

Original Article

INVESTIGATION OF THE EFFECT OF PH VALUES ON THE PHYSICAL CHARACTERISTICS OF COPPER OXIDE NANOSTRUCTURES BY GREEN ASSISTED METHOD USING *Origanum vulgare* PLANT EXTRACTS

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ABSTRACT

Copper oxide (CuO) nanoparticles were synthesized using the green method. The extract of the *Origanum vulgare* (*O. vulgare*) leaves has been used as a capping and reducing agent of CuO nanoparticles. The influence of pH values in the green growth solution (7.5–10.5) on nanoparticle properties was investigated using various characterization techniques. The obtained results from both optical properties and FTIR analysis confirmed that the *O. vulgare* plant extract is a highly efficient reducing and stabilizing agent. The UV-Visible spectrum of the *O. vulgare* plant extract shows two distinct absorption peaks at 287.439 nm and 324.15 nm at ambient temperature. The FESEM results revealed that the pH of the growth solution significantly affects the morphology, shape, dimensions, and orientation of CuO nanoparticles. The spherical and rod-like nanoparticles were produced as the pH value changed from 7.5 to 10.5, with an average diameter in the range of (16.89–28.50) nm. The obtained results of XRD analysis confirmed that the fabricated CuO nanoparticles have a monoclinic crystal structure with a crystalline size in the range of (10.7–17.81) nm. UV-Visible spectra of CuO nanoparticles at different pH values show strong UV absorbance < 300 nm and have band-gap energies in the range of (1.82–1.98) eV. The FTIR analysis of the nanoparticles confirmed their purity and suggested that nanoparticles synthesized at pH 10 had improved quality and properties.

Keywords: copper oxide, nanoparticle, green synthesis, *Origanum vulgare* extract, pH value.

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1. INTRODUCTION

Nanotechnology is a rapidly advancing field that focuses on the design, production, surface analysis, and use of materials at the nanoscale, typically spanning 1 to 100 nm (Gebreslassie & Gebremeskel, 2024). These substances, referred to as nanoparticles (NPs), exhibit distinctive physicochemical and biological characteristics owing to their tiny dimensions and extensive surface-area-to-volume ratio. Consequently, they demonstrate considerable discrepancies in characteristics relative to their bulk equivalents (Liberty *et al.*, 2024). Nanostructured materials possess the potential to improve energy efficiency and mitigate environmental damage, thereby impacting our planet positively (Keles *et al.*, 2024). Nanoparticles can be synthesized from various metals and metal oxides (Schiopu *et al.*, 2024). A wide range of research has indicated that metal (e.g., Cu, Ag, Pt, Au, Mg, Zn) and metal oxide (e.g., ZnO, TiO₂, CuO, Ag₂O) nanoparticles have substantial therapeutic advantages (Keles *et al.*, 2024). CuO NPs have recently attracted considerable attention among metal oxide NPs due to their unique optical, electrical, magnetic, biological, and catalytic properties. The intriguing properties of CuO nanoparticles have led to a comprehensive investigation across several domains, including energy, electronics, cosmetics, biosensors, storage devices, supercapacitors, catalysis, food and agriculture, and

healthcare (Yasin *et al.*, 2022).

Copper (II) oxide (CuO) is a proficient semiconductor utilized in catalytic photodegradation methods to decompose organic pollutants (Khedr & Ali, 2024). Copper oxide (CuO) has been widely employed to remove hazardous colors from wastewater (Yasin *et al.*, 2022). Although CuO NPs can infiltrate the environment through anthropogenic and natural processes, they accumulate in aquatic and terrestrial ecosystems (Keller *et al.*, 2017). Ultimately, nanoparticles can enter the human body either directly through ingestion or respiration (Areecheewakul *et al.*, 2023; Su *et al.*, 2023) or indirectly (Rajput *et al.*, 2020).

According to quantum confinement effects, CuO nanostructures exhibit exceptional thermal stability, mechanical strength, electrical conductivity, electron correlation effects, spin dynamics, electrochemical properties, magnetic characteristics, superhydrophobicity, photovoltaic capabilities, multiferroicity, and biocidal activity (Atchaya & Meena Devi, 2024). Copper oxide nanostructure is a p-type semiconductor with a low band gap ranging from 1.21 to 2.51 eV (Moroda *et al.*, 2025). In contrast, CuO crystallizes in a monoclinic crystal form characterized by the space group C2/c. The unit cell of cupric oxide has four CuO molecules (Tran & Nguyen, 2014). Because the monoclinic crystal structure of cupric oxide exhibits remarkable physical and chemical characteristics, a significant surface area

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(Katwal *et al.*, 2015; Yang & He, 2011). Consequently, there is an increased interest in developing sustainable and efficient technologies for nanoparticle synthesis. Conventional nanoparticle synthesis methods often rely on hazardous chemicals, high temperatures, and energy-intensive procedures, leading to environmental issues and potential toxicity. In response to these challenges, green synthesis has garnered significant interest as a viable alternative. Green synthesis, or environmentally friendly synthesis, involves using natural resources, biomolecules, or other eco-friendly materials to produce NPs (Aswathi *et al.*, 2023; Madani *et al.*, 2022; Noah & Ndagili, 2022), and the capacity for large-scale manufacturing (Harish *et al.*, 2022; Szczyglewska *et al.*, 2023). The biosynthesis of CuO NPs employs extracts from plants and microorganisms, such as bacteria and fungi. Biological synthesis obviates the necessity for chemical reducing and capping agents. Plant extracts comprise phytochemicals such as Alkaloids, phenols, flavonoids, and terpenoids, which are found in plants (Mohammad inejad *et al.*, 2016). Microorganisms have proteins and enzymes. These natural components serve the dual aims of reduction and capping, yielding nontoxic, biocompatible, and highly stable NPs (Abhimanyu *et al.*, 2023; Ahmed *et al.*, 2016; Ahmed *et al.*, 2017; Nzilu *et al.*, 2023).

Plants have attracted considerable attention as sources of CuO NPs because they are easy to use, provide numerous stabilizing and reducing compounds (Jeronsia *et al.*, 2019; Priya *et al.*, 2023), and enable rapid, economical synthesis of highly pure and concentrated products (Heinlaan *et al.*, 2008). Plants are the most common substrates for nanoparticle synthesis due to their capacity for large-scale manufacturing and for forming stable nanoparticles in diverse sizes and forms (Karam & Abdulrahman, 2022). Prior research has effectively shown the synthesis of CuO NPs via a straightforward method that entails combining copper salt with plant extract. This mixture responds within minutes to hours in a standard lab setting. Phytochemicals make it easier to break down copper salts, which makes nanoparticles. The dramatic change in color was the first proof that CuO NPs were being made (Narasaiah *et al.*, 2017; Singh *et al.*, 2017). Various factors influence the amalgamation of the solution, such as the pH of the reaction solution, the concentration of the plant extract, and the concentration of the metal salt. Additional reaction parameters, such as temperature and duration, significantly influence the quality, morphology, and size of biosynthesized NPs (Abdulqudos & Abdulrahman, 2022). Copper oxides have been documented in various plants; this research concentrates on the extract of *Origanum vulgare*, a highly prized fragrant species with a long history of use as a spice and medicinal herb, as well as a source of critical phytotherapeutic compounds used in modern medicine. In ethnomedicine, oregano is used for its emmenagogue, expectorant, stomachic, and carminative properties. It has been recommended as a tea or tincture for digestive and respiratory conditions, as well as for wound treatment in ointments. Its conventional use encompasses indigestion, diarrhea, cough, and bronchitis (Meretoudi *et al.*, 2021).

The production of CuO NPs using plant extracts has been the subject of considerable investigation. Recently, Mohammadinejad, R., *et al.* (2016) used licorice leaf extract to synthesize CuO NPs and evaluated their photocatalytic, antibacterial, and anticancer properties. Characterization methodologies confirmed the successful synthesis of CuO NPs (Masood & Al Sieadi, 2025). Khedr, A. I., & Ali, M. H. (2024) developed a biocompatible synthesis of CuO NPs using the peel extract of *Citrus aurantium* as a natural reducing and stabilizing agent, with potential applications for effectively reducing methylene blue dye. The characterization revealed monoclinic and spherical nanostructures with a characteristic size range of (53.25–68.02) nm (Khedr & Ali, 2024).

Veisi *et al.* (2021) fabricated nanoparticles of CuO from the floral extract of *Stachys lavandulifolia* in herbal tea form. The fabricated CuO NPs have a mean size of 15–25 nm and an aspherical shape

(Shahid ul *et al.*, 2023). Sukumar *et al.* (2020) also exhibited a practical synthesis of CuO NPs from *Caesalpinia bonducella* seed extract.

The thus-synthesized CuO NPs were further compared for antibacterial performance against *Aeromonas* (Gram-negative) and *Staphylococcus aureus* (Gram-positive) bacteria, indicating improved antibacterial efficiency in both cases (Sukumar *et al.*, 2020). In addition, Qamar *et al.* (2020) prepared CuO nanorods, using *Momordica charantia* fruit extract. The nanorods proved highly antibacterial against multidrug-resistant bacteria as well as fungi, with antiviral potency against the Newcastle disease virus. Green synthesis of CuO NPs was reported by Sharma *et al.* (2021) through the aid of *Aloe barbadensis* leaf extract.

The nanoparticles were spherical, with an average width of 5–6 nm, indicating potential use as antibacterial agents and catalysts. The study emphasized the benignity as well as the economy of plant-mediated synthesis (Sharma *et al.*, 2021). Similarly, Ikhiyoy *et al.* (2023) effectively prepared CuO NPs from *Moringa oleifera* leaf extract, using it as both a reducing and capping agent. The NPs exhibited supercapacitive characteristics, low reflectance in ultraviolet light, along with a 2.5 eV band gap energy, thereby being eligible for energy storage applications. Further, Yasin *et al.* (2022) prepared CuO NPs from leaf extracts of *Passiflora edulis* that possessed exceptional photocatalytic activity in removing methylene blue dye from color (degrading 93% within 160 minutes), along with antioxidant properties.

This study aims to produce CuO nanoparticles using *Origanum vulgare* leaf extract in a green method. The effect of several pH values of the green fabrication mixture (7.5 to 10.5) on the quality of GS CuO NPs has been studied. The optimal pH for bio-growing CuO NPs was determined. The significance of this research is evident from a detailed examination of the factors that affect the shape, size, particle distribution, optical properties, crystal structure, and energy band structure of CuO NPs. The role of green plant biomolecules in the bioreduction of metal salts during nanoparticle extraction is demonstrated.

2. MATERIALS AND METHODS.

Copper nitrate trihydrate ($\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$), with a molecular weight of 241.60 g/mol and a purity exceeding that procured from Sigma-Aldrich. Sodium hydroxide pellets (NaOH), with a molecular weight of 40 g/mol, were utilized in this experiment without modification. A pH meter (PH9500 Research-grade Benchtop pH Meter Kit), magnetic stirrer (MR Hei-End, Germany), and oven (Memmert 100–800, Germany) were essential for the entire manufacturing process and for the synthesis of CuO nanostructures.

Preparation of the *Origanum Vulgare* Plant Extract

Origanum vulgare leaves were employed for the manufacture of copper oxide nanostructures; *Origanum vulgare* is a fragrant herb. Additionally, it is a perennial, robust plant with pinnate leaves and purple blooms, classified in the Lamiaceae family (Benedec *et al.*, 2018). It is indigenous to the mild-temperate zones of western and southwestern Eurasia and the Mediterranean region. It is prevalently located throughout the Mediterranean region of Asia and a portion of Europe (Benedec *et al.*, 2018). Dehydrated pulverized leaves of *Origanum vulgare* were obtained from a local market. 40 g of ground dried *Origanum vulgare* leaves were weighed and rinsed multiple times with deionized water to remove dust and contaminants before use. Subsequently, 40 mL of deionized water was added. Following this, it was heated at 65°C for 1 hour, then brought to room temperature, and then filtered through a plastic filter. The resulting solution was stored at 4°C for further analysis (Moroda *et al.*, 2025). The technique is summarized schematically in Figure 1 as follows:

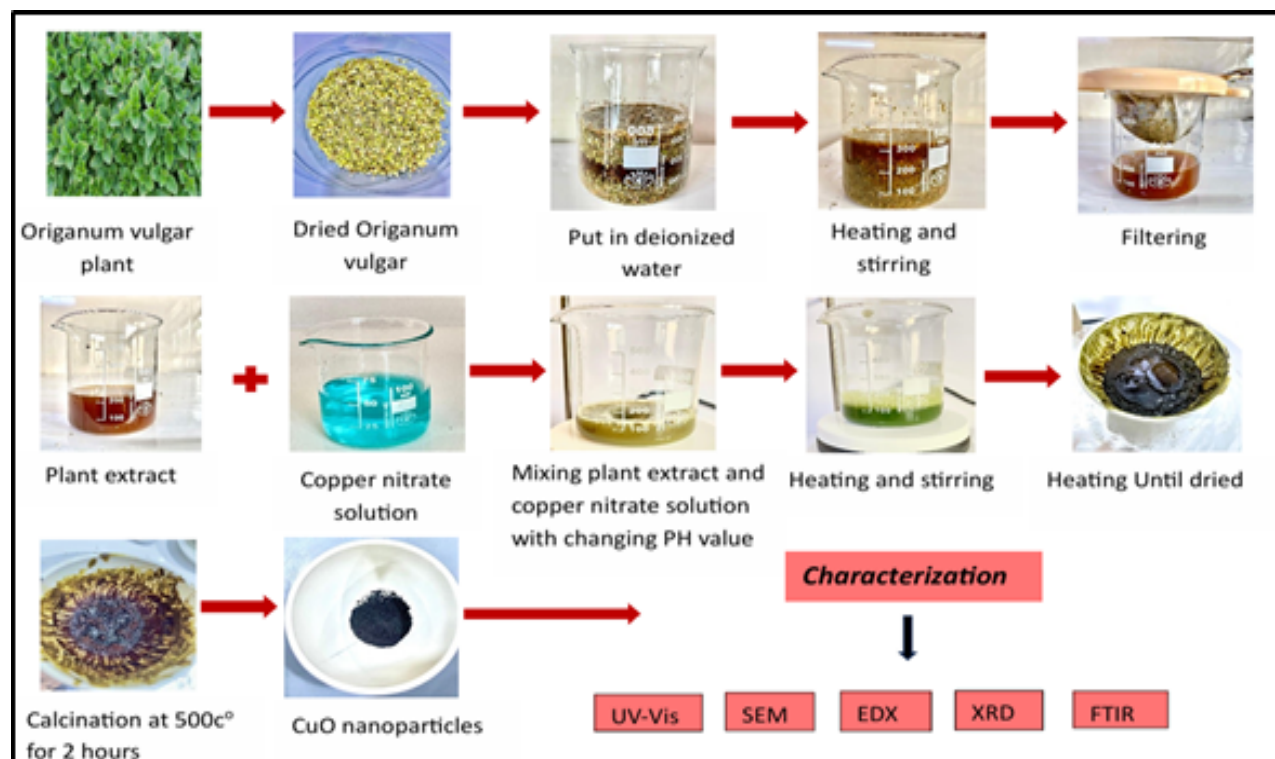


Figure 1: Schematic diagram illustrating the green synthesis of GS CuO nanoparticles utilizing extracts from *O. vulgare*

Green synthesis of CuO nanoparticle

The CuO NPs were synthesized utilizing an environmentally friendly approach with *Origanum vulgare* leaf extract at varying pH levels. Following the procedures detailed in the previous subsection for obtaining the *O. vulgare* leaf extract, 50 mL of the extract was added dropwise to a 0.1 M $[\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}]$ solution in a 1:1 ratio at room temperature for 45 minutes under magnetic stirring at 2000 rpm. Drops of sodium hydroxide (NaOH) were then added to the prepared GS mixture to bring its pH to the required level. While under magnetic stirring, the pH of the solution was consistently measured using a pH meter. As the pH was adjusted, the mixture's appearance and clarity changed until the color became milky dark green, indicating that the metallic salt reaction and the plant extracts occurred and that

CuO NPs were formed. The setup was kept under magnetic stirring at 70°C for four hours until the solution became a hunter-green jelly paste. After that, the remaining mixture was transferred into a crucible and heated at 150°C for four hours to evaporate the water. The resulting dry solid paste was ground into a fine powder and subsequently annealed in a furnace at 500°C for two hours to achieve calcination and obtain pure CuO NPs.

In addition, Ethanol was used to clean the solid powder at temperatures ranging from 75 to 100°C for two cycles to remove impurities and organic materials. Lastly, as shown in Figure 1, the GS CuO NPs results were kept in a tube for characterization. The exact process was carried out for different GS mixture pH values, specifically (7.5, 8.0, 8.5, 9.0, 10.0, 10.5). The green growth mechanism of the CuO NPs is shown in Figure 2.

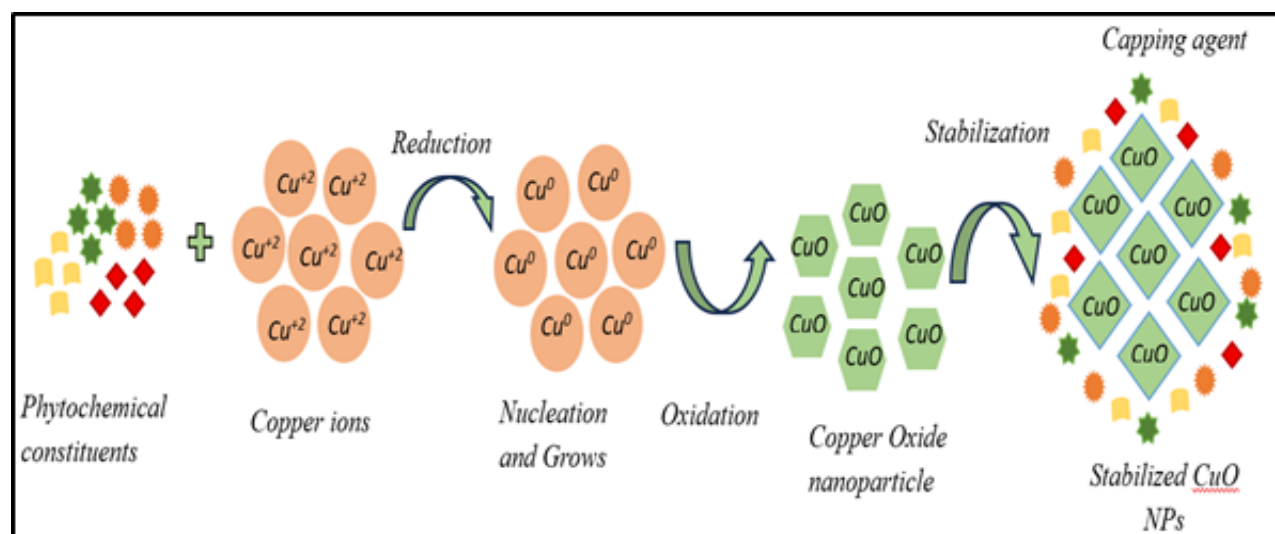


Figure 2: CuO NPs' green growth mechanism (Gebremedhin *et al.*, 2023).

Characterization of Copper Oxide Nanoparticles

The optical properties of *Origanum vulgare* extracts and produced Copper Oxide nanoparticles (CuO NPs) were analyzed using a Cary 100 UV-Vis double-beam spectrophotometer from Agilent Technologies. The absorbance of both CuO NPs and leaf extracts was measured across a wavelength spectrum of 200, 250 to 800 nm. Numerous functional groups in *Origanum vulgare* extracts and CuO NPs produced at varying pH levels and at 500°C were identified. A thorough surface examination, encompassing morphology, chemical composition, particle size, and orientation of the CuO NPs, was performed. Additionally, the crystalline structure, strain, stress, and overall quality of synthesized NPs at different pH levels were evaluated using Fourier Transform Infrared Spectroscopy (FTIR) with a Nicolet IS10 instrument from Thermo Scientific, spanning a range of 400 to 4000 cm^{-1} . Additional characterization was conducted utilizing Energy-Dispersive X-ray Spectroscopy (EDX) and Scanning Electron Microscopy (SEM, Quanta 450). High-resolution X-ray diffraction (HR-XRD) analysis was performed using a Pro MRD (PAN X-Pert) with $\text{CuK}\alpha$ radiation (wavelength 1.54050 Å) over a scanning angle range of 5° to 80°.

3. RESULTS AND DISCUSSION

Characterization of Origanum Vulgare Extract.

In recent years, researchers have employed plant-derived phytochemicals to fabricate Copper Oxide nanoparticles (CuO NPs), thereby effectively controlling their form, morphology, orientation, and size through green means. The phytochemicals found in each component of metal ions are transferred to the metal nanoparticles by a plant extract (Yasin *et al.*, 2022). Consequently, the plant extract is influenced by a reducing and stabilizing agent. UV-Visible spectroscopy was successfully employed to investigate and acquire this chemical mechanism. The UV-Vis spectrum exhibited a peak absorption corresponding to surface plasmon resonance (SPR), which also captures conduction-band electron oscillations. Electromagnetic waves indicate the production of nanoparticles and the reduction of metal ions (Yasin *et al.*, 2022).

Figure 3 illustrates the UV-Visible spectra of the *Origanum vulgare* plant extract. Two prominent peaks were observed at 287.439 nm and 324.15 nm, potentially associated with the predominant phytochemicals in *Origanum vulgare*. Due to the widespread use of OH groups as stabilizing agents, bio-reduction can be employed to synthesize NPs (Jeronsia *et al.*, 2019). Due to their antioxidant effects and absence of harmful compounds, these phytochemicals are highly effective in reducing metal ions and stabilizing them at the nanoscale. They can also produce nanoparticles with various sizes and morphologies. The copper ions form a $\text{Cu}(\text{OH})_2$ compound via the hydroxide groups. Three mechanisms are involved in the phase transition from $\text{Cu}(\text{OH})_2$ to CuO: reprecipitation of dissolution, in situ crystallization, and solid-solid phase transition. Additionally, Khedr and Ali (2024) have suggested that the lattice lets water out during the solid-to-solid phase change.

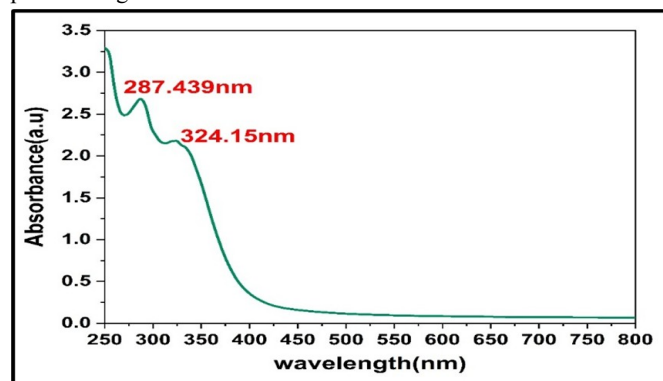


Figure 3: UV-Vis. Spectrum analysis of the *O. vulgare* extracts.

FTIR stands for Fourier Transform Infrared Spectroscopy. Researchers have used infrared spectra of plant extracts to identify functional groups associated with reductive biomolecules and to identify those that facilitate the reduction of CuO nanoparticles (Jeronsia *et al.*, 2019; Khedr & Ali, 2024). The FTIR spectra of the *O. vulgare* plant extract are depicted in Figure 4, revealing many peaks within the range of 400 to 4000 cm^{-1} . The FTIR spectra are essentially categorized into two regions. The fingerprint region, spanning from (0–1500) cm^{-1} , is where the plant extract can be observed and utilized for the synthesis of CuO nanoparticles. The functional group region spans from 1800 to 4000 cm^{-1} . FTIR spectra often provide insights into the bonding characteristics of materials throughout all states of matter.

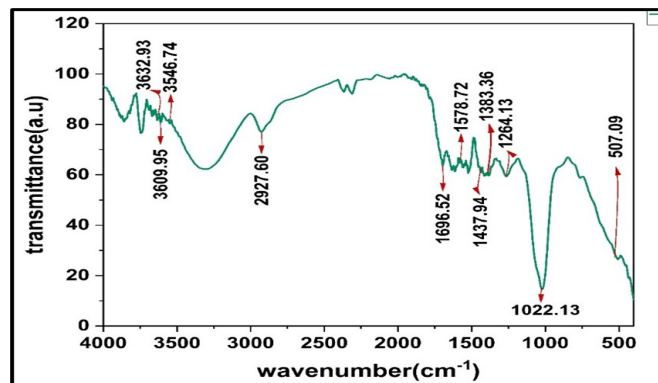


Figure 4: FTIR Spectrum analysis of the *O. Vulgaris* plant Extracts

The peak absorbed at 507.09 cm^{-1} corresponds to the S–S stretch of polysulfides and aryl disulfides in thiols and thio-substituted compounds. The observed peak at 1036 cm^{-1} corresponds to the vibrations of the cyclohexane ring and the methylene ($> \text{CH}_2$) groups of saturated aliphatic compounds (alkenes/alkyls). The measured absorption peak for aromatic ethers, specifically the aryl–O stretch of ether and oxy compounds, is at 1264.13 cm^{-1} . The gem-Dimethyl or "iso" (doublet) is observed at the absorption peak of 1383.36 cm^{-1} . The Methyl C–H asymmetric/symmetric bend is observed at a peak of approximately 1437.94 cm^{-1} , corresponding to the Methyl ($-\text{CH}_3$) bond in saturated aliphatic compounds (alkene/alkyl). The analyzed signal at about 1578.72 cm^{-1} corresponds to the Carboxylate (carboxylic acid salt) of a Carbonyl molecule. The peak at 1696.52 cm^{-1} corresponds to the aromatic combination bands of the aromatic ring (aryl). Nevertheless, the apex at 2927.60 cm^{-1} corresponds to the asymmetrical/symmetrical stretching of Methylene C–H, specifically for Methylene ($> \text{CH}_2$) in saturated aliphatic compounds (alkene/alkyl). The observed peak at 3546.74 cm^{-1} corresponds to the internal OH stretch of alcohols and hydroxy compounds. In contrast, the nonbonded hydroxy group (OH) stretch of alcohols and hydroxy compounds is located at 3609.95 cm^{-1} . A peak at 3632.93 was found, indicating the presence of secondary alcohol, as well as the OH stretch characteristic of alcohols and hydroxy compounds (Kim *et al.*, 2001).

Characterization of the GS CuO Nanoparticles

FTIR Spectroscopic Study of CuO NPs

The FTIR spectra were used to analyze the structure, identify functional groups, and determine the chemical composition of CuO nanoparticles. The FTIR spectrum of the copper oxide nanostructures synthesized using *Origanum vulgare* extract revealed multiple functional groups, indicating successful phytochemical-assisted synthesis. FTIR spectra of the solid phase were recorded between 4000 cm^{-1} and 400 cm^{-1} and compared with standard FTIR spectra.

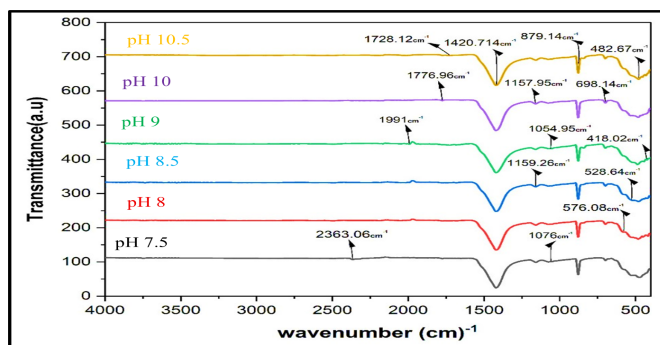


Figure 5: FTIR Spectrum Analysis of the CuO nanoparticles using the *O. vulgare* plant extract at Different pH values.

The FTIR spectrum of the sample exhibited many characteristic absorption peaks (Atchaya & Meena Devi, 2024; Santos *et al.*, 2011). In Figure 5, six biosynthesized samples of CuO NPs were produced at varying pH values. Significant peaks were seen at 476.04 cm^{-1} , 482.67 cm^{-1} , and 486.98 cm^{-1} , indicative of the S–S stretching vibrations of polysulfides, characteristic of thiols and thio-substituted chemicals. An extensive absorption peak between 418.02 cm^{-1} and 481.23 cm^{-1} signifies the existence of aryl disulfides (S–S stretch) linked to thiols and thio-substituted chemicals. This region corresponds to peroxides and C–O–O– stretching vibrations of ethers and other oxygenated molecules. Three absorption bands were identified at 518.58 , 531.51 , and 528.64 cm^{-1} , resembling the C–I stretch of aliphatic iodo compounds within organohalogen compounds. An absorption peak at 576.04 cm^{-1} is attributed to the C–S stretch of disulfides in thiols and thio-substituted chemicals. The peaks at 698.14 cm^{-1} , 877.71 cm^{-1} , and 879.14 cm^{-1} correspond to $\text{CH}_2\text{--S}$ (C–S stretch) bending and trans-C–H out-of-plane bending vibrations, indicating the existence of thioethers and olefinic structures. The absorption peak at 1054.95 cm^{-1} is ascribed to the methylene ring vibrations of a cyclohexane configuration. A notable peak identified within the range of $1073\text{--}1076\text{ cm}^{-1}$ is attributed to C–O stretching vibrations of ethers and various oxygenated compounds, including alkyl, cyclic, and substituted ethers, as well as big-ring structures.

The peak at 1157.95 cm^{-1} is due to the CN stretching vibration of secondary amines. The signal at 1159.26 cm^{-1} is related to tertiary alcohols, indicating C–O stretching of alcohol and hydroxy groups. The C–H in-plane bending vibrations are shown by peaks at 1419.27 cm^{-1} , 1420.71 cm^{-1} , and 1422.14 cm^{-1} . An absorption signal at 1728.12 cm^{-1} indicates the presence of a carbonyl group from an ester. Two more peaks observed at $(1775.52)\text{ cm}^{-1}$ and $(1776.96)\text{ cm}^{-1}$ are ascribed to peroxides, alkyl carbonates, or five-membered ring anhydrides. Conversely, the peak at 1991.00 cm^{-1} is indicative of the C–O stretching vibration in phenolic compounds. A robust absorption band at 2363.06 cm^{-1} indicates the presence of cyanate groups, which is also consistent with nitrogen-containing compounds exhibiting various functionalities.

Optical Properties of the GS CuO NPs

The spectroscopic features of GS Copper Oxide nanoparticles (CuO NPs) synthesized at different pH values have been investigated and validated using double-beam (UV-Visible) spectrophotometry by analyzing the absorption spectra. The absorption spectra of GS CuO NPs synthesized, utilizing extracts from the leaves of the *O. vulgare* plant at varying pH levels within the wavelength range of 220 to 700 nm are illustrated in Figure 10. The GS CuO NPs powder demonstrates strong ultraviolet absorption across a wide pH range, with an effective absorbance below 400 nm, along with outstanding clarity and a narrow observable absorption spectrum. Crystal quality, lattice parameters, crystal dimensions, oxygen vacancies, and donor impurities, which explain the lower absorption values at longer wavelengths, all affect defects in GS CuO NPs (Atchaya & Meena Devi, 2024). Within the wavelength range of 246.7 to 253 nm, there have

been reports of strong exciton absorption and a pronounced ultra-violet absorption border that are related to the optical energy band gap (E_g) of GS CuO NPs at varying pH levels. Upon sharpening the peaks, monodisperse CuO NPs are generated via surface plasmon resonance (SPR), exhibiting a maximum UV-Vis absorption peak at 246.253 nm. CuO NPs generally exhibit absorption maxima across a broad spectrum of 270–380 nm, influenced by particle size, shape, crystallinity, and synthesis conditions.

The proximity of the observed peak to the lower edge of this range indicates the formation of small nanoparticles, leading to a pronounced blue shift in the absorption spectra as the pH varies. This alteration is frequently associated with an elevation in band gap energy resulting from the quantum confinement effect, a defining attribute of nanoscale materials (Jabeen *et al.*, 2024).

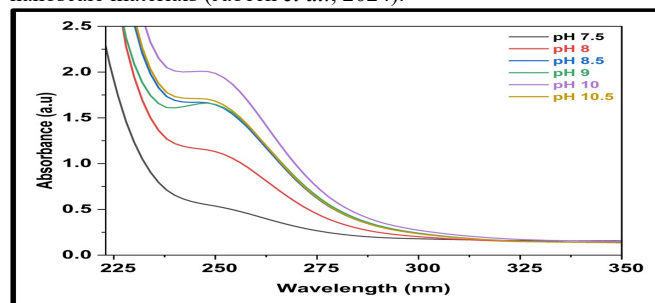


Figure 6: The UV-Visible spectra analysis of biosynthesized the GS CuO NPs using the extracts of *O. Vulgare* plant at Different pH values.

The exponential relationship of $(\alpha h\nu)^{1/2}$ against $(h\nu)$ is depicted by transmittance spectra, as seen in Figure 7. The Tauc formula is utilized to determine the optical band-gap energy of GS CuO nanoparticles employing *O. vulgare* plant extracts at varying pH levels:

$$(\alpha h\nu)^{1/2} = A(h\nu - E_g) \quad (1)$$

In this case, α , $h\nu$, A , and E_g stand for the absorption coefficient, photon energy, constant, and optical band gap energy, respectively. Also, n depends on the transmission and equals $1/2$ for allowed direct transmission. The primary focus of the (α) coefficient container represents the transmittance spectrum, utilized by Atchaya and Meena Devi (2024).

$$\alpha = \frac{\ln\left(\frac{1}{T}\right)}{d} \quad (2)$$

While "T" and "d" denote the transmissions and thicknesses that are intended for CuO samples.

In this study, the energy band gap is calculated to be indirect. Figure 7 shows that the transition energy of the synthesized GS CuO NPs at various pH levels ranges from 1.82 eV to 1.98 eV. This shows the optical band gap, which occurs when the valence and conduction bands switch places, corresponding to the optical energy gap of the semiconductor GS CuO NPs. This range is higher than the published band gap for CuO NPs, 2.1 eV. The significant disparity in energy levels between the valence and conduction bands in the nanoparticles can be attributed to quantum confinement, a size-dependent effect. Consequently, the band gap energy increases as particle size diminishes, and the nanocrystals become smaller than the Bohr radius of the excited electron-hole pair. This finding aligns with prior research (Jabeen *et al.*, 2024). The surface atom exhibits a reduced coordination number, raising the energy of the highest valence band and reducing the intensity of the lowest unoccupied conduction band. As a result, the band gap energy increases. The graphs in Figure 7 indicate that the produced GS CuO NPs exhibit energy gaps (E_g) at various pH values: 1.82 eV, 1.91 eV, 1.94 eV, 1.98 eV, 1.85 eV, and 1.95 eV for pH levels of 7.5, 8, 8.5, 9, 10, and 10.5, respectively. The energy band gap diminishes as the pH of the green-produced mixture increases. The reduction in the E_g of the nanoparticles is attributed to changes in the lattice constant of the leaf extracts; the use of plant extracts often results in a diminished band gap [63]. This finding, primarily noted in GS NPs, aligns with instances of quantum confinement effects (Masood & Al Sicadi, 2025).

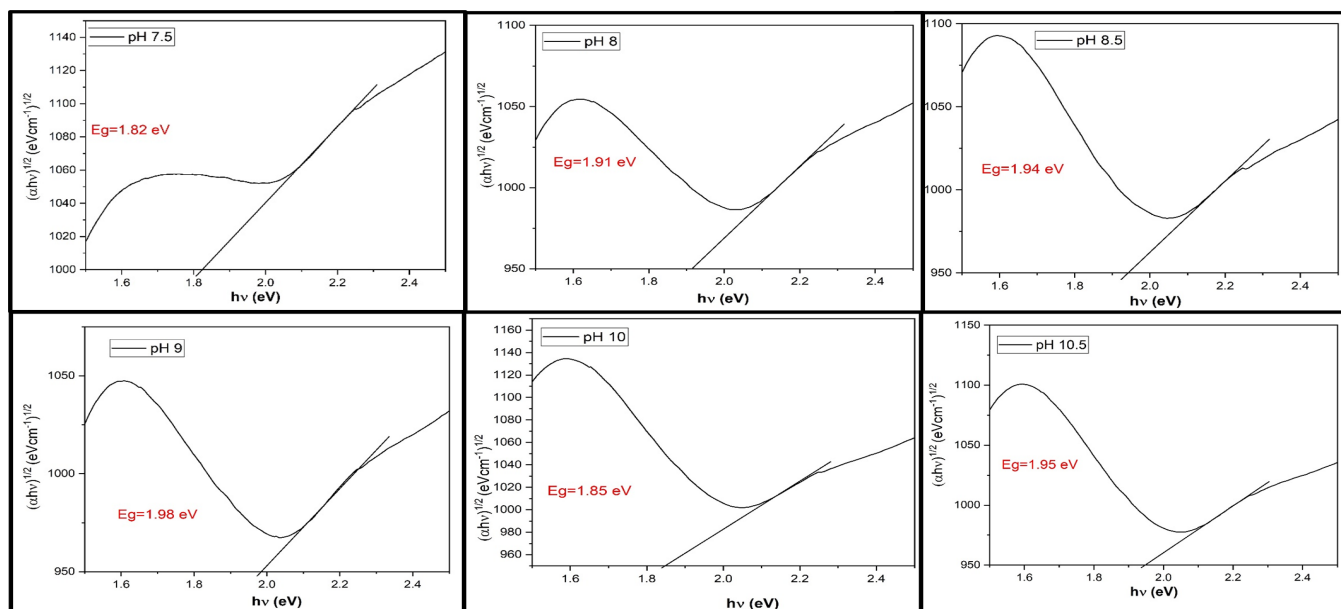


Figure 7: Tauc-plot vs. energy band gap of GS CuO nanoparticles utilizing *O.vulgar* extracts at varying pH values.

Energy Dispersive X-ray Spectroscopy (EDX) Analysis of ZnO NPs

EDX spectroscopy is used to analyze and quantify the chemical composition of the GS CuO NPs. Figure 8 shows the EDX (or EDS) analysis, indicating that both Oxygen (O) and Copper (Cu) elements are present. This means there are no signs of faults within the EDS limits associated with the structural features of the GS CuO NPs (Priya *et al.*, 2023).

The EDX spectra exhibited two prominent peaks corresponding to Cu at around (1.1) keV and (8.7) keV, together with an oxygen

peak at (0.5) keV, indicating the distinctive features of CuO NPs (Ali *et al.*, 2023). The atomic ratio of copper (Cu) to oxygen (O) remains consistent among all GS CuO NPs synthesized at different pH values. The molar ratio of copper to oxygen in the synthesized nanoparticles is 1:1, as determined by quantitative EDX analysis, indicating that the analyzed nanoparticles are pure GS CuO. The molecular percentages of copper (Cu) and oxygen (O) are as follows: Cu (52.22%, 57.02%, 50.80%, 61.96%, 95.14%, and 57.04%) and O (57.77%, 42.97%, 49.19%, 38.03%, 4.85%, and 42.95%) at different pH levels of 7.5, 8, 8.5, 9, 10, and 10.5, respectively.

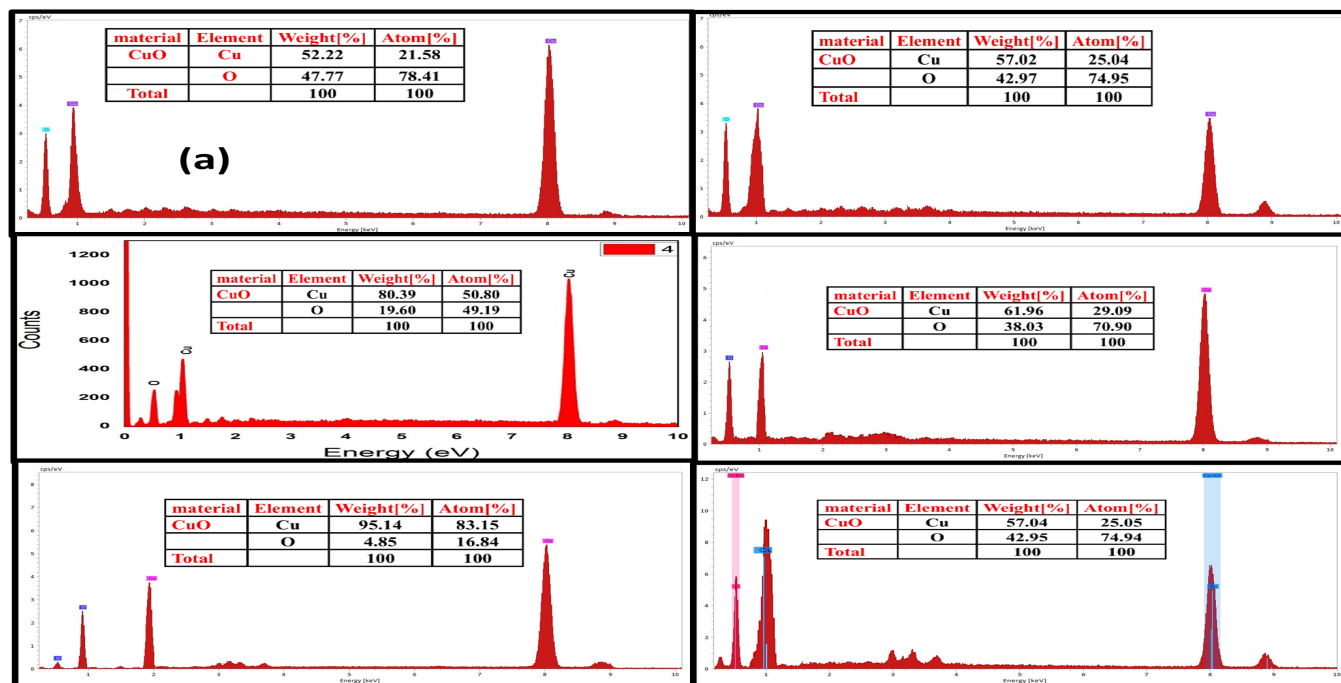


Figure 8: The *O.vulgar* extracts at the following pH values were used in the EDX analysis of the GS CuO NPs: (a) pH 7.5, (b) pH 8, (c) pH 8.5, (d) pH 9, (e) pH 10, and (f) pH 10.5.

The Morphological Analysis of the GS CuO NPs

Figure 9 shows the top view of the shape of GS CuO nanoparticles that were fabricated using *O. vulgare* plant extracts at different pH levels. This was done using SEM analysis. Figure 9 shows that changing the pH of the green manufacturing mixture from 7.5 to 10.5 has significantly altered the dimensions, morphology, topography, orientation, and dispersion. The CuO NPs have sizes of less than 100 nanometers. In Figure 9 (a), and at pH 7.5, GS CuO NPs are produced with a heterogeneous distribution, predominantly including aggregated irregular spherical nanoparticles, mixed with sporadic elongated rod-like structures, leading to moderately dense clusters due to inadequate electrostatic stabilization and significant aggregation with an average size of 24.02 nm. Figure 9(b) illustrates that raising the pH to 8 increased the average nanoparticle size to 28.01 nm, along with a more uneven distribution. This distribution exhibits a higher ratio of rod- and needle-shaped particles interspersed within tightly packed aggregates of fused nanoparticles, indicating a shift towards anisotropic development under moderately elevated alkalinity.

Raising the pH value to 8.5, as illustrated in Figure 9(c), results in a morphology characterized by massive, elongated nanostructures with smoother, more uniform surfaces, forming substantial structures.

23.31 nm enhanced anisotropic crystal formation, and intensified particle agglomeration facilitated by elevated pH, respectively. At pH 9, as shown in Figure 9(d), the nanostructures displayed irregular quasi-spherical particles characterized by significant aggregation and constrained form definition, with a particle size of 16.48 nm. In Figure 9 (e), increasing the pH to 10 resulted in particles measuring 23.51 nm, which facilitated the formation of well-defined rod-like and plate-like structures within a fine nanoparticle matrix, indicating better anisotropic growth and improved structural regularity. At a pH of 10.5, the particles exhibited an average particle size of 17.23 nm, aggregating with irregular morphology and diminished separation, indicating excessive nucleation and unregulated growth. The observations suggest that pH 10 offers the most advantageous conditions for generating homogenous, well-oriented nanostructures with little aggregation, establishing it as the optimal synthesis condition among the evaluated pH levels. Comprising a combination of spherical and rod-like CuO nanostructures, some exhibiting polycrystallinity, with discernible grain boundaries in larger fused aggregates, indicates the coalescence of numerous initial nanoparticles during growth. The overall morphology suggests restricted form regularity, perhaps resulting from rapid nucleation and development at elevated pH levels (Yang & He, 2011).

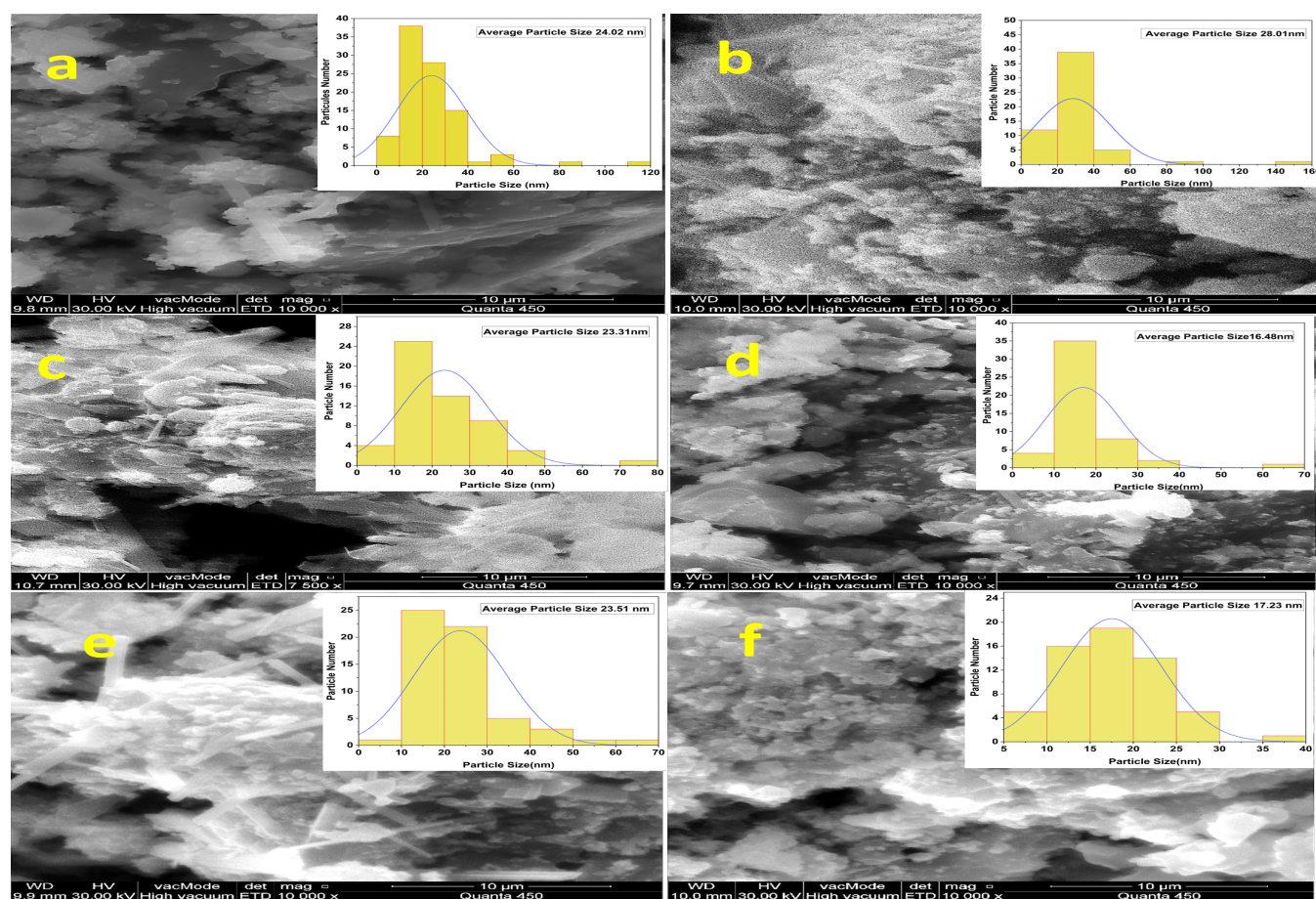


Figure 9: The SEM patterns of the GS CuO NPs by using the Leaf extracts of the *O.vulgare* Plant at Different pH values: (a) pH 7.5, (b) pH 8, (c) pH 8.5, (d) pH 9, (e) pH 10, and (f) pH 10.5

The Structural Characteristics of GS CuO Nanoparticles

The nondestructive XRD technique was employed to investigate the structural parameters of GS CuO nanoparticles produced from *O. vulgare* plant extracts at different pH values. The polycrystalline crystal structure of the CuO NPs is illustrated in Figure 10. And all diffraction peaks in the XRD patterns examined have been classified as monoclinic. Additionally, the diffraction peaks in all XRD patterns were

indexed and correlated with the standard spectrum database (JCPDS card number 98-062-8612). Further, the absence of any peak diffraction from other defects (impurities) was confirmed, indicating that the GS CuO NPs were fabricated and grown with a very high purity of CuO NPs from the *O. vulgare* plant extracts at varying pH values. In Figure 10, the two predominant and distinct diffraction peaks, 002 and 111, exhibiting variable intensities, are given in the X-ray diffraction patterns at all pH levels. The intensity of these diffraction peaks

changed as the pH of the GS solution changed. Also, the diffraction peaks along the (101) plane are more noticeable. The obtained XRD results are in complete agreement with prior studies (Aroob *et al.*, 2023; Javid-Naderi *et al.*, 2023; Taghavi Fardood *et al.*, 2023).

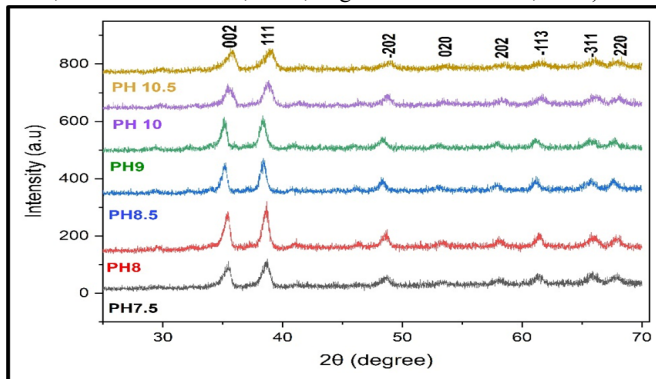


Figure 10: XRD Patterns of GS CuO nanoparticles utilizing *O.vulgar* plant extract at different pH values.

The Full Width at Half Maximum (FWHM) value of the GS CuO NPs can be determined by analyzing the XRD patterns to determine the distribution of compositional phases and the purity of the crystalline structure. The Debye-Scherrer equation was successfully used to find the average size of GS CuO NPs at different pH values (002) and (111) diffraction peaks, which are both substantial and robust. The results of the investigation are summarized in Table 1.

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (3)$$

where K, D, diffraction angle, X-ray beam wavelength, and crystallite size shape factor are, in turn, the full width at half maximum (FWHM) of the peak. The pH transition from 7.5 to 10.5 has resulted in a modification of the average particle size of GS CuO NPs. The thermal energy during the annealing process causes the particles to reorient, which in turn diminishes the defects in the crystal structure. As a result, the typical particle size of the CuO NPs fluctuates.

Table 1: XRD results and the average size of the particulates under investigation.

Sample	peak	FWHM	Pos. [° 2Th.]	D (nm)	$\delta (\text{\AA}^{-2}) \times 10^{-5}$	d (Å)
pH7.5	002	0.7085	35.5420	11.78	7.206	2.52590
pH8	002	0.3542	35.4777	23.55	1.801	2.53033
pH8.5	002	0.3542	35.2704	23.54	1.803	2.54472
pH9	002	0.3936	35.1781	21.18	2.228	2.55118
pH10	002	0.5510	35.4154	15.14	4.361	2.53463
pH10.5	002	0.5510	35.8281	15.15	4.351	2.50638
pH7.5	111	0.5510	38.6918	15.28	4.278	2.32722
pH8	111	0.5117	38.7320	16.46	3.689	2.32489
pH8.5	111	0.4723	38.4049	17.81	3.149	2.34394
pH9	111	0.4723	38.3994	17.81	3.149	2.34426
pH10	111	0.7872	38.7556	10.7	8.730	2.32353
pH10.5	111	0.6298	39.1153	13.39	5.575	2.30299

The corresponding equation is used to ascertain the dislocation density (δ) at various pH levels, subsequently integrating it with the dislocation density of the synthesized CuO nanoparticle.

$$\delta = \frac{1}{D^2} \quad (4)$$

D denotes the crystallize size.

Table 1 indicates that the pH of the green manufacturing combination strongly influences the number of flaws in GS CuO NPs. The table shows that adjusting the pH significantly affects the interplanar distance of biosynthesized CuO NPs. The subsequent equation has been utilized for this study (Lakshmanan *et al.*, 2021):

$$\frac{1}{d^2} = \frac{1}{\sin^2 \beta} \left(\frac{h^2}{a^2} + \frac{k^2 \sin^2 \beta}{b^2} + \frac{l^2}{c^2} - \frac{2hl \cos \beta}{ac} \right) \quad (5)$$

Where:

d_{hkl} is the interplanar spacing for the Miller indices (hkl).

h, k, l are the Miller indices, which are integers representing the planes.

a, b, c are the lattice parameters of the monoclinic crystal.

β is the angle between the lattice vectors a and c .

$\sin^2 \beta$ and $\cos \beta$ are the sine and cosine of the angle β .

Conclusion

CuO nanoparticles were successfully biosynthesized with *O. vulgare* leaf extracts. The investigation explored nanoparticle properties as a result of varying extract concentration and pH levels. The results showed that *O. vulgare* extracts offer a highly efficient and environmentally friendly route for synthesizing CuO nanoparticles under different pH conditions. SEM characterization revealed nearly spherical and nanorod-shaped CuO nanoparticles of (16.89–28.50) nm in size, with pH-dependent association and morphology. UV-Visible spectra exhibited intense absorbance below 300 nm, with bandgap energy decreasing from 1.82 eV to 1.98 eV as pH increased. The FTIR characterization proved the purity of the resulting CuO nanoparticles. In summary, CuO nanoparticles prepared at 10 pH exhibited improved structural quality and good optical performance compared with those prepared at other pH values.

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Ethical Statement

None

Conflict of Interest

The authors declare no competing interests.

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None

Author Contributions

H. A. H., Data Collection, Laboratory Analysis, Design of the Project, Data Analysis and Writing. A. F. A., Supervision, Project, Administration, Validation, Review & Editing.

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