

ORIGINAL RESEARCH ARTICLE

Study of the effect of doped methyl blue and methylene blue dyes on the Optical and structural properties of (PMMA-CuO) thin films

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ABSTRACT

The two laser dyes used were methyl blue and methylene blue, which exhibited favourable optical properties. The linear optical properties were enhanced by incorporating methyl blue or methylene blue dyes into the copper oxide nanoparticles added to polymethyl methacrylate dissolved in Acetone, improving the optical properties important for future applications in laser materials. The prepared samples were examined by a UV-Vis spectrophotometer at wavelengths ranging from 200 to 800 nm. The results showed that the incorporation of methylene blue dye into the (CuO-PMMA) nanocomposite thin films significantly increased some optical properties, including absorbance, absorption coefficient, extinction coefficient, and refractive index. Meanwhile, the transmittance and energy gap decreased with increasing dye concentration. The addition of methyl blue dye had less effect on these properties and yielded values close to those of the nanocomposites that did not contain the dye. The structural properties of the nanocomposites were studied. The structural properties included Field emission scanning electron microscopy (FE-SEM) images and XRD analysis. The SEM images showed the distribution of copper oxide nanoparticles within the PMMA polymer and demonstrated the diffusion of laser dyes within the nanocomposites. Methyl blue dye forms rough, irregular aggregates on the surface, creates darker areas and reduces solubility. In contrast, methylene blue forms a uniform thin layer that covers the nanoparticles and enhances surface contact through hydrogen bonds, which masks the material crystallinity and affects its mechanical and optical properties.

Keywords: (PMMA-CuO) thin films; Laser dyes; UV-Vis spectroscopy; optical properties; Field emission scanning electron microscopy (FE-SEM)

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1. Introduction

Copper oxide (CuO) nanoparticles-based thin films have garnered significant attention in recent materials science due to their unique linear optical properties, including tunable absorption ^[1], refractive index modulation, and promising applications in optoelectronics and optoelectric devices. Optical properties of CuO thin film are primarily determined by its particle size, shape, and the dielectric environment ^[2], and it is thus regarded as a potential candidate for photonic coatings, sensors, and solar cells ^[1].

At the same time, the laser dyes such as methylene blue (MB) and methyl blue (MeB) exhibit strong absorption and fluorescence in the visible region, which makes them attractive for respective dye lasers, bioimaging, and photodynamic therapies ^[2].

A dye laser is a laser that uses an organic dye as the lasing medium, usually as a liquid solution, compared to other alternatives (gases and

bacrystals). Organic dyes present an absorption band wider than in the ultraviolet region, in the visible spectral range of the electromagnetic radiation. This characteristic is possessed only by organic compounds having an extensive system of alternating tissues of conjugates of single and double bonds. In a dye laser, the dyes are either solubilized in an organic solvent or a crystalline form. It has been demonstrated that dyes can act as lasers in solid, liquid, and gas media; however, in liquid and solid forms, dyes have had their most significant impact as a laser medium^[3].

These dyes doped in a poly(methyl methacrylate) (PMMA) polymer matrix possess enhanced photo stability, controlled dispersion, and tailored optical behaviours ^[4].

Due to the high optical transparency, mechanical rigidity, and its compatibility with organic dyes, PMMA is a good host material ^[5].

The linear optical properties (absorbance, transmittance, and refractive index) of CuO NP thin films and the spectral characteristics of MB and MeB doped in PMMA(Poly(methyl methacrylate)) are analyzed in this work. The combination of plasmatic nanoparticles (CuO) and organic dyes in a polymer makes them unique materials for advanced optical materials, optical sensors, and light-harvesting media ^[6].

2. Experimental work

Copper oxide (CuO) nanoparticles (0.01 g) and poly(methyl methacrylate) (1 g, PMMA) are added to Acetone. dyes are methylene blue 10^{-4} M, methyl blue 10^{-4} M, Solvent is Acetone(soluble with PMMA).Ultrasonic disperser for approximately 15 minutes to ensure the homogeneity of the material and Magnetic stirrer. Dispersion of CuO Nanoparticles: CuO nanoparticles (0.01 g) are weighed and dispersed in 30 mL of solvent (Acetone). Exposing 1-2 hours to form discrete agglomerates and to promote uniform dispersion. PMMA Dissolution: In a separate beaker, dissolve PMMA in the same solvent (0.5g of PMMA in 10mL of Acetone). The stirring is continued for 1–2 hours at 40–50°C until finally all the PMMA has dissolved. Doping of CuO in PMMA: The precipitated CuO particles are slowly added to the PMMA solution under stirring.

Stir for 2 to 3 hours to ensure uniformity. Dye use: 10^{-4} M solutions of methylene blue and methyl blue were prepared in the same solvent. Add the dye mixtures stepwise to the CuO/PMMA mixture. After the first hour, stir for another hour to help distribute evenly.

Film Casting: Transfer the final mixture to a clean glass Petri dish. Pour the solution slowly at room temperature (Casting) to form a film.

3. Theory

Linear optical properties are properties that are related to the interaction of materials with light at low intensities. Examples of such properties include refractive index, absorption, and reflection. Refractive index, absorption, and reflection are proportional to the electric field of the light. The user is likely involved in optics or materials science^[7,8].

Copper oxide massif comprises semiconductor thin layers. The metal has a band gap of approximately 2.0-2.2 eV; it can absorb visible light the former property results in a reddish view of CuO massif. The latter property gives CuO metal a smaller bandgap of about 1.2-1.5 eV. This allows the metal to absorb near infrared and red light, which in turn makes it look black or grey. Moreover, their optical properties depend significantly on the differing methods of fabrication^[9].

PMMA is a transparent polymer that is widely used in optics. The material an excellent transparency in the visible spectral range. PMMA also has a small refractive index, approximately 1.49, and it barely absorbs.

Such material is often used as a substrate or a medium for other substances. This way, the dyes can be inserted into the PMMA matrix or can be attached to the glass wafer^[10].

Methylene blue and methyl blue are organic laser dyes, with a cationic structure. For instance, the former absorbs at around 664 nm, and it emits a deep red/NIR^[11].

Methyl blue has a slightly relative absorption (~650-680 nm). Both dyes high foundations' molar extinction coefficients. The optical properties of the two dyes mainly depend on their concentration. Some applications are Copper Oxide Solar Absorbers, Photo detectors, Sensors, PMMA Optical waveguides, Micro lenses, Laser substrates, PMMA dyes, Solid-State dye lasers, and filters[12,13].

4. Results and discussion

4.1. Optical properties

4.1.1. Absorbance (A)

The absorbance spectra of PMMA, CuO, and the dyes are distinct and shown in the UV less 400 nm region. Besides these combinations, relatively higher absorbance was observed for PMMA, CuO, and methylene blue (MB) at 350 nm as range. Absorbance of MeB at 350 nm. The results for (PMMA, CuO, and MeB),(PMMA, CuO, and MB)and(PMMA, CuO,) were 0.960, 0.955, and 0.945, respectively as Figure 1. Optical studies revealed that the addition of dyes causes significant changes in the absorbance properties of PMMA/CuO composites, resulting in intermediate absorbance. Principal Insights are high absorbance at 350 nm spectrum is associated with electronic transitions in CuO nanoparticles and the $\pi \rightarrow \pi^*$ or $n \rightarrow \pi^*$ transitions in the dye molecules^[8].

The structures of the different dyes indicate that methylene blue (MB) is expected to have a more substantial UV absorbance than methyl blue (MeB), due to the more extensive conjugation in the light absorption mechanism^[9].

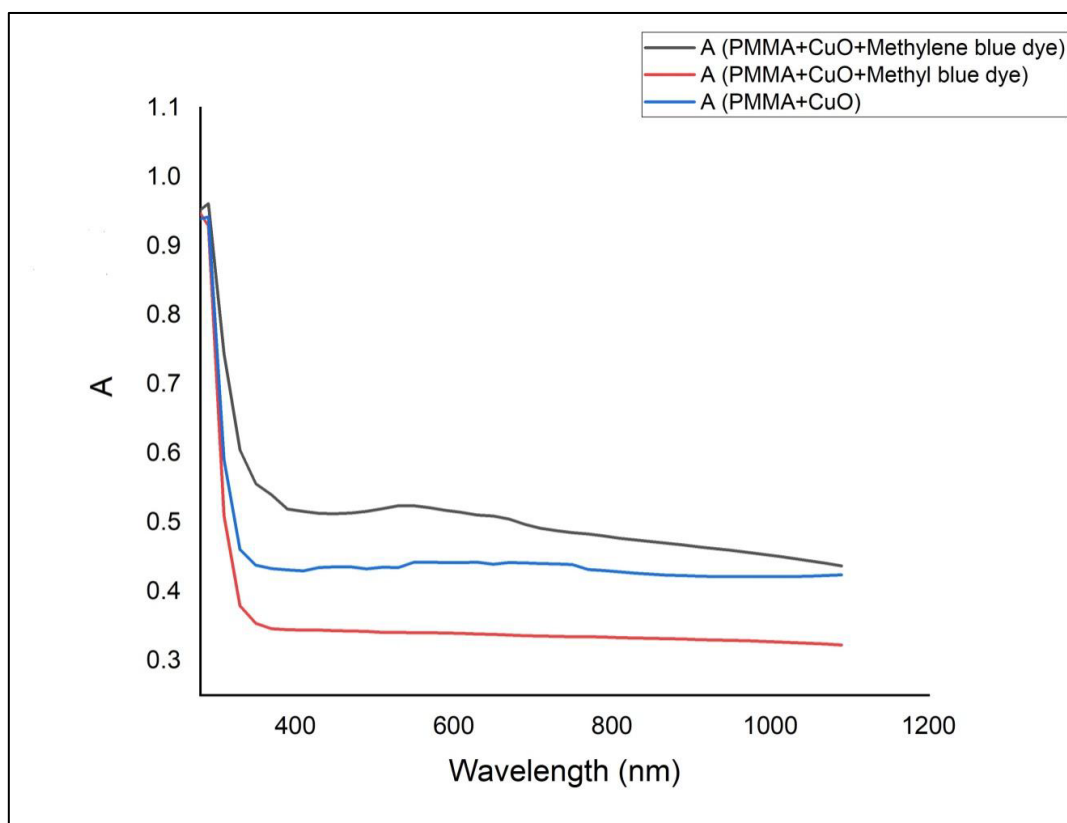


Figure 1. Absorbance between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

Table 1. Absorbance Peaks

Sample	Peak Absorbance (A)	Wavelength (nm)
PMMA/CuO/Methylene Blue	0.955	350
PMMA/CuO/Methyl Blue	0.960	350
PMMA/CuO (Control)	0.945	350

4.1.2. The transmittance (T)

Transmittance (T) characteristics of presumed CuO nanocomposites in the ultraviolet and visible Spectrum. Below is shown the summary of the Principal Observations and the results for (PMMA, CuO, and MeB)>(PMMA, CuO,>(PMMA, CuO and MB) at wave length 400nm, 550nm, and 900nm, as Figure 2: UV Region ($\lambda = 400$ nm), High concentration ($T = 0.450$ or 45%), which indicates significant UV absorption^[10].

An intense decrease at 310 nm implies an optical band gap of nearly ~ 4 eV (calculated using $E_g = 1240/\lambda$, where λ is in nm). This is based on reported values of CuO nanocomposites. Visible Spectrum (400–700 nm):

The transmittance increases, indicating that the molecules become progressively more transparent to visible light^[12].

The combination of high UV absorption and low visible light transmission makes it suitable for UV absorption applications, including coatings, glasses, and other protective films.

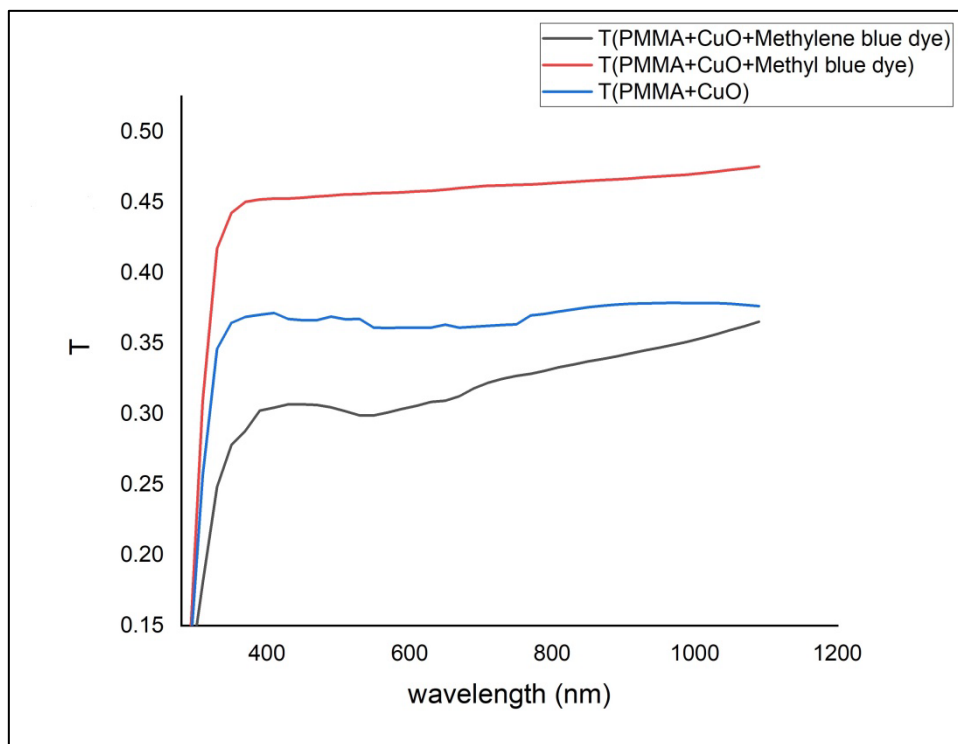


Figure 2. Transmittance between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

Table 2. Showing transmittance

Wavelength (nm)	PMMA/CuO/Methylene Blue (T)	PMMA/CuO/Methyl Blue (T)	PMMA/CuO (T)
400	0.310	0.450	0.360
550	0.299	0.456	0.361
900	0.340	0.459	0.368

4.1.3. Reflectance (R)

Negative UV reflectance indicates more absorption than reflection, signifying the dominance of the absorption process, where the material absorbs most of the UV light rather than reflecting it. That's typical of materials that block UV.

The results show reflectance for (PMMA, CuO, and MeB) > (PMMA, CuO) > (PMMA, CuO and MB) at waves length 320nm, 330nm and 390nm, respectively as Figure 3.

The partially visible light reflectance manifested here is suitable for optical coatings that demand controlled reflection and transmission, e.g., anti-glare or protective coatings.

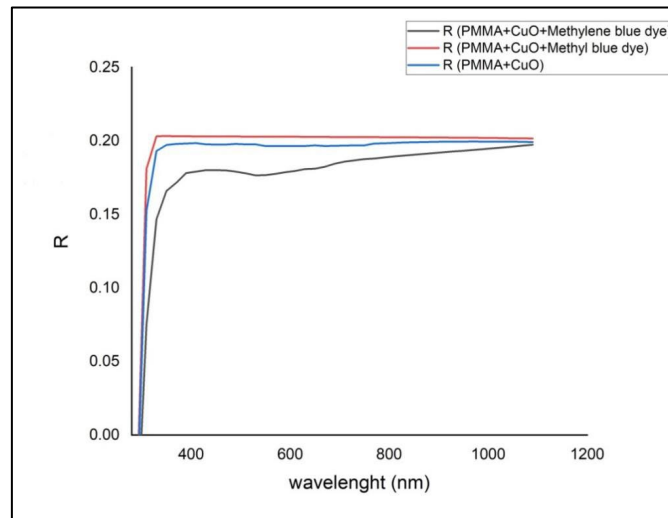


Figure 3. Reflectance between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

4.1.4. Absorption coefficient (α)

High α in UV: Confirms high UV blockage, required for UV protective applications (e.g., sunglasses, UV protective coatings).

The α value of the methyle blue sample is higher than that of other samples, which indicates an enhancement of UV-blocking ability as well. This can be explained by its molecular structure, which can absorb UV light effectively.

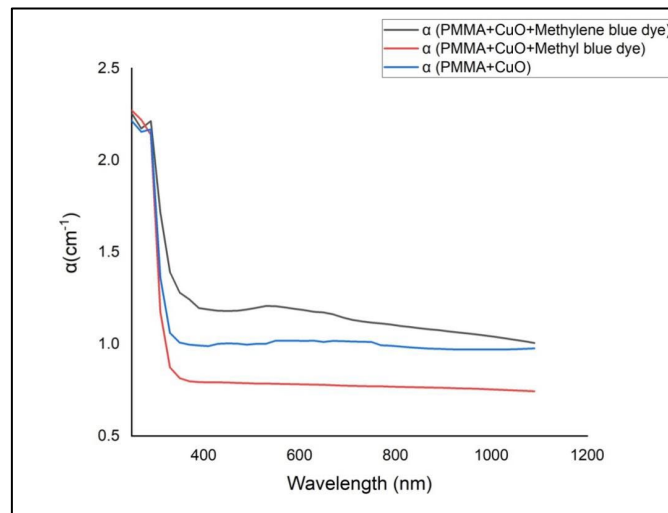


Figure 4. Absorption Coefficient between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

4.1.5. Refractive index (n)

As the refractive index (n) decreases , the wavelength begins to decrease, Then it increases again, demonstrating normal dispersion (typical of most materials), where shorter wavelengths (UV/blue) are refracted more than longer wavelengths (red/IR), as shown in Figure 5. Among the samples, the one containing methyl blue exhibited the highest refractive index, indicating a greater optical density and thus, a higher degree of light-matter interaction. For optical filters, sensors, or photonic devices that benefit from high refractive index material as well, this feature is advantageous.

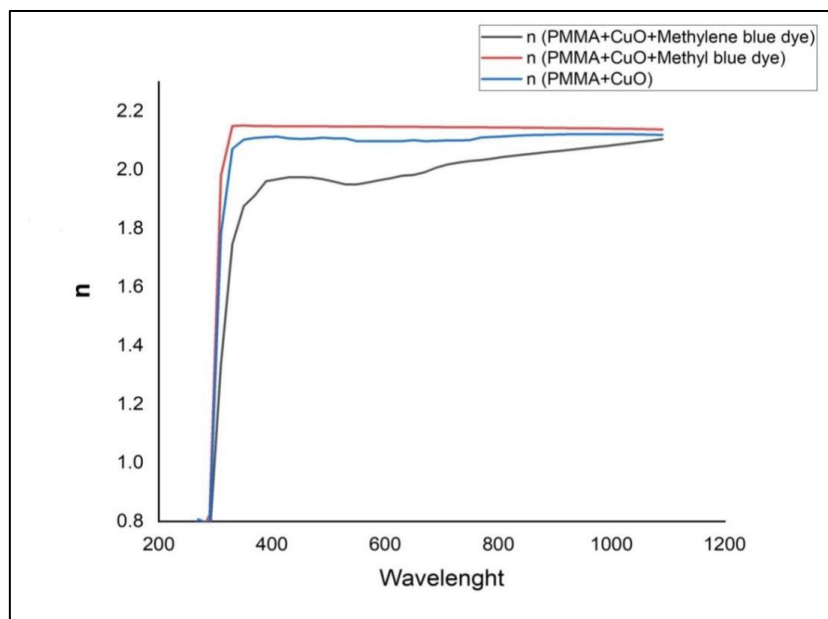


Figure 5. Refractive index between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

Table 3. Refractive Index Comparison

Sample	Refractive Index (n) at 550 nm
PMMA/CuO/Methylene Blue	1.950
PMMA/CuO/Methyl Blue	2.148
PMMA/CuO	2.098

Table 4. Optical Parameters at 550 nm

Parameter	PMMA/CuO/Methylene Blue	PMMA/CuO/Methyl Blue	PMMA/CuO
Absorbance (A)	0.955	0.960	0.945
Transmittance (T)	0.299	0.456	0.361
Refractive Index (n)	1.950	2.148	2.098

The drastic drop of transmittance at the wavelength of 310 nm confirms a value of optical band gap of ~4 eV, which is characteristic of CuO nanocomposites 7–10. Some Possible applications include UV-shielding materials, which allow for low transmission of UV light and can be applied to coatings for sunscreens, eyeglasses, and UV-block textiles. Optical filters: Tunable absorption makes them applicable for selective wavelength filters for image or sensor technologies. Solar Energy: regulated light absorption can be used in dye-sensitized solar cells (DSSCs). Promising Applications of Composites: Ultraviolet protection coatings for

windows, packaging, and exterior materials. Tunable optical filters for the suppression of selected wavelengths. Innovative packaging with built-in protection from UV.

4.1.6. Energy gap

Figure 6 shows Energy in eV: ≈ 1 to 7 eV from the absorption coefficient (α) vs. photon energy ($h\nu$), and $(\alpha h\nu)^2$. The band gap examination (E_g), The band gap energy (E_g) of different compositions is graphically represented as follows:

PMMA/CuO/methylene blue $E_g=3.52\text{eV}$ PMMA/CuO/methyl blue $E_g=3.32\text{eV}$ PMMA/CuO alone $E_g=3.68$ electron volts .This indicates that the presence of dyes decreases the band gap compared to self-PMMA/ CuO. Notably, As new energy states are created within the gap, additional particles such as dye molecules create new electronic energy levels within the original band gap of PMMA/CuO. These new levels act as sublevels or intermediates, facilitating the transition of electrons from the valence band to the conduction band, thereby reducing the band gap. Methyl blue has a more pronounced effect than methylene blue.

The research in this graph may come from new technologies related to the use of materials such as PMMA/ CuO, looking at the influence of dyes on optical properties.

They are dyes that minimize the band gap and can enhance visible-light absorption.

The lower activity level observed with methylene blue compared with methyl blue is possibly due to structural or electronic differences^[13].

E.g., values are labelled explicitly, which makes the graph quite good for qualitative data.

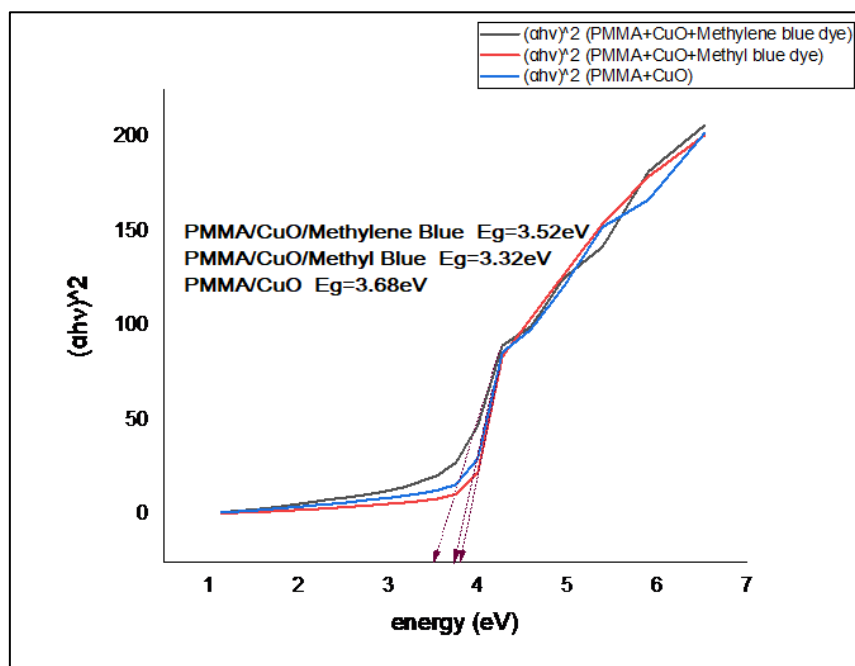


Figure 6. Energy gap between (PMMA/ CuO/ Methylene Blue) , (PMMA/ CuO/ Methyl Blue) and (PMMA/ CuO).

5. The structural properties

5.1. Field emission scanning electron microscopy (FESEM)

A FESEM Study of copper oxide/PMMA thin films that were generated under the identical deposition conditions differ significantly in their morphology, as shown by the field emission scanning electron microscopy (FESEM) figure(7) in (a), (b), and (c). Every sample had the same thickness (8.7–8.9 nm) ^[14].

The typical smooth morphology of the baseline sample (without dye) with nano-dispersed copper oxide nanoparticles (CuO) scattered throughout the PMMA matrix. The significant disparity between the inorganic and organic components makes the nanoscale particles visible, and this uniform distribution is also typical of films [15]. Only faint process markings from the coating are still visible, and the low surface cracking indicates perfect preparation [16].

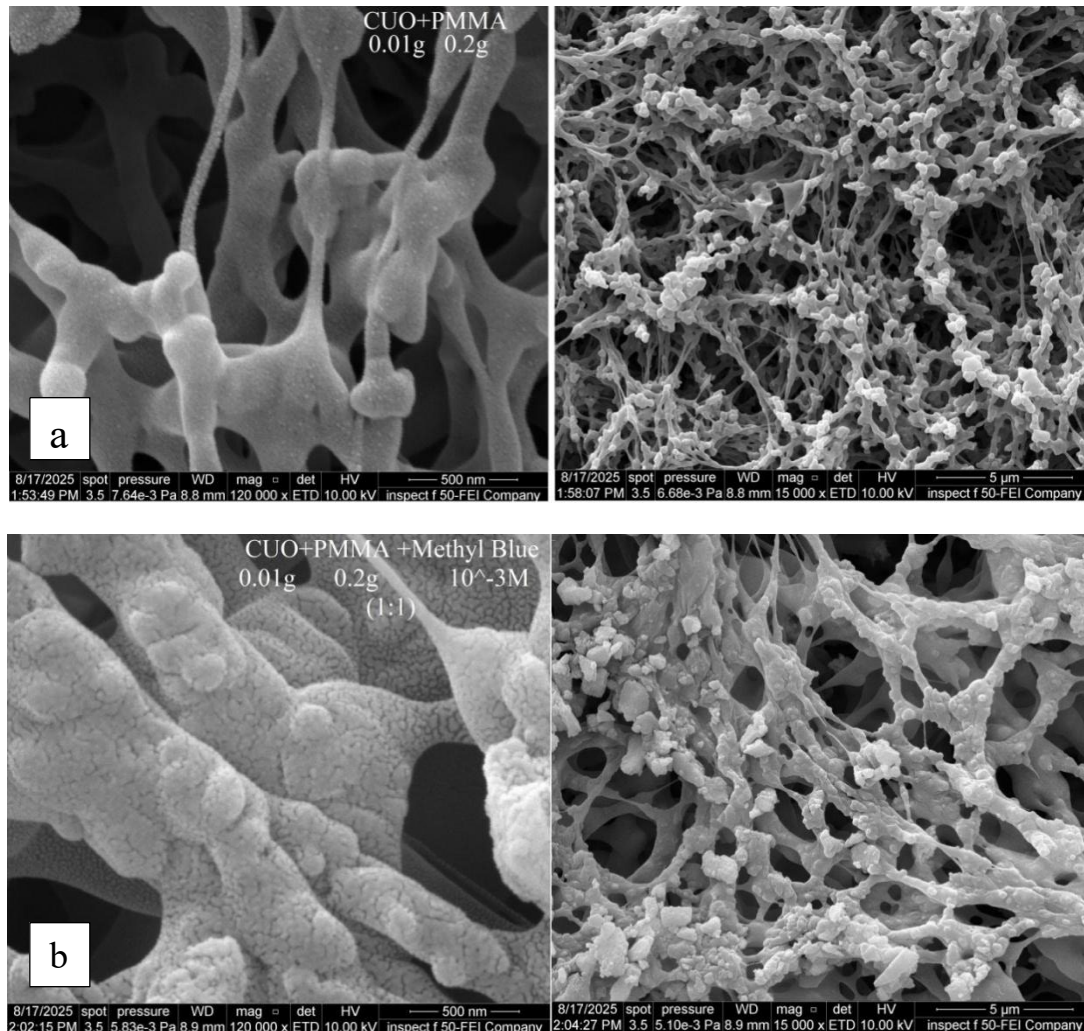


Figure 7. Shows morphology of (a) CuO/PMMA, (b) CuO/PMMA/Methyl and (c) CuO/PMMA/Methylen.

The FESEM images change considerably in areas where methyl blue dye is applied. A coarser topography is created by aggregates that are irregularly spaced. This phenomenon, which results in more saturated black areas on the photographs, is caused by methyl blue's low solubility in PMMA [17]. As previously seen in similar systems, the observed scattering implies that the dye interacts with the copper oxide surfaces, perhaps producing nanoscale clusters [18].

The methylene blue-containing sample looks different. Although the dye coverage appears more consistent than with methyl blue, it forms a thin over layer that covers and covers the nano copper oxide particles underneath. Stronger interfacial contacts are produced by these hydrogen bond complexes between the hydroxyl groups on the oxide's surface and the NH_2 of the dye molecules [19].

The associated Cu oxide NPs show clear crystallinity (phase CuO), according to the XRD curves of the pure CuO NPs. However, both colours helped to conceal crystallinity, with methylene blue's masking impact being enhanced by greater surface contacts [20]. In all situations, dye insertion preserves the PMMA matrix's

amorphous Form even while it increases inter-chain spacing, which may have an impact on mechanical properties [21].

Defect analysis shows that the baseline sample has only minor polymer shrinkage cracks after drying [22]. While the methylene blue samples exhibit tiny holes, likely due to gas generated during dye solidification, the Methyl blue samples display defects associated with dye aggregation resulting from inadequate mixing [23].

Their morphologies differ greatly, suggesting that their uses are also highly distinct. Electrochemical sensors and protective coatings benefit significantly from the undoped film's consistent surface [24]. Methyl blue has been shown to have improved surface properties after modification, making it a candidate for photocatalysis and antibacterial coatings [25]. Its optical behaviour has also been shown to be suitable for the photonic applications of colourimetric sensors [26].

5.2. X-Ray Diffraction (XRD)

Particle size and strain variations occur (property of CuO: $V=0.0811\text{nm}^3$), however. The CuO + PMMA sample has a low dislocation density ($4.27\times10^{13}\text{nm}^{-1}$) and an average particle size of 153 nm. However, the exceptionally high Strain (5.749%) suggests that PMMA and CuO are interacting. Massive particles (363-379 nm), near-zero dislocation densities ($\sim7\times10^{12}\text{nm}^{-1}$), low Strain (300 nm), and shifts in bulk fraction as a function of either full width at half maximum (FWHM) errors or additive-induced crystallization effects are all present in samples with methyl additions.

Upon the addition of methyl blue or methylene blue dyes, the reaction work to structural transformation to is shift the structure from a state of high strain to a more stable and ordered crystalline arrangement. Promotion of particle growth stimulates significant growth in crystallite size (D); this is physically evidenced by the narrowing of the full width at half maximum (FWHM) of the peaks in the XRD pattern. Crystalline phase alterations induce changes in the crystalline phase or peak intensities, indicating that the dye does merely adsorb onto the surface but chemically intercalates and interacts with the nanocomposite matrix.

The effect of adding methyl blue and methylene blue dye additives to CuO/Poly(methyl methacrylate) (PMMA) thin films was investigated in this work. It was explained how the films were prepared and characterized.

The structural changes brought about by various dye additions in spin-coated CuO/PMMA nanocomposite films are clarified by the whole XRD pattern.

CuO/PMMA samples of clean CuO exhibit distinctive diffraction peaks for tenorite CuO at 35.5° (002) and 38.7° (111) 2θ . The pure composite has distinctive, highly intense, sharp peaks that suggest well-developed crystallinity [27]. Let's compare the outcomes of the dye-modified samples. Greater relative intensities of CuO peaks decreased by about 40%.

The prominent peaks' expansion (FWHM rose by 1.2°). At $25^\circ 2\theta$, the new amorphous halo appears.

There are even more striking results for the methylene blue sample. Near-complete suppression of CuO crystalline signals is a prominent trait of the amorphous nature at $22^\circ 2\theta$, with a complete loss of definition between 35° and 40° [28].

Our FESEM results show a correlation with these XRD patterns. The methyl blue samples' altered peak broadening and intensity correspond to the microscopically observed nanoparticulate aggregation. The continuous overlayer creation (in the production of etch pits using electron microscopy) is correlated with excellent crystalline masking in the methylene blue samples. These $22\text{--}25^\circ$ amorphous halos originate from. PMMA chain packing disorder brought on by interactions between polymers and dyes

CuO crystallites need to experience the consequences of nanoscale confinement, specifically the formation of a Dye Molecule-Oxide Interfacial Complex.

Additionally, the following outcomes are significantly impacted by the deposit methods: dye distribution as a result of solvent evaporation dynamics, using film thickness to regulate the depth of X-ray penetration, despite their apparent consistency ^[29]. The 8.7-8.9 mm thickness exhibits different scattering behaviours, as illustrated below. These include variations in mass absorption coefficients, electron density profile comparisons, Changes in interface roughness, and X-ray scattering at small angles for nanostructure characterization. For stability ranges, XRD varies with temperature. This comprehensive XRD analysis shows how much molecular additives can alter the microstructure of the nanocomposite under specific deposition circumstances. These findings significantly impact the design of functional coatings with particular, customized qualities. The FESEM microscopy confirms the X-ray diffraction (XRD) calculations, where the increased size of the crystalline grains leads from fine lattices under the influence of a condenser to a more efficient particle mass.

Table 5. shows the granular size, particle size, Dislocation density and Strain.

Sample	Dominant diffraction peak "2 θ " degree	Crystallite size "D" nm	Particle size (nm)	Dislocation density= $1/D^2$ (nm ⁻¹)	Strain
CuO/PMMA	16.5174	47	153	4.27×10^{13}	5.74906
CuO/PMMA/Methyl	34.645	292.36	379	6.96×10^{12}	0.00442
CuO/PMMA/Methylen	12.0089	280.64	363	7.59×10^{12}	0.01312

6. Conclusion

Employing methyl blue dye to augment the optical properties of synthesized (CuO + PMMA), the absorption, absorption coefficient, and absorption of the dyes methyl blue doped with were found to be superior to those of both methylene blue-doped (CuO/ PMMA) composites and the undoped composite. The enhanced absorbance of (CuO/ PMMA/ Methyl Blue) results from constructive light absorption, related to the optimal dye concentration and maximal photon absorption. The PMMA polymer effectively interacts with CuO nanoparticles and the dye, improving charge transfer and light-harvesting capabilities. This system (CuO/ PMMA/ methyl blue) is a promising contender for optical applications, including laser traps and sensors, due to its significant absorption of specific wavelengths—light-activated gadgets and photodynamic treatment (enhanced light absorption results in increased efficiency).

The addition of a dye fundamentally alters the films' characteristics. methylene blue provides sufficient coverage but can obscure the desired nanofeatures, whereas methyl blue creates problematic aggregates. The intended application will determine which is best, and in either scenario, there is still much process optimization to be done for optimal performance. This comparative analysis can help with the customization of functional nanocomposite films to meet technological requirements.

Conflict of interest

The authors declare no conflict of interest

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