institute of Technology (Indian School of Mines), Dhanbad, 826004, India

⁴ School of Physics, Indian Institute of Science Education and Research, Thiruvananthapuram, Kerala 695551, India

* Corresponding author email: - lijakjoy@gmail.com

3. Results and Discussions

3.1 Structural and Compositional Studies

3.1.2 FTIR studies

Table S1. Vibrational bands of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.08$)

Composition	ν_t (cm^{-1})	ν_o (cm^{-2})
0.00	536.16	289.16
0.02	541.84	306.64
0.04	546.00	315.93
0.06	550.14	329.92
0.08	552.49	335.86
0.10	549.25	330.15

3.1.3 XPS Studies

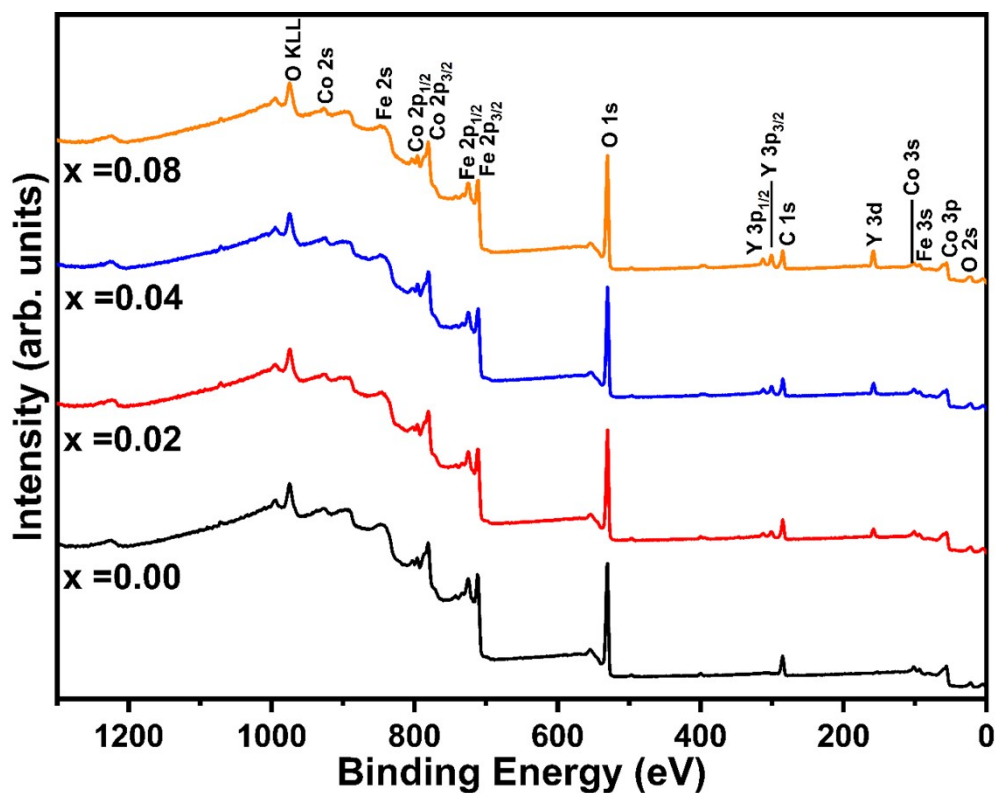


Figure S1. Wide scan survey spectrum of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.08$) at room temperature.

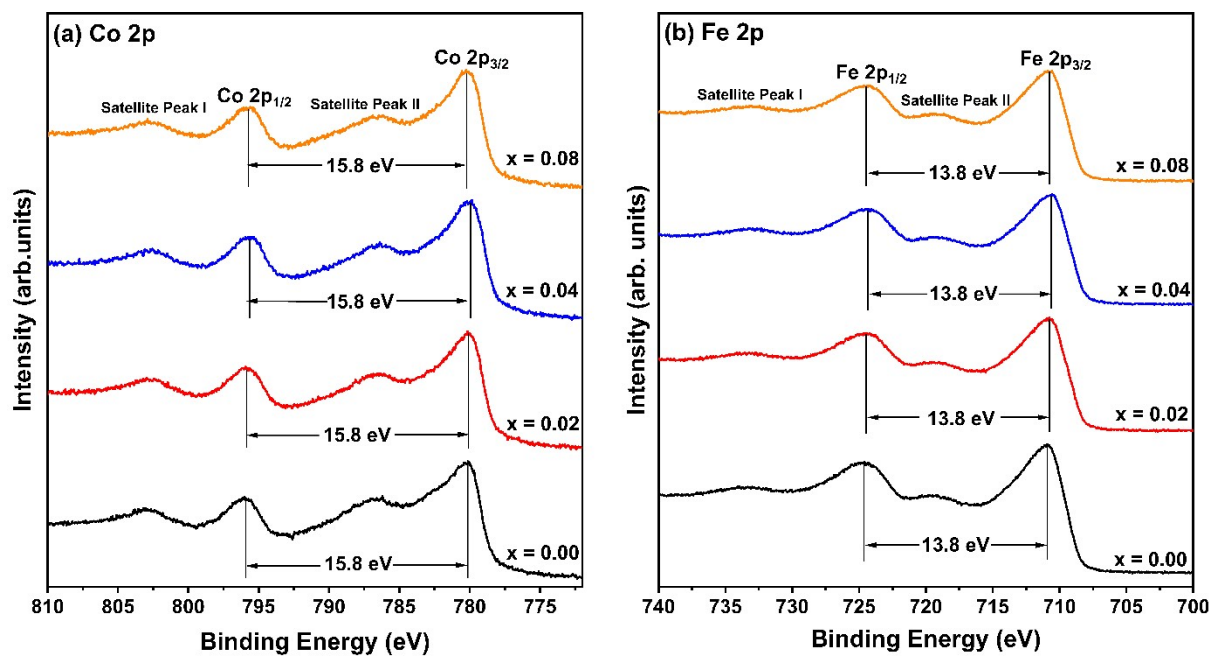


Figure S2. (a) Co 2p core level XPS spectra and (b) Fe 2p core level XPS spectra of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.08$)

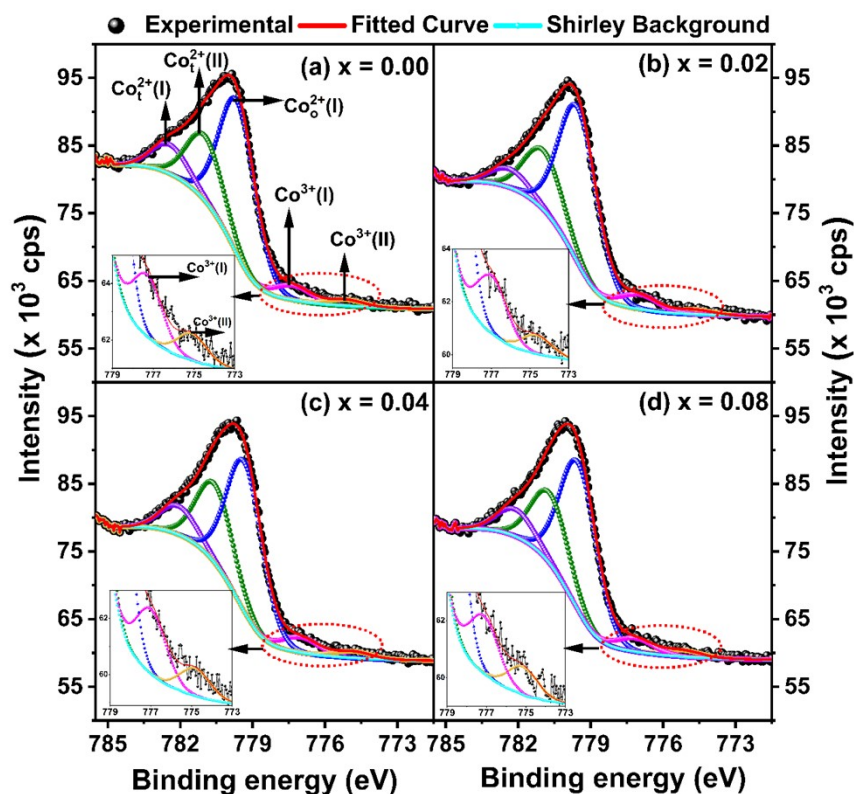


Figure S3. Deconvoluted Co $2p_{3/2}$ XPS spectra of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04$ and 0.08). The inset shows the magnified image of $\text{Co}^{3+}(\text{I})$ and $\text{Co}^{3+}(\text{II})$ peaks in $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$.

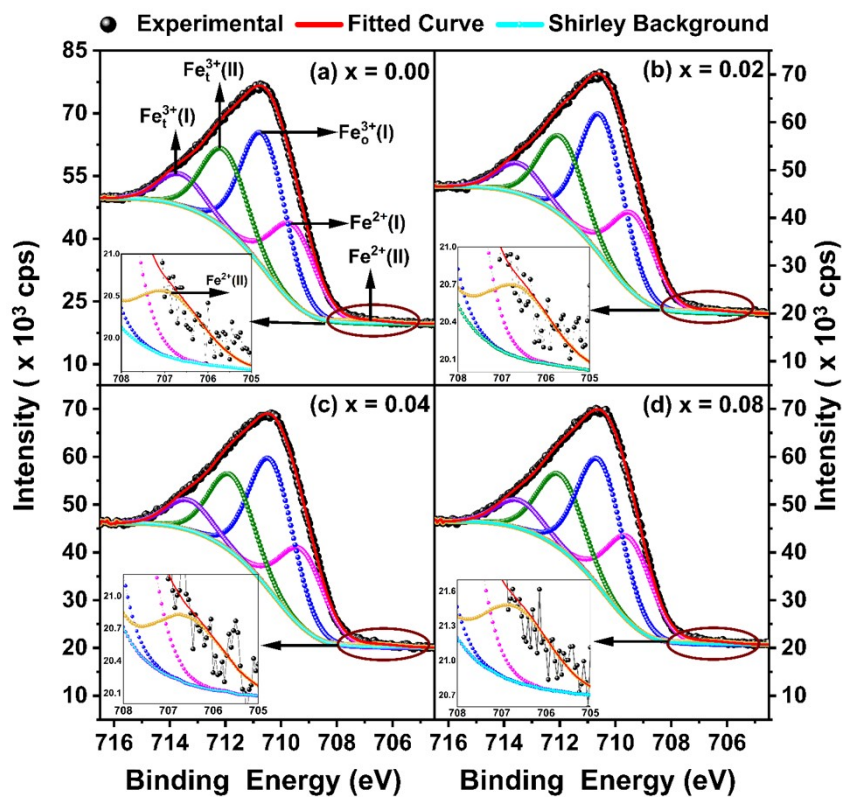


Figure S4. Deconvoluted Fe $2p_{3/2}$ XPS spectra of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04$ and 0.08). The inset shows a magnified image of the $\text{Fe}^{2+}(\text{II})$ peak in $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$.

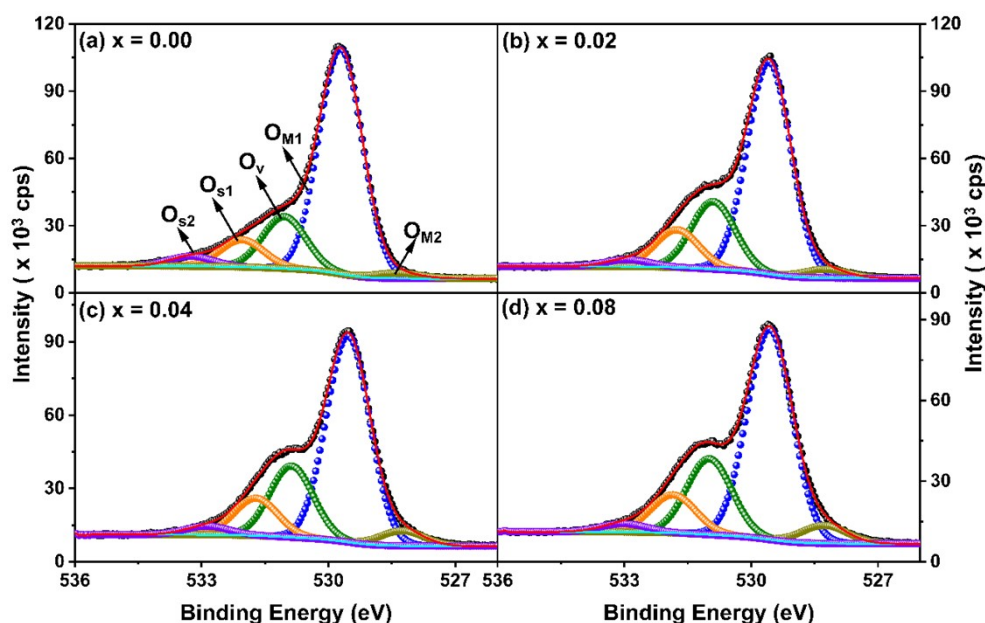


Figure S5. Deconvoluted O1s XPS spectra of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04$ and 0.08).

Table S2: (a) Binding energy of Co, Fe, and Y in $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04$, and 0.08) system.

Binding Energy in eV					
Spectrum	Assignment	Composition			
		x = 0.00	x = 0.02	x = 0.04	x = 0.08
Co 2p _{3/2}	$\text{Co}_t^{2+} (I)$	782.43	782.37	782.06	782.11
	$\text{Co}_t^{2+} (II)$	780.97	780.95	780.57	780.69
	$\text{Co}_o^{2+} (I)$	779.62	779.52	779.32	779.50
	$\text{Co}^{3+} (I)$	777.31	776.99	777.00	777.15
	$\text{Co}^{3+} (II)$	775.11	774.66	774.83	775.17
Fe 2p _{3/2}	$\text{Fe}_t^{3+} (I)$	713.60	713.46	713.30	713.48
	$\text{Fe}_t^{3+} (II)$	712.04	711.94	711.77	711.97
	$\text{Fe}_o^{3+} (II)$	710.62	710.49	710.34	710.54
	$\text{Fe}^{2+} (I)$	709.55	709.33	709.24	709.48
	$\text{Fe}^{2+} (II)$	706.92	706.66	706.58	706.72
Y 3d _{3/2}	Y_o^{3+}	--	159.37	159.28	159.63
	Y_{ov}^{3+}	--	--	--	158.58
Y 3d _{5/2}	Y_o^{3+}	--	157.29	157.21	157.58
	Y_{ov}^{3+}	--	--	--	156.45
O 1s	O	533.25	532.93	532.87	533.02
	O _s	532.04	531.75	531.71	531.84
	O _v	531.04	530.90	530.87	530.99

	O _{M1}	529.71	529.58	529.53	529.56
	O _{M2}	528.37	528.24	528.25	528.27

3.1.4 EDS Analysis

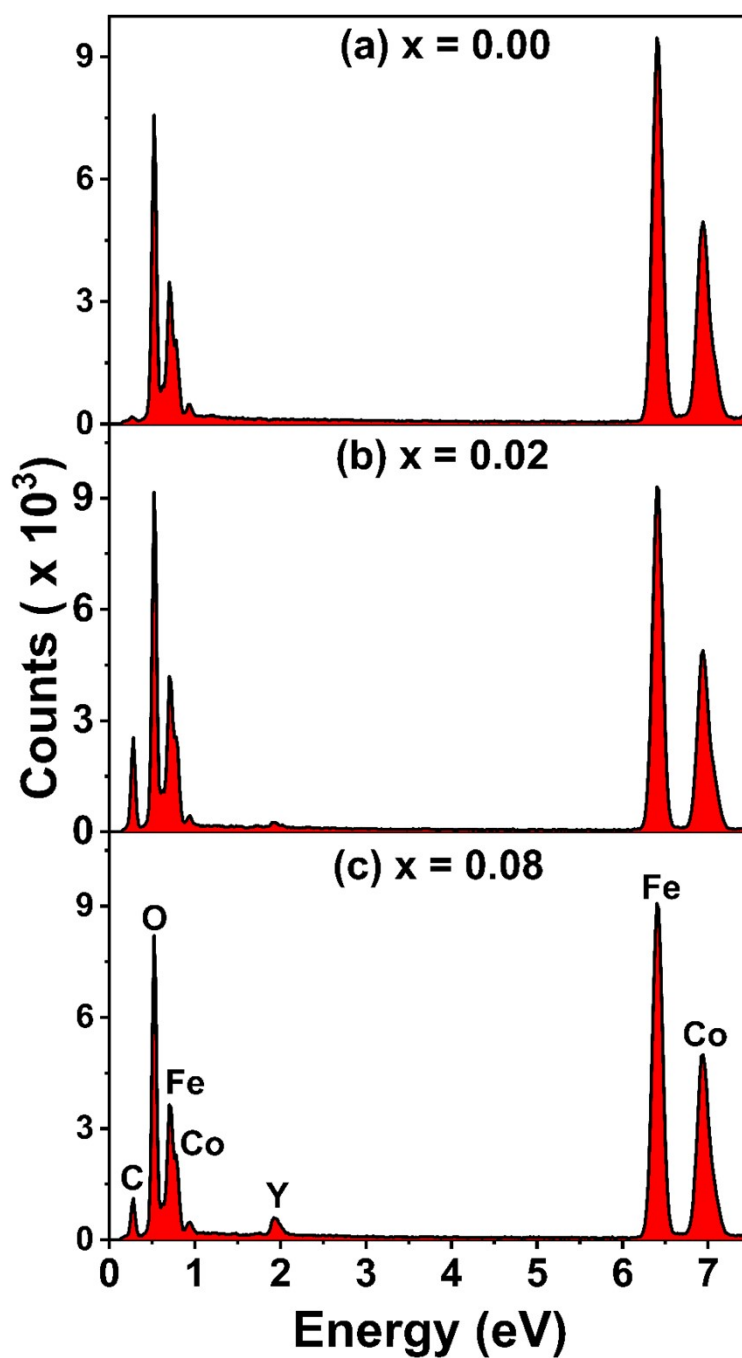


Figure S6. EDS micrographs of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02$ and 0.08)

Table S3. The atomic ratio of Co/Fe and Y/Fe in $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02$ and 0.08).

Composition	Co/Fe ratio		Y/Fe ratio	
	From EDS	Theoretical	From EDS	Theoretical
0.00	0.55	0.5	0	0
0.02	0.53	0.51	0.01	0.01

0.08	0.58	0.52	0.03	0.04
------	------	------	------	------

3.2 Morphological studies

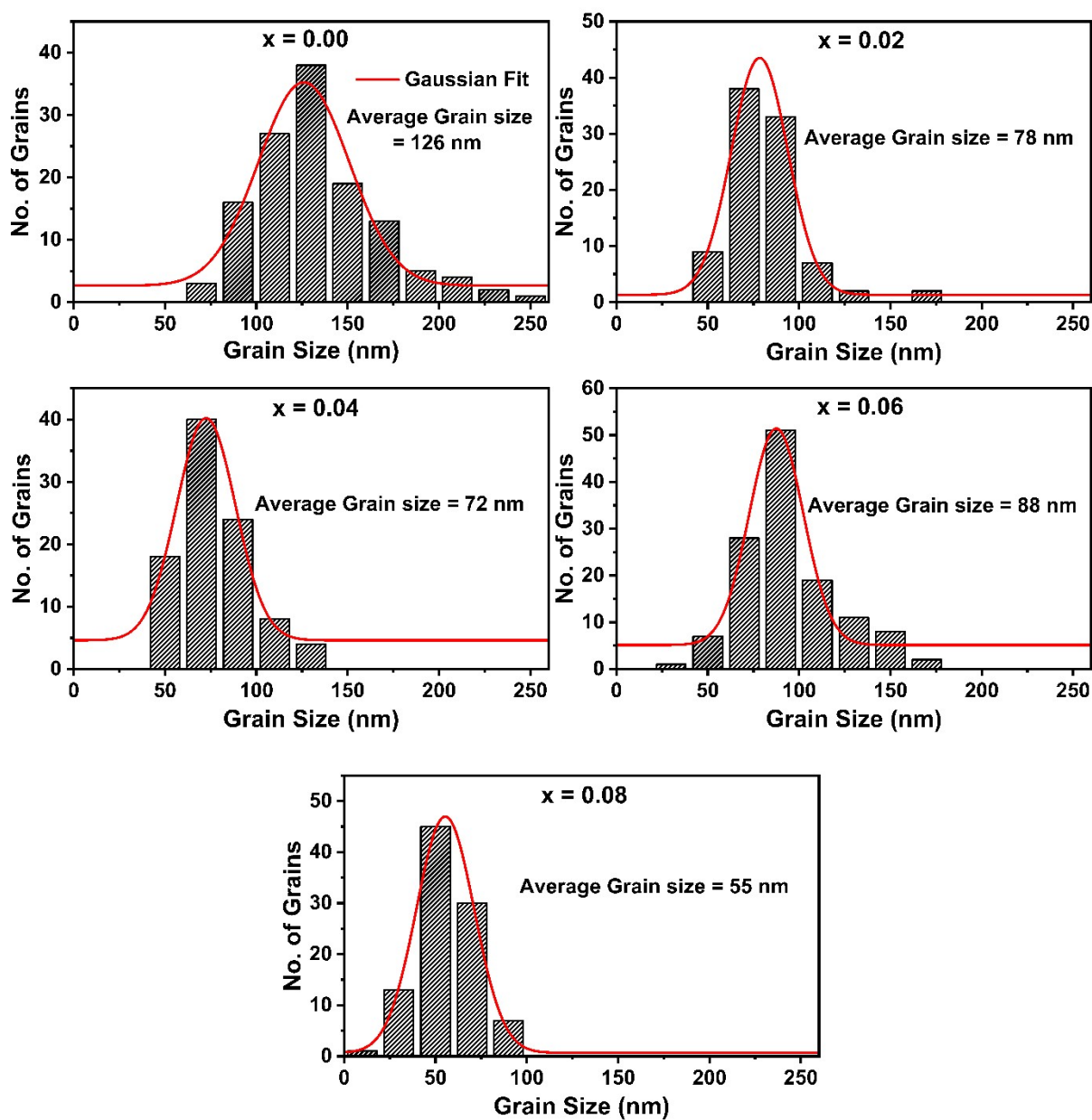


Figure S7 Grain distribution curves of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06$, and 0.08)

3.3 Optical Studies

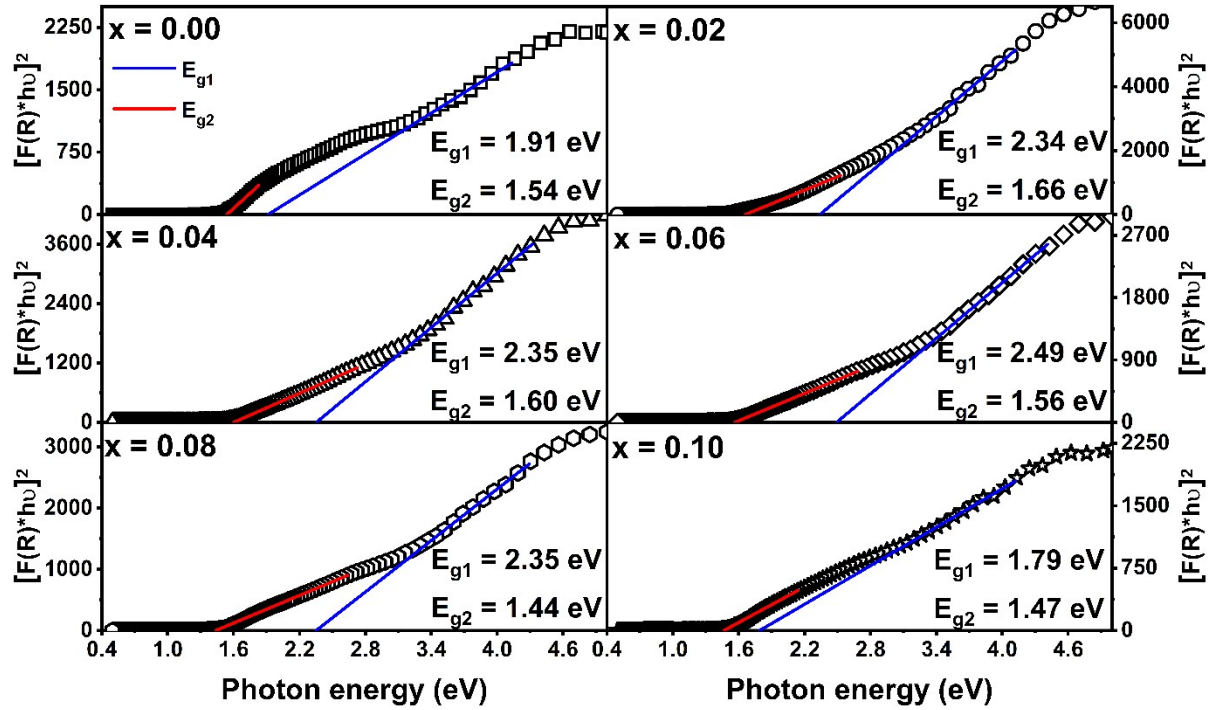


Figure S8. Direct band gap calculation of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06, 0.08$ and 0.10)

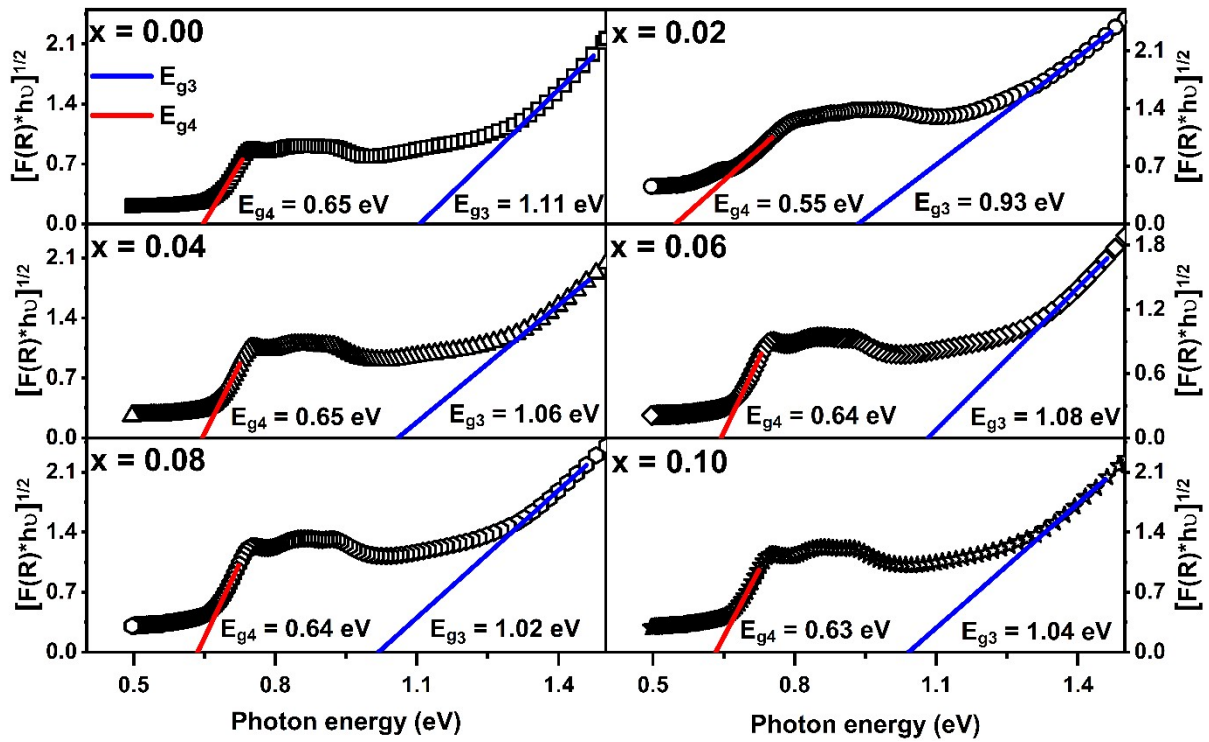


Figure S9. Indirect band gap calculation of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06, 0.08$ and 0.10)

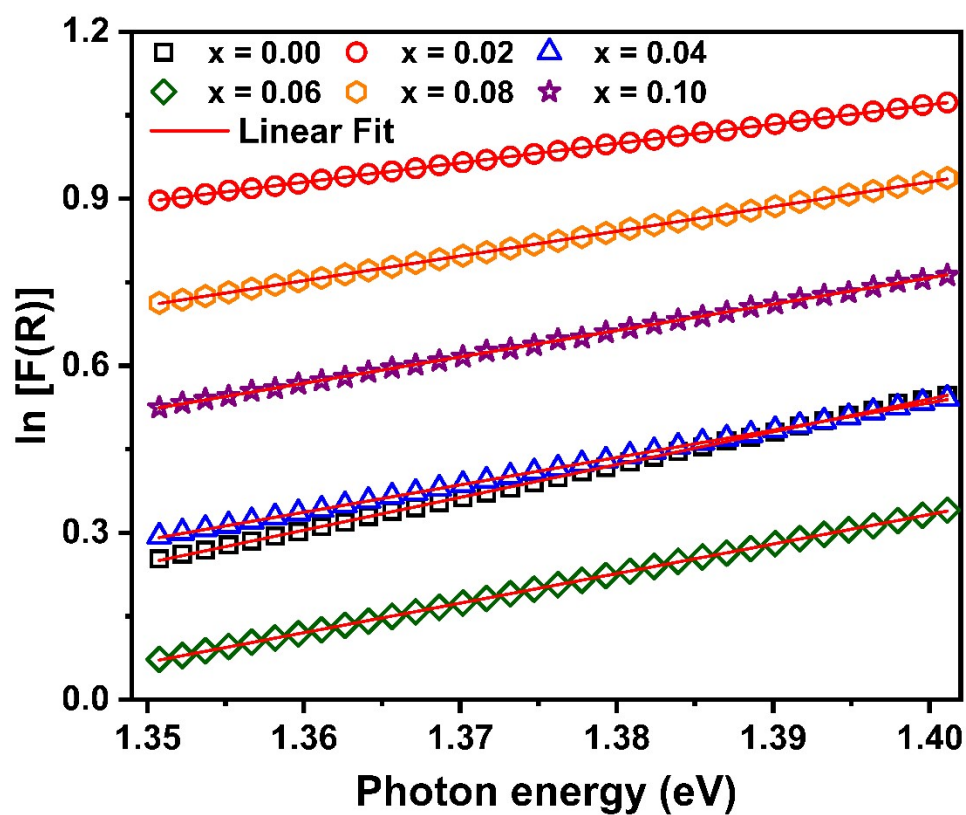


Figure S10. Estimation of Urbach energy of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06, 0.08$ and 0.10)

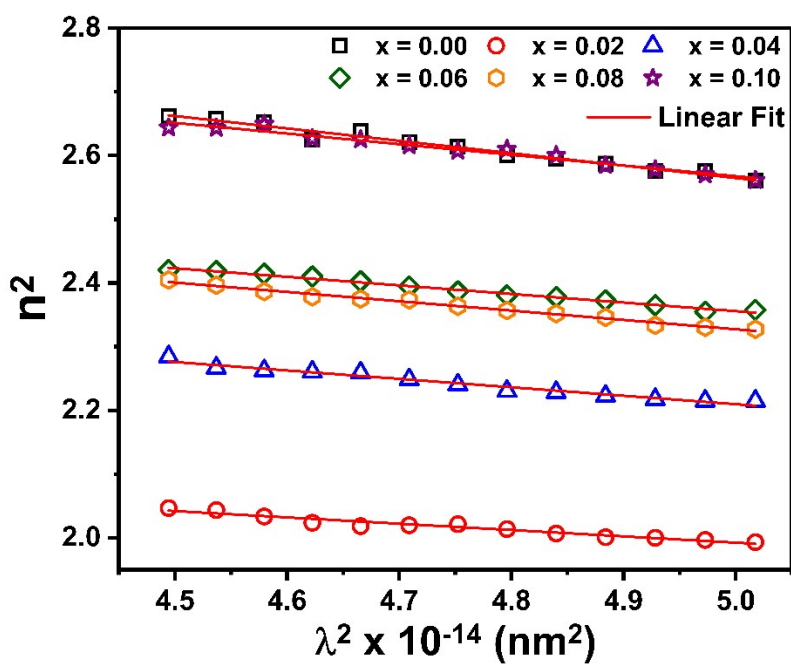


Figure S11. Estimation of ϵ_L and N/m^* of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06, 0.08$ and 0.10)

3.4 Dielectric and Impedance Formalism

3.4.1 Frequency-dependent Studies

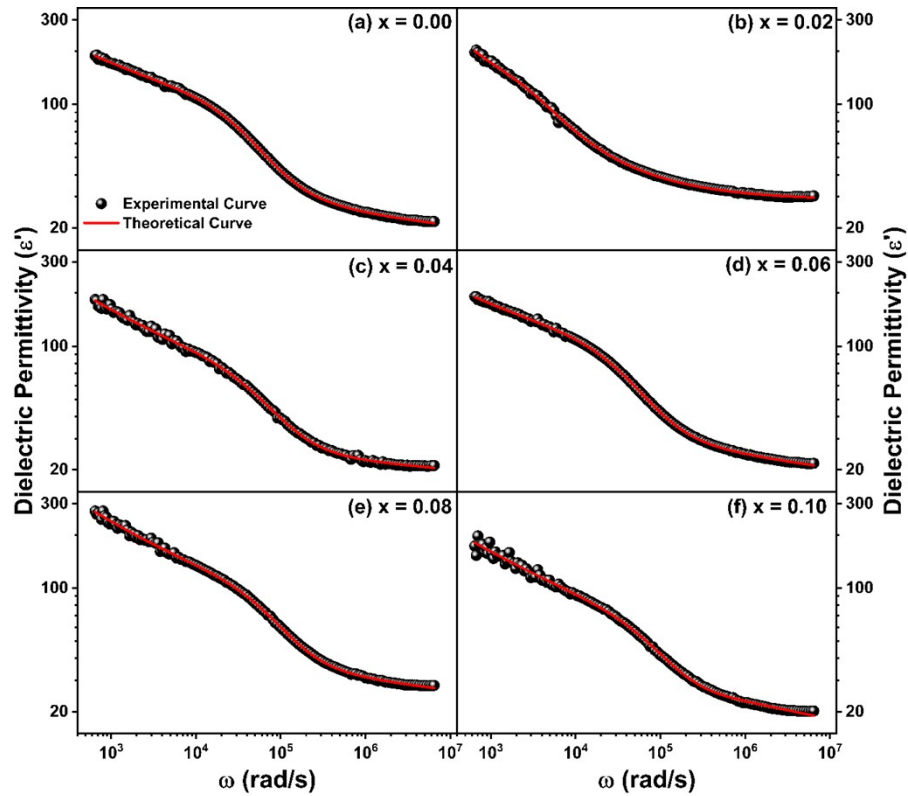


Figure S12. Theoretical fit using modified Havriliak Negami relaxation model associated with the conductivity of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$

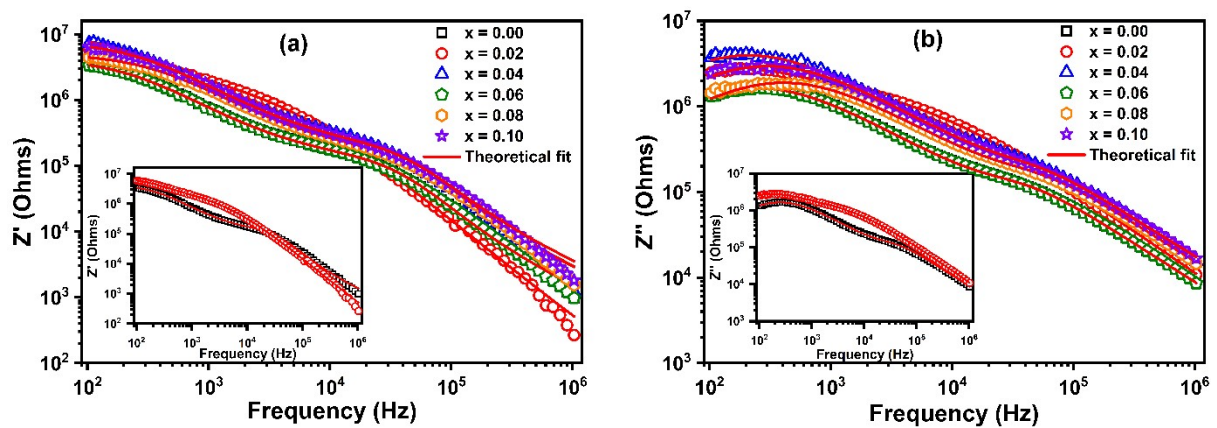


Figure S13. (a) variation of real impedance and (b) variations of the imaginary impedance of the $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

Table S4. Variations of resistance, capacitance, and n value of grain and grain boundaries in $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

x	R_g ($M\Omega$)	C_g (pF)	R_{gb} ($M\Omega$)	Q_{gb} (pF/ Ω)	n_{gb}	C_{gb} (pF)	Q_{il} (pF/ Ω)	n_{il}
0.00	0.11	38.47	4.39	552.44	0.82	147.3	-	-
0.02	1.62	378.90	13.68	13501.00	0.35	613.57	16.25	0.99
0.04	0.21	21.84	10.47	256.8	0.82	70.04	-	-
0.06	0.11	38.45	4.37	559.11	0.82	149.32	-	-
0.08	0.14	23.491	5.12	332.5	0.81	74.52	-	-
0.10	0.16	22.711	8.00	278.87	0.81	66.6	-	-

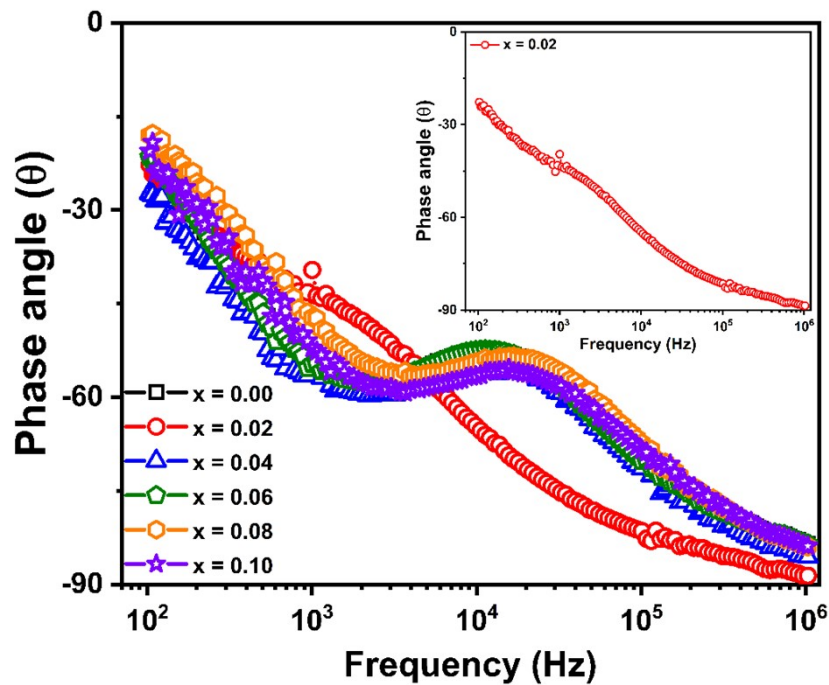


Figure S14. Phase angle variations of the $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

3.4.2 Temperature-dependent studies

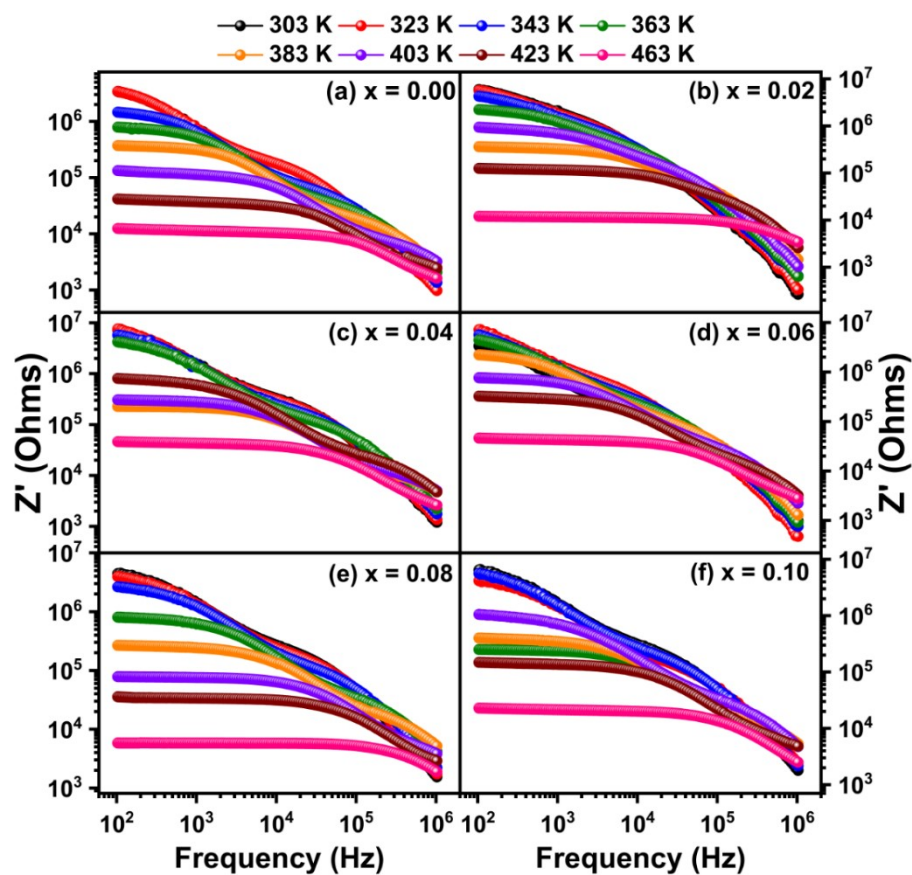


Figure S15. Temperature-dependent real impedance dispersion curve of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

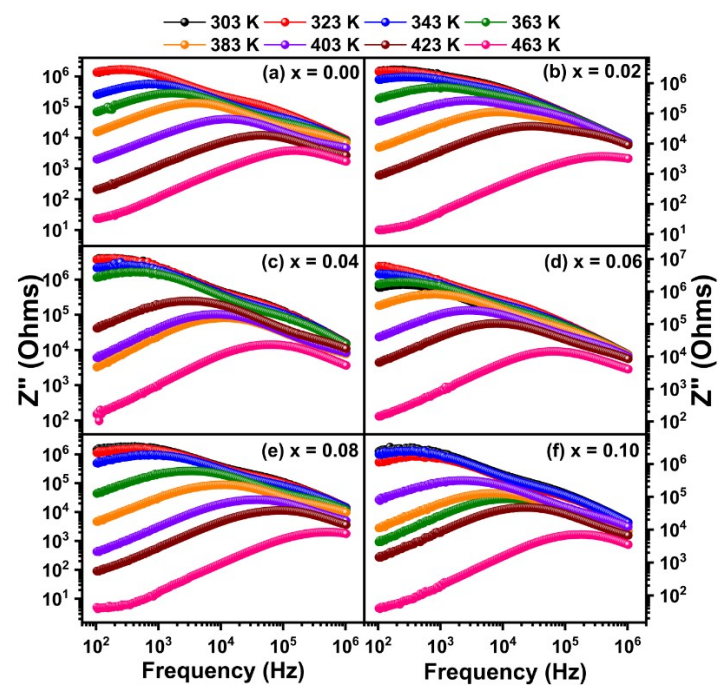


Figure S16. Temperature-dependent imaginary impedance dispersion curve of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

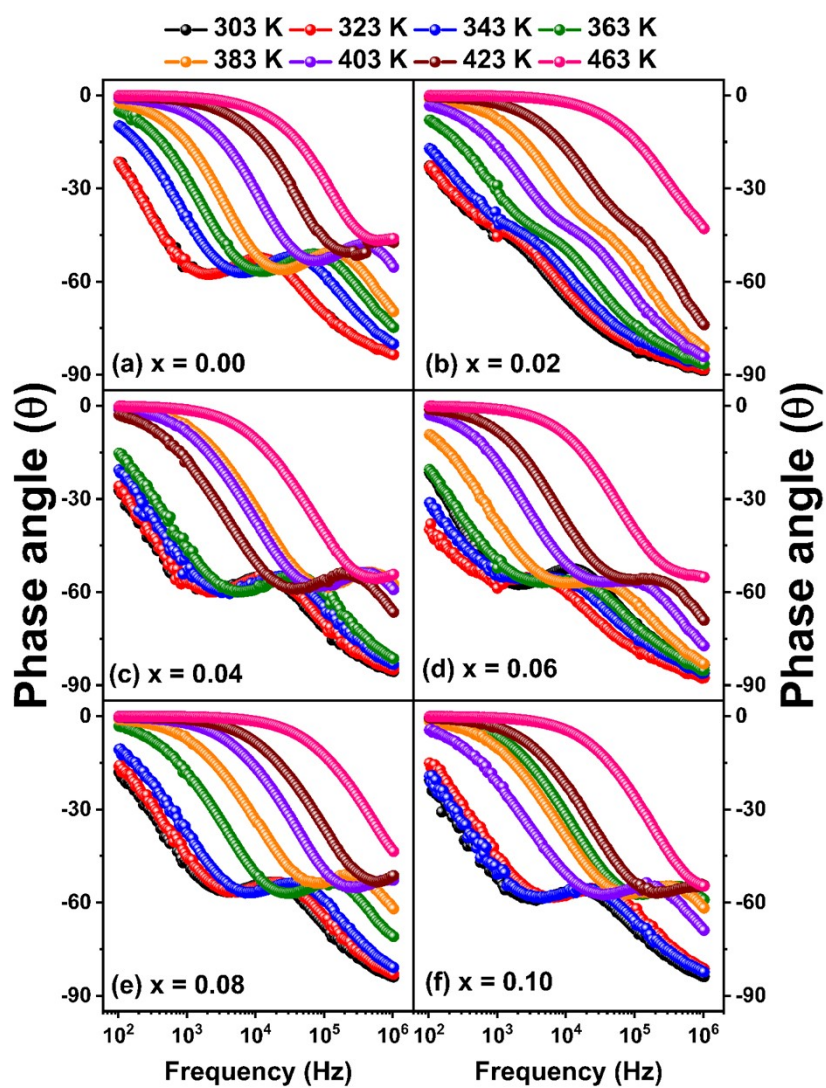


Figure S17. Temperature-dependent phase angle dispersion curve of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

3.5 A.C. conductivity studies

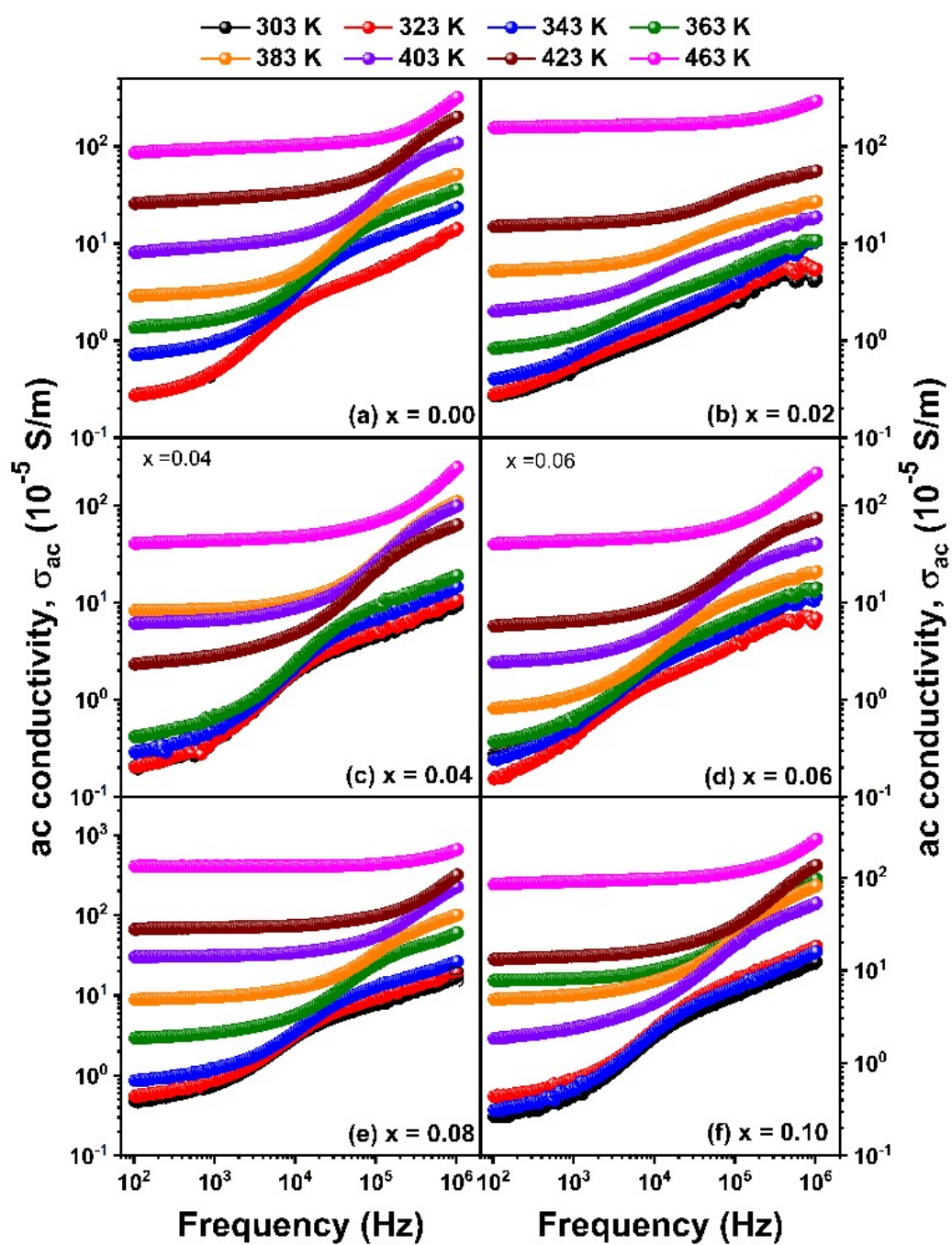


Figure S18. Temperature-dependent frequency response AC conductivity curve of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

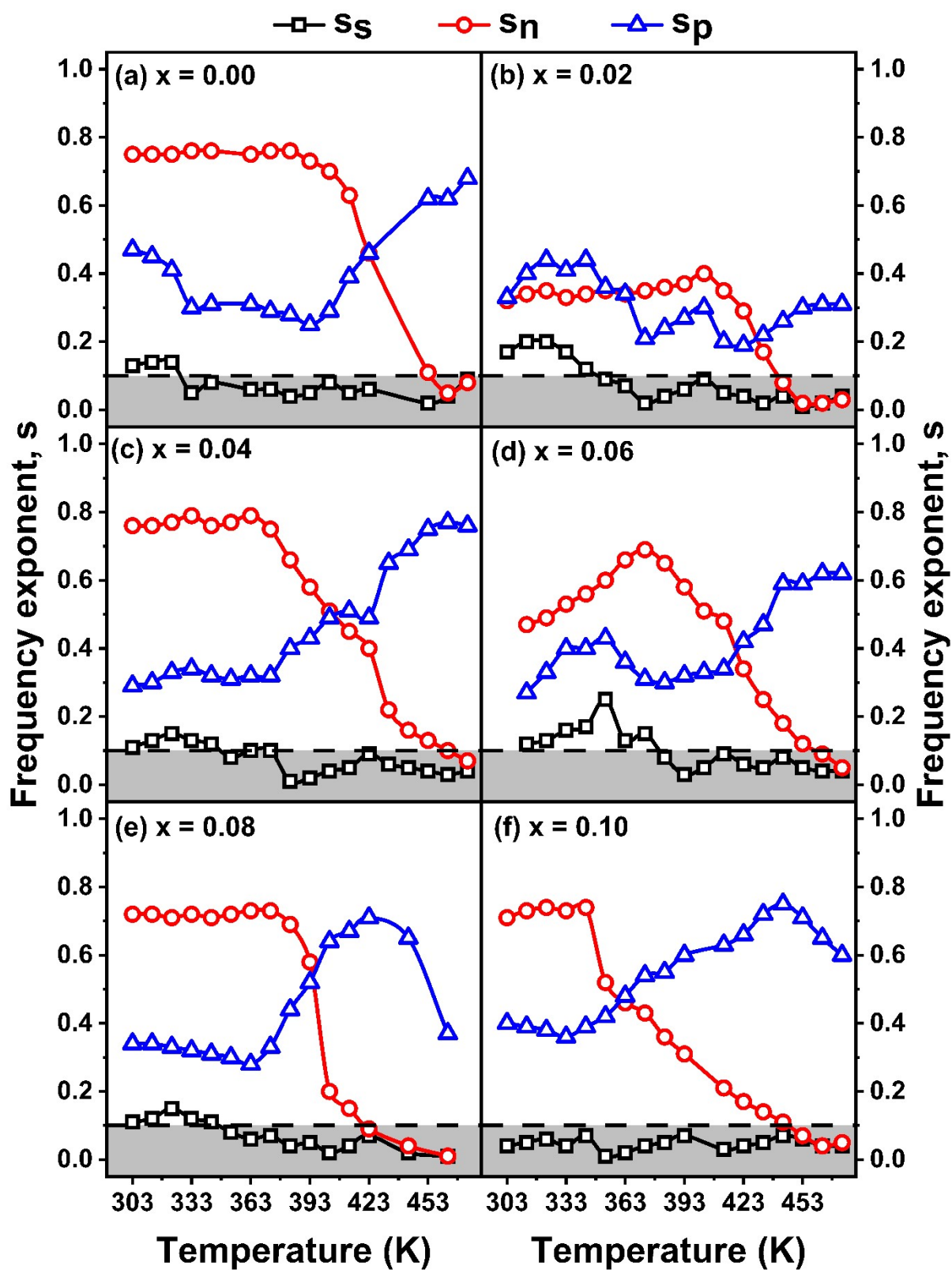


Figure S19. Variations of frequency exponent of different charge carriers of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system with respect to temperature

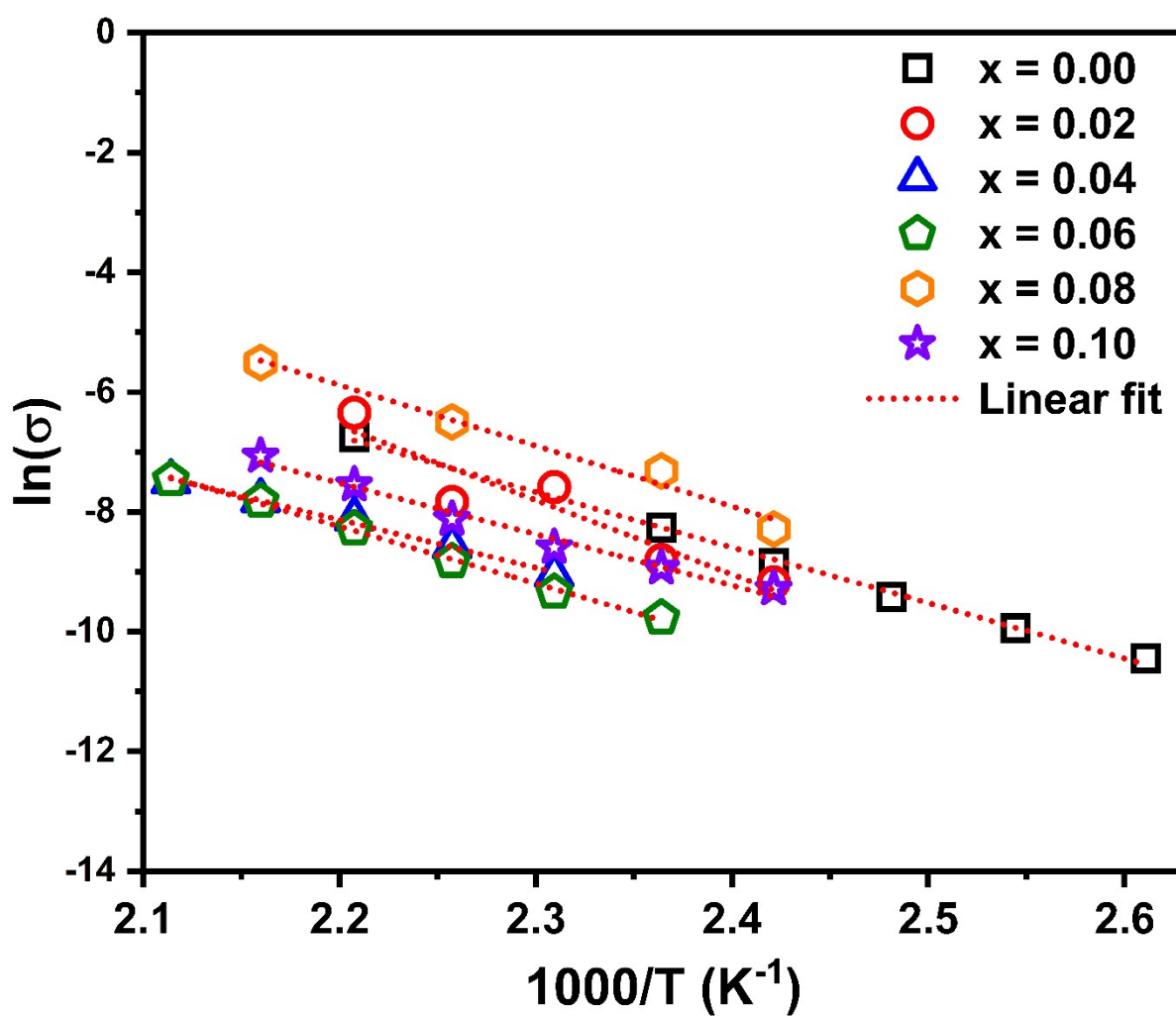


Figure S20. Estimation of activation energy (E_a) of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ ($x = 0.00, 0.02, 0.04, 0.06, 0.08$ and 0.10)

Table S5. Activation energy of $\text{CoFe}_{2-x}\text{Y}_x\text{O}_4$ system

Composition, x	Activation Energy (eV)
0.00	0.80
0.02	1.07
0.04	0.69
0.06	0.82
0.08	0.87
0.10	0.74