# Development of PVA/Chitosan based CuO Reinforced Polymer Composites with Enhanced Dielectric Performance for Energy Storage Applications

Rahmat Ullah, Tajamal Hussain*, Adnan Mujahid, Ahsan Sharif and Ejaz Ahmed
*School of Chemistry, University of the Punjab, Lahore-54590, Pakistan*
tajamalhussain.chem@pu.edu.pk*

(Received on 2nd September 2025, accepted in revised form 19th November 2025)

**Summary**: The increasing demand for energy in the past two decades intensified the development of efficient and sustainable energy storage systems. In this context, the use of polymer-based composites is a promising approach as dielectric materials for next generation capacitors as a quick and burst energy source. A blend of polyvinyl alcohol (PVA) and chitosan (CS) was used as the matrix, while CuO particles acted as fillers. This work reports, for the first time, the synergistic interaction between PVA and chitosan matrices with CuO nanoparticles for dielectric investigations for advanced energy storage applications. The successful formation of the composite was confirmed by UV–visible and FTIR analyses. SEM images revealed a textured surface due to CuO incorporation, while TGA demonstrated improved thermal stability, highlighting the reinforcing effect of CuO in the composite. The dielectric constant as a function of frequency has also been measured for a series of composites, with a maximum increase of 270 % in dielectric constant at 300Hz against a loading of 7 wt% CuO. The results of this study demonstrate that ternary composites are promising dielectric materials for use in efficient and flexible capacitors, offering a sustainable solution for the development of efficient energy storage systems.

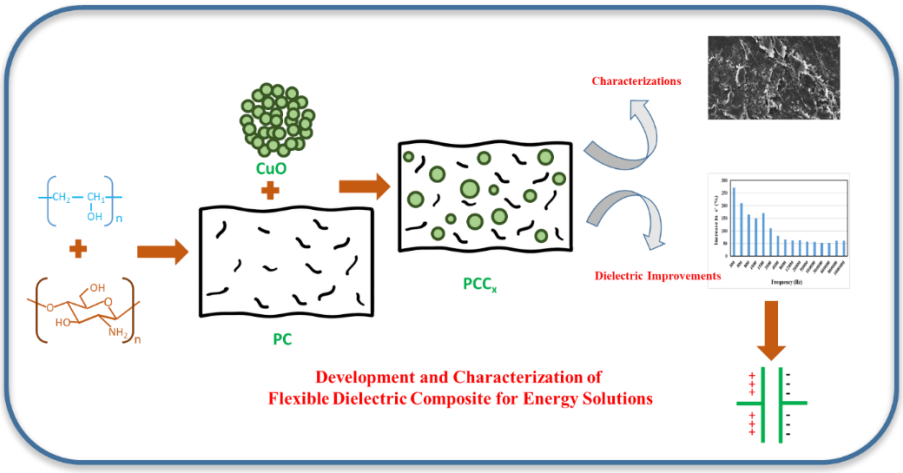


Fig. 1: Graphical Abstract

**Keywords:** Polymer nanocomposites, Flexible composites, Energy storage, Capacitors, Dielectric material.

## Introduction

The development of polymer composites has emerged as an important research area due to their capacity to improve the characteristics of traditional polymers. Polymer-based composites find applications across various fields, including biotechnology, nanoelectronics, actuators, supercapacitors [1] fuel cells, light emitting diodes (LEDs), biosensors, and even in mechanical domains such as radar systems and solar cells. Polymer nanocomposites are composed of fillers (nanoparticles) and polymers. The type of nanoparticles is crucial in determining the increase in optical, mechanical, thermal and electrical properties [3, 4]. The polymer's characteristics also influence the interfacial bonding between nanoparticles and polymer chains, where molecular bridges formed through such interactions strengthen the hybrid material. The consistent distribution of nanofillers across the polymer matrix is a challenging and crucial component in achieving optimal performance and broadening their range of applications [6].

Among various polymers, PVA is the most studied polymeric dielectric material due to its versatile properties, having strong dielectric values [7, 8] and good film-making ability [9]. Because of its non-toxic[10], biodegradable [11], and biocompatible qualities[12], this thermoplastic polymer is both

diverse and highly desired. However, its inherent limitations, including a relatively low dielectric constant and moderate thermal stability, restrict its broader applications. To overcome these challenges, blending PVA with natural biopolymers such as chitosan (CS) has emerged as an effective strategy to reduce costs, enhance biodegradability, and realize the profits of combined biopolymers [13, 14].

Considerations over the ecological issues caused by the disposal of synthetic polymers have led to an increase in the use of biological and sustainable natural polymers like CS [15]. CS has a variety of active groups that facilitate the synthesis of composites with unique properties and attributes. The primary and secondary –OH, amino, acetamide groups, and glycosidic bonds make it suitable for diverse sensing and electrochemical processes due to its high hydrophilicity and ability to be modified chemically and physically [16]. PVA is a suitable polymer to combine with CS to produce a blend (PC) that is both biocompatible and soluble in water [17]. Hydrogen bonding interactions are formed between them, promoting the development of physical crosslinks [18]. Nanoparticles used as fillers interact synergistically with the PC blend matrix, leading to enhanced overall properties.

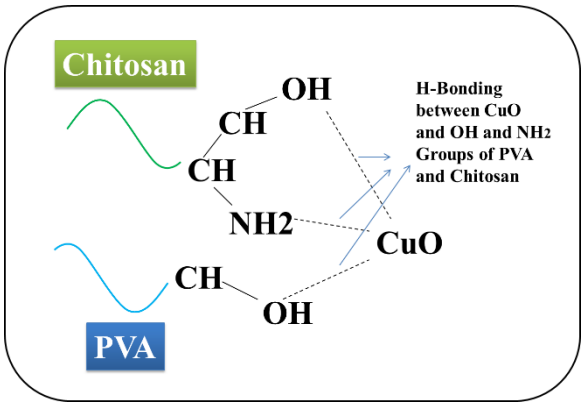


Fig. 2: Interaction between CuO nanoparticles and PVA & Chitosan polymer.

There is a variety of literature available discussing the relations between polymers like PVA, CS. Vinyl alcohol and co-vinyl acetate combine to form a biodegradable, physiologically inert copolymer known as PVA [19]. It dissolves well in water, is gas permeable, and can withstand severe tension[20]. PVA serves as an excellent polymer to combine with CS, producing a water-soluble, biocompatible material with strong mechanical rigidity [17]. Furthermore, nanofillers composed of dispersed nanoparticles are more efficient than micron-sized fillers at improving the characteristics of polymers [21]. Crystalline cupric oxide (CuO) nanoparticles (Nps) can be used in photovoltaic and photocatalytic applications due to their very narrow energy band gap, lower as compared to that of ZnO Nps [22, 23]. CuO has reinforced its importance as a significant technology material due to the physicochemical properties such as its semiconducting characteristics, great chemical stability, low toxicity, and chemical and physical stability, as part of the nanocomposites [24, 25]. Moreover, it has become a promising option for use in energy storage systems.

Customizing the characteristics of nanocomposites has been made possible by the addition of nanofillers to the polymer. Furthermore, the nanofillers enhance the dielectric characteristics of the polymer matrix by forming connections within the polymer chains. A significant interaction was observed between CS and CuO. PVA based composites with well-dispersed CuO were fabricated and showed improvements in terms of toughness, dielectric properties, mechanical properties, and optical properties [26]. Similarly, CuO particles were dispersed well in a system composed of PEO/CS blend and showed optimal properties change for their use in microelectronics [27]. Previous studies have reported similar improvements in nanocomposites reinforced with metal oxides such as $TiO_2$, $ZrO_2$ and ZnO, demonstrating increased electrochemical properties, dielectric constant and thermal stability [13, 28-30]. Here, it is expected that CuO particles will develop an association with the PC matrix through the existence of functional polar groups. Owing to their high surface area and excellent strength-to-weight ratio, the incorporation of CuO particles into the PC matrix is expected to significantly enhance both the surface morphology and electrical properties of the composites. In short, the ultimate purpose of this study is to improve the dielectric characteristics of PC by introducing CuO particles with different amounts into the matrix, resulting in $PCC_x$ composites ($PPC_1$ with 1wt% of CuO, $PCC_3$ with 3wt% of CuO, $PCC_5$ with 5wt% of CuO, and $PCC_7$ with 7wt% of CuO. The addition of CuO in PC blend aims at developing flexible composites with improved functionality of the PC, particularly, dielectric properties, for their application as dielectric materials in capacitors. In this study, a novel combination of PVA and chitosan matrices incorporated with CuO particles is explored to develop high-performance dielectric composites. The unique combination shows strong synergistic effects, resulting in improved dielectric behavior for energy storage.

## Experimental

### *Materials and Chemicals*

PVA Powder (M.W=14000 g/mol approx.) was bought from SAMCHUN. Chitosan, a natural biopolymer (M.W=1526 g/mol) was used, and Copper sulphate pentahydrate (M.W=249 g/mol) was bought from Sigma Aldrich. Other reagents, sodium hydroxide and acetic acid, were also purchased from Sigma Aldrich. Distilled water was used to dissolve PVA and prepared CuO particles separately, and was also employed as a solvent for film preparation. All the chemicals have been used as received without any further purification steps.

### *Instrumentation*

Fourier Transform Infrared (FTIR) spectrometer (Cary 630) of Agilent Tech. and Ultraviolet/Visible (UV/VIS) spectrometer ($T90^{+}$) of PG Instrument Ltd. are used to monitor the synthesis of composites. Thermogravimetric analyser (TGA-50H) was used to study the thermal behavior of the composite film and samples were heated from room temperature to 650 °C at the heating rate of 5°C/min. FEI Inspect 50 scanning electron microscope (SEM) with Everhart-Thornley (ET) detector was employed to study the morphology and particle distribution. The LCR Meter (E4980AL) of Keysight Ltd was used to study the resistance and capacitance of the composite film at various frequencies. The frequency range used was from 300 Hz to 1 MHz throughout the experiment.

### *Fabrication of CuO Particles*

The production of the CuO particles was done using the precipitation technique. 0.5g of $CuSO_4.5H_2O$ was first dissolved in distilled water. After continuously stirring the copper sulphate solution at 60 °C, NaOH solution was added drop by drop until the pH 11 was achieved. Brownish black precipitates of copper hydroxide began to form, and the solution was kept on stirring at a temperature of 60 °C for 12 hours to ensure the complete conversion of $CuSO_4.5H_2O$ to copper hydroxide. After filtration and washing twice with distilled water, the synthesized copper hydroxide particles were dried in an oven at 80 °C for 12 hours. Afterwards, dried particles were calcined for four hours at 400 °C in a muffle furnace to convert copper hydroxide to copper oxide particles. These were subsequently used in composite formation.

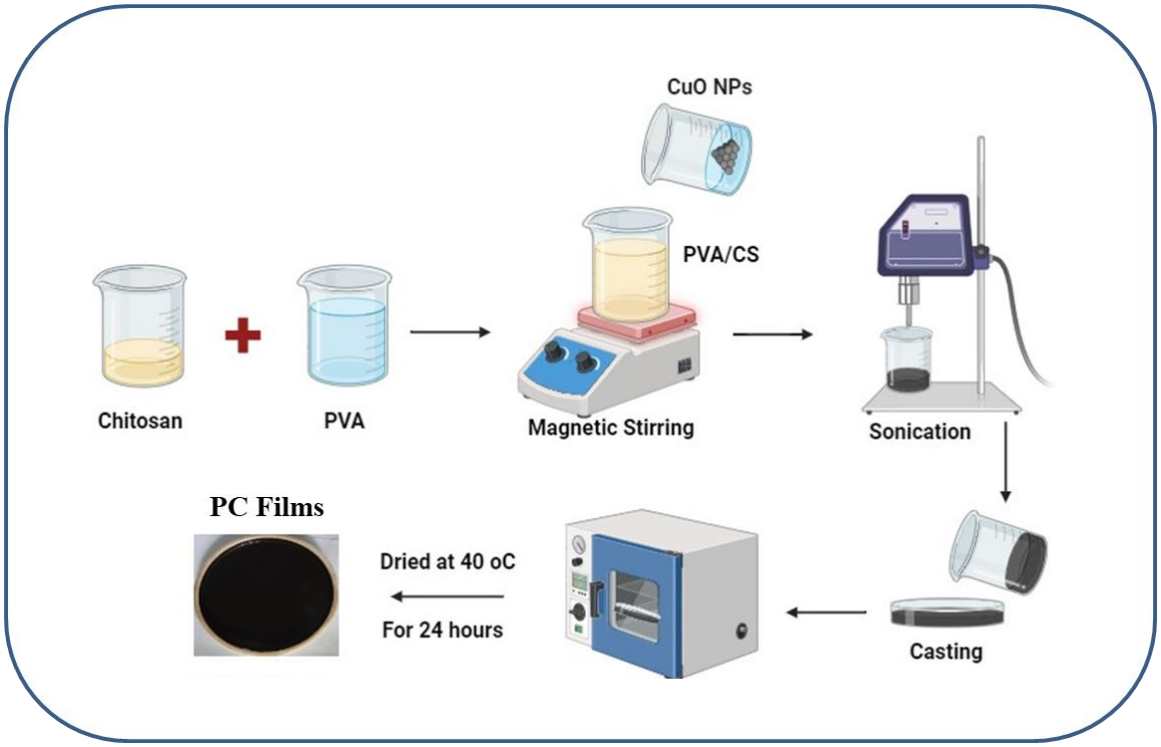


Fig 3: Schematic diagram for PC synthesis.

*Film Fabrication Process*

PVA based ternary composite film was prepared by the solution casting method. In this study, CS was employed as a biopolymer. PVA serves as a water-soluble synthetic polymer in the matrix. The biodegradable nature of PVA and CS was induced by CS, which is a distinguishing attribute of this substance. Four composite films were developed: $PCC_1$, $PCC_3$, $PCC_5$, and $PCC_7$. One film (PC) is formed without adding CuO particles using a blend of PVA and CS. $PCC_1$, $PCC_3$, $PCC_5$, and $PCC_7$ were produced by adding 1, 3, 5, and 7 wt. % CuO particles into the PC blend matrix, respectively. These films were further used for different characterizations. The whole experimental process is illustrated in Fig 3.

## Results and Discussion

*UV-Visible and FTIR Spectroscopic study of molecular interactions*

The plot of optical absorption versus wavelength for PC, $PCC_1$, $PCC_5$, and $PCC_7$ flexible composite thin films is shown in Fig. 4. A single sharp absorption peak is reflected in the spectrum of PC without any noticeable shoulder peak, which can be attributed to two possible reasons. First, the high miscibility of PVA and CS results in a PC behaving as a single system and which is indicated in UV-visible as a single peak. Second, while pure PVA typically exhibits a maximum absorption near 220nm and chitosan in a region of 300nm, the absorption maximum of the PC blend appears at near 215nm. This shift is attributed to the dominant contribution of PVA (80%) relative to CS in the blend. In the UV-Visible spectra of $PCC_1$, $PCC_5$, and $PCC_7$, an absorption maximum is observed at the same position, i.e., 215 nm; however, an extended strong absorption is observed up to 400nm, accompanied by a distinct shoulder peak near 368nm, which belongs to CuO.

Table-1: Summary of amounts used to develop polymer blend and composites.

| Sr. No | PVA (g) | CS (g) | CuO NPs (g) | Amount of CuO (%) |
|---|---|---|---|---|
| 1 | 0.8 | 0.2 | 0 | 0 |
| 2 | 0.79 | 0.2 | 0.01 | 1 |
| 3 | 0.77 | 0.2 | 0.03 | 3 |
| 4 | 0.75 | 0.2 | 0.05 | 5 |
| 5 | 0.73 | 0.2 | 0.07 | 7 |

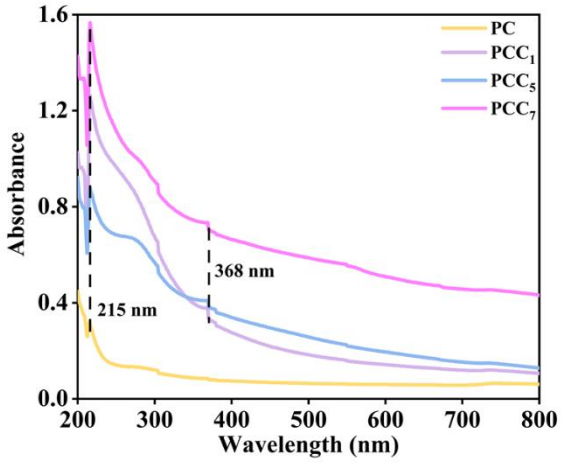


Fig 4: UV-Visible Spectra of PC and PCCx; ( x=1, 5 and 7).

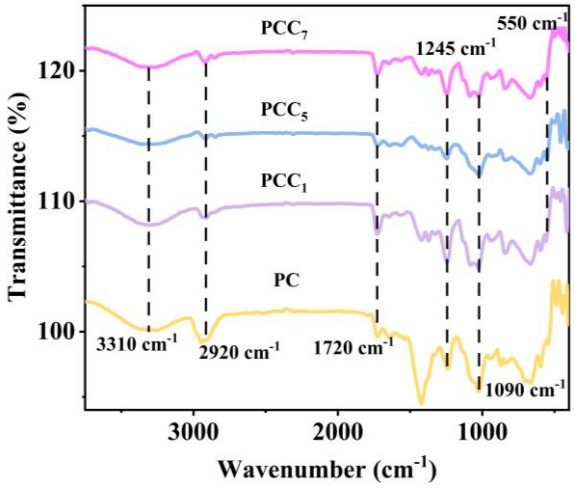


Fig. 5: FTIR Spectra of PC and PCCx; (x=1, 5 & 7).

FTIR is used to examine the functional groups and chemical structure of the synthesized composites. Fig 5 displays the FTIR spectra of PC and $PCC_x$ composites. The large peaks for C–H stretching vibrations occurred at 2920 $cm^{-1}$ and a broad peak near 3310 $cm^{-1}$ belongs to –O–H stretching vibrations for PC and composites[31-33]. The stretching C=O peak is associated with 1720 $cm^{-1}$. The peak at approximately 1400 $cm^{-1}$ corresponds to $–CH_3$ wagging, and the small band at 1105 $cm^{-1}$ is due to the anti-symmetric stretching of C–O–C present in CS[32]. CuO exhibits a stretching peak around 400-600$cm^{-1}$ and in this region, PVA also exhibits peak [34]. Change in shape of peak in UV-Visible spectra and change in intensity of the peak observed in FTIR

spectra for $PCC_x$ as compared to PC, indicate the successful development of the composites.

*Thermal Stability Analysis of PC and $PCC_x$*

The thermal degradation behavior of PC blend and $PCC_x$ composites was examined by thermogravimetric analysis (TGA), as shown in Fig 6. All samples exhibited multistep weight loss patterns, characteristic of polymeric systems. The initial weight loss (almost 6.14 %) below ~150 °C can be attributed to the evaporation of physically adsorbed moisture and volatile components. The major decomposition stage, occurring between ~250 °C and 450 °C, corresponds to the thermal degradation of the polymer backbone (deacetylation of chitosan and chain scission of PVA). A final and maximum weight loss (26.84 %) observed above 450 °C is attributed to the further breakdown of char residues and the complete degradation of the polymer matrix. For $PCC_7$, the residual waste was 10 % and for PC and $PCC_5$, it was 1% and 5%. Hence, in contrast to PC, both $PCC_5$ and $PCC_7$ composites displayed slightly higher thermal stability, as evidenced by the delayed onset of decomposition and higher residual weight at elevated temperatures. The improvement becomes more pronounced with 7% CuO loading, indicating that CuO nanoparticles act as effective thermal stabilizers by restricting polymer chain mobility and enhancing the barrier effect against volatile degradation products. The higher char yield in $PCC_x$ further confirms the protective role of the inorganic filler. These observations confirmed that the incorporation of CuO particles effectively integrated within the polymer matrix, enhancing its overall thermal stability. Thus, incorporation of CuO nanoparticles into the PC matrix enhances the thermal stability and flame resistance ability of the composites, making them more suitable for high-temperature applications and reliable energy storage devices. Similar thermogravimetric behaviours were also observed in the reported metal oxide-PVA based nanocomposite films [35, 36].

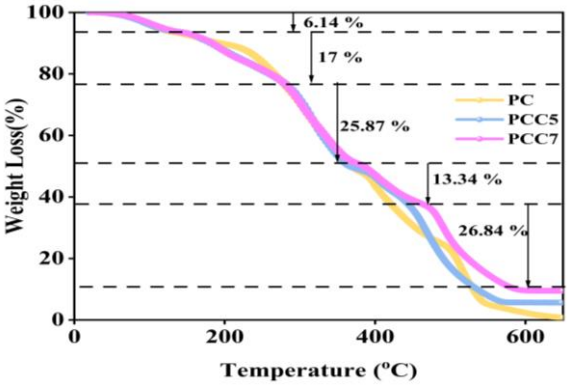


Fig. 6: TGA profile of PC and $PCC_5$ and $PCC_7$

The surface morphology of the pristine PC blend and its CuO-incorporated composites was examined using SEM images given in Fig 7. As shown in SEM image, the PC blend exhibits a relatively smooth and planar surface, indicating good miscibility between the two polymers and the absence of significant phase separation[37]. In contrast, the $PCC_7$ composite displays a uniform, rougher and more textured morphology[38]. The observed uniform roughness can be attributed to the homogeneous dispersion of CuO nanoparticles within the PC blend, which introduces interfacial interactions.

*Surface Morphological Analysis by SEM images*

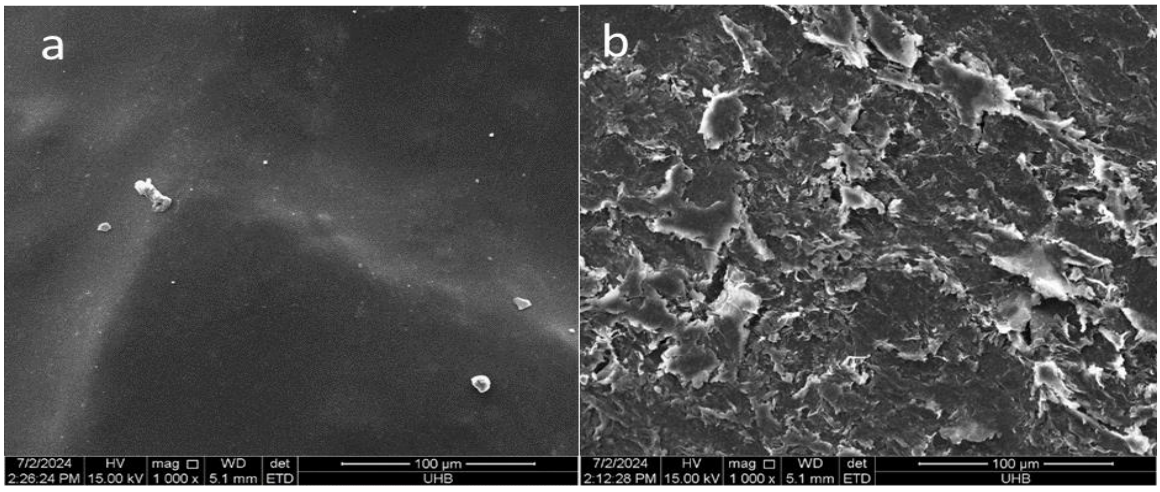


Fig. **7:** SEM images) of PC (a) and $PCC_7$ (b).

This morphological transformation is particularly significant for dielectric applications. The rough and textured composite surface suggests improved interfacial polarization, which can contribute to a higher dielectric constant and stability under an applied electric field. These features highlight the potential of composite as a promising dielectric material for capacitor applications.

*Dielectric and Conductive Behaviour*

Dielectric constant (ε') of prepared composites as a function of CuO loading measured at a series of frequencies is shown in Fig 8. The dielectric constant of PCCx composites showed negligible variation at 1 wt% to 5 wt% CuO loading across frequencies (300 Hz – 1 MHz), indicating insufficient filler connectivity to alter polarization. However, at 7 wt% CuO—the percolation threshold—a sharp enhancement in ε' occurs. The enhancement of dielectric properties primarily results from interfacial (Maxwell–Wagner–Sillars) polarization. Conductive CuO particles provide additional charge carriers and increase interfacial polarization, raising the dielectric constant. This effect was highly dependent on frequency: a bar graph, showing the increase relative to PC, displayed a 270% rise in ε' at 300 Hz, which gradually decreased to just 60% at 1 MHz. Similar trends of dielectric constant have been observed in the literature [39, 40]. This attenuation aligns with the slow kinetics of interfacial charge migration, which cannot follow the rapidly oscillating field at higher frequencies. This substantial improvement in the dielectric constant value for the prepared composites is quite encouraging.

It has been reported in the literature that CuO has been incorporated into PVC, and the maximum increase in dielectric constant observed was just 18% on the incorporation of 15% filler. In another reported work, CuO nanoparticles have been used to prepare a composite by incorporating them into a PVDF matrix; a 30-35% increase has been observed at low frequency upon incorporation of 10% metallic Cu [41]. A large amount of addition of metallic oxide or metal particles results in a compromise in the flexibility, density, and ease of processing of the materials. In a recently reported research work, a more than 300% increase in dielectric constant has been observed upon the incorporation of $TiO_2$/Ag hybrid nanofiller in natural rubber. Although, exact amount of the hybrid filler is not specified, this improvement is attributed to the synergistic effect of the dielectric polarization modulated by the highly conductive nature of Ag nanoparticles [42].

Results of the percentage increase in electrical conductivity and dielectric constant for composites prepared with maximum filler loading are displayed in Fig 9(a-b). Simultaneously, electrical conductivity (σ) exhibited an inverse trend: the largest enhancement occurred at 1 MHz, while the smallest increase was observed at 300 Hz. This complementary behavior confirms that at low frequencies, energy is dominantly stored via interfacial polarization (high ε'), whereas at high frequencies, the interconnected CuO network facilitates charge transport (high σ), reducing the relative contribution of capacitive storage. The coexistence of giant low-frequency ε' and high-frequency σ at 7 wt% underscores the dual role of percolative filler networks in dictating the dynamic electrical response.

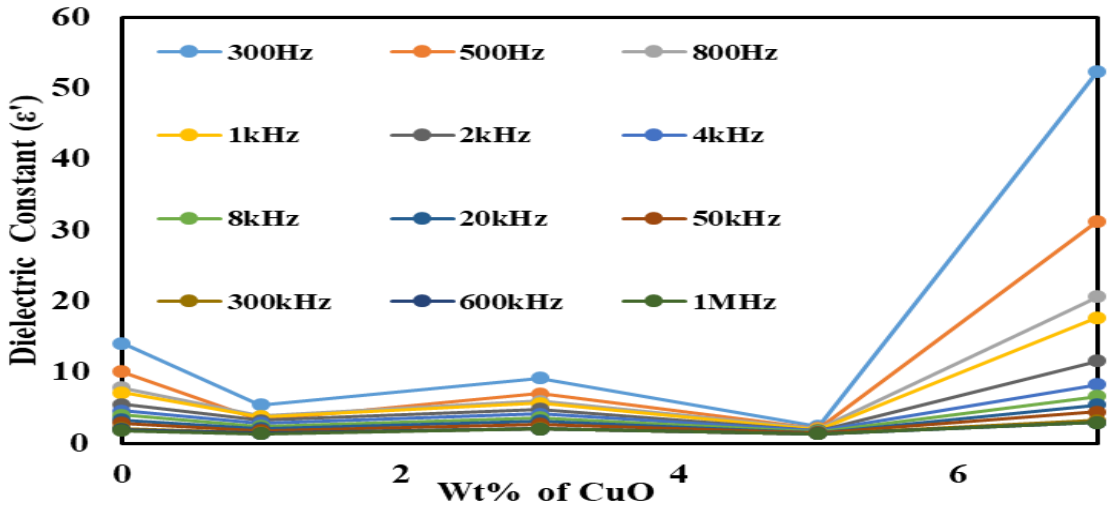


Fig. 8: Dielectric constant as a function of CuO loading (%) measured at a range of frequencies (300Hz to 1MHz).

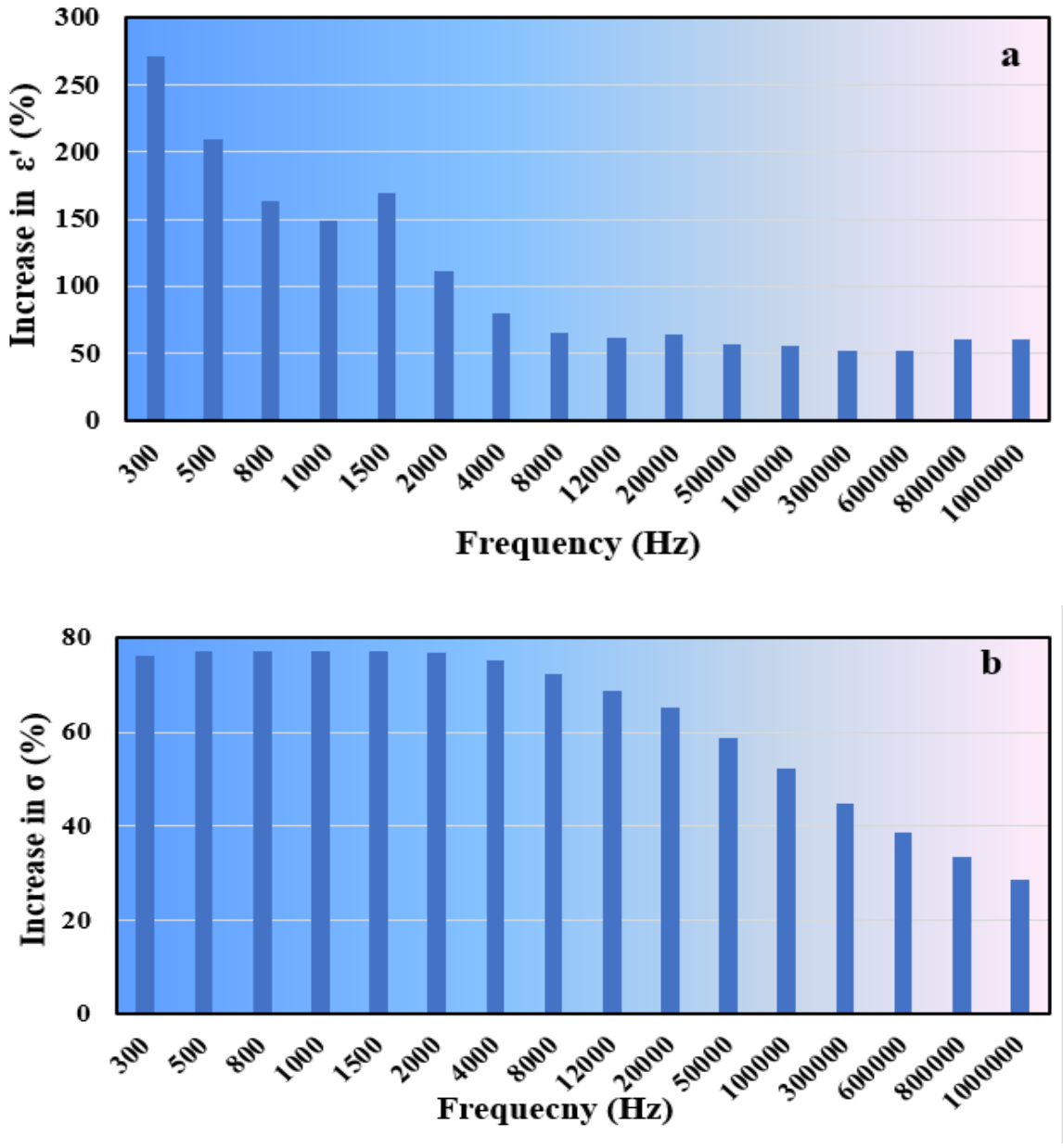


Fig. 9: Percentage increase in Dielectric constant (a) and Conductivity (b) observed at different frequencies for $PCC_7$.

**Conclusion**

In the current research work, PVA based composites were successfully fabricated utilizing CuO and CS by a simple and sustainable solution mixing procedure. The synergistic effect of the blend of PVA and CS serves the purpose of developing a robust and compatible matrix with improved features, while the uniform dispersion of CuO nanoparticles markedly improved the dielectric, thermal, and structural characteristics of the composite. Results of the structural analysis performed using FTIR and UV-Visible spectroscopy confirmed the successful synthesis of the blend of PVA and CS and composites. TGA analysis confirmed that the incorporation of CuO particles effectively integrated within the polymer matrix, enhancing its overall thermal stability. Among all samples, PCC7 exhibited the highest enhancement—showing a 270% increase in dielectric constant at 300 Hz —demonstrating the strong interfacial polarization and efficient charge transport introduced by CuO incorporation. This unique ternary

design highlights the potential of combining bio-derived polymers with metal oxides to achieve high-performance dielectric materials. Overall, the present work provides valuable insight into tailoring polymer–filler interfaces for the development of flexible, efficient, and environmentally benign dielectric composites suitable for next-generation capacitors.

**Acknowledgement**

This research work is fully funded by the University of the Punjab, Lahore, through its Annual Research Project 2024-2025. Tajamal Hussain gratefully acknowledges the Office of Research, Innovation and Commercialization (ORIC), University of the Punjab, Lahore, for facilitating this project. Tajamal Hussain would also like to thank Dr. Amir Habib (University of Hafr Al Batin, Hafr Al Batin, KSA) for his assistance in conducting SEM and TGA analysis.

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