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Eco-Friendly Biosynthesis Of Copper Nanoparticles Using Phyllanthus Emblica And Their Characterization

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Abstract
Nanotechnology has transformed numerous domains, encompassing medicine, energy, and environmental restoration, owing to the distinctive attributes of nanoparticles (NPs). Among these, copper nanoparticles (CuNPs) have attracted scrutiny for their economical viability, antimicrobial, antioxidant, and catalytic capabilities. Conventional synthesis methodologies frequently entail perilous chemicals, necessitating the pursuit of environmentally benign alternatives. This research investigates the environmentally sustainable synthesis of CuNPs utilizing Phyllanthus emblica bark extract, capitalizing on its abundant phytochemical composition as a reducing and stabilizing agent. The synthesized CuNPs were characterized employing UV-Vis spectroscopy, Fourier-transform infrared spectroscopy (FTIR), dynamic light scattering (DLS), scanning electron microscopy (SEM), and X-ray diffraction (XRD). UV-Vis analysis corroborated NP formation with a surface plasmon resonance peak at 254 nm. FTIR elucidated the participation of hydroxyl, amine, and carbonyl groups in capping and stabilization. DLS indicated a Z-average dimension of 240 nm and a zeta potential of -3.44 mV, implying moderate stability. SEM imagery depicted spherical to quasi-spherical morphology with slight aggregation, while XRD validated the monoclinic crystalline architecture of CuO NPs. The research underscores the efficacy of P. emblica in generating stable, crystalline CuNPs, presenting a sustainable alternative to traditional methodologies. These NPs exhibit potential for biomedical and industrial applications, consistent with the principles of green chemistry and sustainable nanotechnology.
Keywords: *Nanotechnology, Green synthesis, Copper nanoparticles (CuNPs),* Phyllanthus emblica, *Sustainable nanotechnology*

INTRODUCTION

Nanotechnology has acquired considerable prominence in recent years owing to the exceptional attributes and extensive applications of nanoparticles across diverse domains including, but not confined to, medicine, energy, and electronics [1]. The distinctive attributes of nanoparticles, encompassing their minuscule dimensions (typically ranging from 1 to 100 nm), vast surface area, and modifiable surface chemistry, render them ideal for precise pharmaceutical delivery, enhanced antimicrobial efficacy, and environmental remediation [2]. Researchers have employed a variety of metals such as gold, silver, copper, zinc, iron, chromium, platinum, and palladium to synthesize nanoparticles, facilitating a multitude of applications [3]. Green synthesis can be utilized to fabricate nanoparticles with dimensions spanning from 1 to 100 nm. Nanoparticles exhibit unique physical and chemical attributes that render them exceedingly desirable for numerous applications in sectors such as medicine, electronics, and environmental restoration [4]. Nanoscale materials are implemented in several industries including textiles, pigment and dye production, nanomedicine, wastewater treatment, food manufacturing, regulation of plant metabolic pathways, lithium-ion battery sensors, and catalytic applications [5]. Traditionally, the synthesis of nanoparticles typically necessitates the use of hazardous chemicals and solvents, which can pose detrimental effects on both the environment and human health. Green synthesis methodologies present a sustainable and ecologically responsible approach to the production of nanoparticles. These technologies utilize natural and renewable resources as catalysts, while minimizing the application of toxic chemicals [6]. A commonly employed technique for generating nanoparticles in an environmentally sustainable manner involves the utilization of plant extracts. The plant extracts encompass a diverse array of biologically active compounds that can function as both reducing and stabilizing agents during the formation of nanoparticles [7]. Microorganisms and enzymes may also be utilized for ecologically sound production of nanoparticles. Bacteria, fungi, and yeast possess the capability to generate nanoparticles by reducing metal ions. Additionally, enzymes such as lipases and proteases can also act as reducing agents

in the fabrication of nanoparticles [8]. Plants are widely recognized as highly effective sources for the synthesis of nanoparticles [9]. Nanoparticles (NPs) can be generated through several methodologies, including physical, chemical, biological, and hybrid processes. There exist two principal methodologies for the generation of NPs: the top-down approach, which relies on physical processes, and the bottom-up method, which employs chemical and biological activities [10]. The top-down strategy entails the reduction of bulk materials to nanoscale dimensions utilizing procedures such as cutting, grinding, and scratching without precise control at the atomic level. The techniques employed to fabricate nanoparticles include laser ablation, vacuum vapor deposition, pulsed wire discharge, and mechanical milling [10,11]. Conversely, the bottom-up methodology involves constructing the structure of NPs by commencing with individual atoms, molecules, or clusters. Chemical reduction, microemulsion techniques, non-chemical reduction, electrochemical, microwave-assisted, and hydrothermal methods are frequently utilized for the synthesis of nanoparticles employing chemical methodologies [10]. The concept of "Green Chemistry" for "Sustainable Development" has been the focus of considerable investigation over the preceding decade [12]. The morphological attributes of the green synthesized nanoparticles are governed by a multitude of factors. The form and dimensions of the nanoparticles are swayed by an array of variables, encompassing the concentration and constituents of the botanical extract, temperature, agitation rate, reaction duration, pH of the reaction mixture, among others [13]. During the synthesis of nanoparticles, the phytoconstituents serve as both stabilizers and reducing agents. The dimensions and morphology of the nanoparticles are influenced by the quantity of bio-reducing agents present in the botanical extract. An elevated concentration of botanical extract enhances the nucleation of nanoparticles, thereby accelerating their formation. The nucleation rate escalates in direct correlation to the concentration of the reducing agent within the botanical extract. However, rapid biosynthesis rates accompanied by high extract concentrations yield smaller nanoparticles [14].

Phyllanthus emblica L (Phyllanthaceae), locally designated as amla or Indian gooseberry, represents an exceptional and widely utilized herb within Indian Ayurvedic practices. It is referred to by the common appellation “the Monarch of Rasyana” due to its remarkable restoration, revitalization, and reconstructive properties [15,16]. In ancient Ayurvedic medical manuscripts (500 BC), it is identified as Charak Smitha. Beyond Indian Ayurveda, it is employed in the Unani therapeutic system. P. emblica is profoundly nutrient-dense and may serve as a significant alimentary source of amino acids, vitamin C, and minerals. P. emblica additionally encompasses a variety of phenolic compounds such as tannins, rutin, phyllembelic acid, phyllembelin, curcuminoids, and emblicol, among others [17]. The entirety of the plant (all components) can be utilized for therapeutic purposes, particularly the fruit, which has been employed in Ayurveda as a formidable rejuvenating agent, the so-called Rasayana. Its fruit has also been utilized in traditional medicine for the management of jaundice, diarrhea, and inflammation. Moreover, various plant components have demonstrated antibacterial, antioxidant, antidiabetic, hypolipidemic, antiulcerogenic, gastroprotective, hepatoprotective, and chemopreventive attributes [18]. P. emblica fruit is a valuable endowment of nature that is indigenous to India. It is predominantly found on hill slopes and coastal regions above 200 m. Additionally, P. emblica has notable applications in enhancing memory, treating respiratory, dermatological, and ophthalmological ailments, as well as in detoxification, including the remediation of snake venom. Furthermore, this review presents retrospective analyses on P. emblica at the molecular level for disease management and control. The fruit of P. emblica is also incorporated in a Thai traditional herbal formulation known as Triphala, which comprises P. emblica, Terminalia bellirica (Gaertn.) Roxb., and Terminalia chebula Retz. [19]. Numerous herbal and patented pharmaceuticals have been developed utilizing the diverse constituents of P. emblica [20].

Copper nanoparticles (CuNPs) are acquiring prominence as economical and effective substitutes for noble metal nanoparticles, attributable to their antimicrobial, antioxidant, anticancer, and catalytic capabilities [21,22]. Traditionally, copper nanoparticles are fabricated through physical and chemical methodologies, which frequently entail hazardous substances, elevated energy consumption, and toxic byproducts, thereby raising apprehensions regarding environmental safety and sustainability [23]. In this regard, green synthesis methodologies, particularly plant-mediated biosynthesis, have emerged as an environmentally friendly and sustainable alternative. This approach employs phytochemicals present in plant extracts that function as reducing, capping, and stabilizing agents, thus obviating the necessity for external chemical reagents [24]. These bioactive compounds not only bestow potent therapeutic attributes but also facilitate the bioreduction of metal ions into nanoparticles. Prior investigations have illustrated

that P. emblica extract can proficiently synthesize silver and gold nanoparticles, signifying its substantial reducing capacity [25]. Nonetheless, scant literature is accessible regarding the biosynthesis of copper nanoparticles utilizing Phyllanthus emblica, and the current study endeavors to bridge this chasm. In the present endeavor, copper nanoparticles were synthesized employing aqueous extract of Phyllanthus emblica bark and meticulously characterized utilizing UV-Vis spectrophotometry, Fourier-transform infrared spectroscopy (FTIR), Dynamic light scattering (DLS), Scanning electron microscopy (SEM), and X-ray diffraction (XRD). These characterization techniques contribute to elucidating the optical properties, functional groups involved in stabilization, particle size distribution, surface morphology, and crystalline nature of the biosynthesized nanoparticles, respectively.

MATERIALS AND METHODS

Preparation of leaf extracts and synthesis of copper nanoparticles

All the chemicals of analytical grade were acquired from Sigma-Aldrich chemicals. Phyllanthus emblica bark of 30 grams was thoroughly washed, air-dried, powdered, and subsequently boiled in 100 milliliters of distilled water for 10 minutes at a temperature below its boiling point to avert the charring of the secondary metabolite. The resultant crude extract was filtered through Whatman filter paper (pore sizes- 25 µM and 0.6 µM). To the 20 milliliters of Phyllanthus emblica bark extract, 80 milliliters of an aqueous solution of 1 mM copper sulfate (CuSO4) was incorporated and maintained under stirring conditions utilizing a magnetic stirrer for 24 hours at room temperature. The incubated nanoparticles were subjected to centrifugation for 10 minutes at 10,000 rpm. Subsequently, the pellet was rinsed and centrifuged with distilled water, and the synthesized nanoparticle was desiccated in a hot air oven at a minimal temperature to prevent charring.

Characterization of synthesized copper nanoparticles

UV–Visible Spectroscopy

The reduction of copper ions and formation of CuNPs was monitored using UV–Vis spectroscopy. The absorption spectra of the colloidal solution were recorded in the range of 200–800 nm using a UV–Vis spectrophotometer (e.g., Shimadzu UV-1800), and the characteristic surface plasmon resonance (SPR) peak was noted [26].

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR analysis was performed using an FTIR spectrometer (e.g., PerkinElmer) in the range of 400–4000 cm^{-1} to identify the possible biomolecules responsible for the reduction and capping of CuNPs. Dried samples of CuNPs were mixed with KBr and pressed into pellets for measurement [27].

Dynamic Light Scattering (DLS) and Zeta Potential

The average particle size distribution and zeta potential of the synthesized CuNPs were determined using a DLS instrument (e.g., Malvern Zetasizer). These measurements help in understanding the stability and dispersion of nanoparticles in aqueous medium [28].

Scanning Electron Microscopy (SEM)

The surface morphology and shape of CuNPs were analyzed by scanning electron microscopy. A thin layer of the dried nanoparticle sample was sputter-coated with gold before analysis. SEM images provided information on the surface texture and particle aggregation [29].

X-ray Diffraction (XRD)

The crystalline structure of the synthesized CuNPs was studied using an X-ray diffractometer (e.g., Bruker D8 Advance) operating at 40 kV and 30 mA with Cu-Kα radiation (λ = 1.5406 Å). The diffraction pattern was recorded in the 2θ range of 20°–80°, and the crystallite size was calculated using the Debye–Scherrer equation [30].

RESULTS AND DISCUSSION

UV- analysis

Ultraviolet-Visible (UV-Vis) spectroscopy quantifies the extinction (scattering + absorption) of light traversing a specimen. Nanoparticles (NPs) possess distinctive optical attributes that are contingent upon the dimensions, morphology, concentration, agglomeration state, and refractive index in proximity to the NP surface, thereby rendering UV-Vis an invaluable apparatus for the identification, characterization, and examination of nanomaterials [31]. The synthesized Copper Nanoparticles (CuNPs) were characterized utilizing the UV/Vis spectrophotometer Lambda 35. The biosynthesized CuNP was

scrutinized by UV-spectrophotometer across an absorbance range from 250 to 800 nanometers. The UV-Vis absorption spectrum reveals peaks indicative of the surface plasmon resonance of CuNPs. The spectroscopic evaluation of synthesized CuNPs manifested the peak absorbance at 254 nanometers, signifying the presence of biosynthesized CuNPs within the reaction mixture (Figure 1). These experimental inquiries were ascertained to align favorably with the findings previously documented in the literature by Singh et al., 2021 [32].

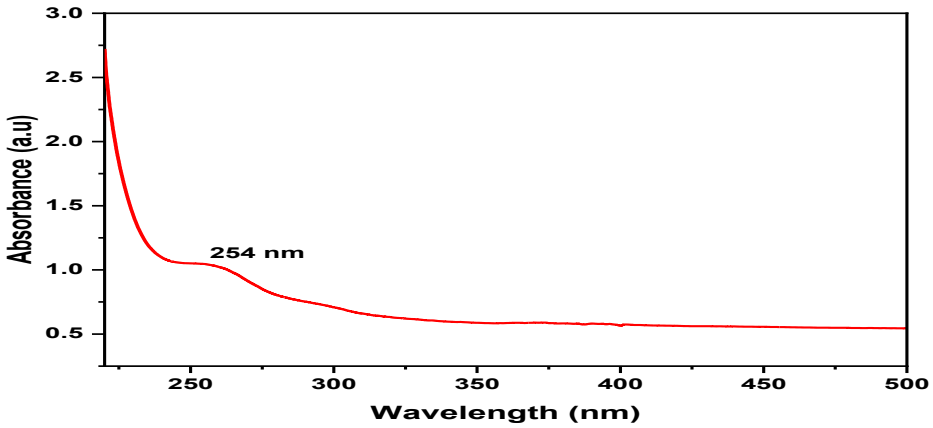

Figure 1: UV spectrum of copper nanoparticle

FTIR analysis

The FTIR spectra of the synthesized CuNPs are presented in Figure 2. The spectrum was recorded in the wavelength domain between 400 and 4000 cm−1. The FTIR spectrum of the CuNPs exhibited substantial absorption bands in the range of 3367–3847 cm^{-1}, signifying the presence of hydroxyl (–OH) groups, potentially attributed to alcohols, phenols, or hydrogen-bonded water molecules. Peaks between 3597 and 3462 cm^{-1} imply the existence of secondary amines or N–H stretching vibrations, whereas a broader band near 3367 cm^{-1} may also indicate carboxylic acids. The peaks in the vicinity of 2526–2200 cm^{-1} are ascribed to triple bond stretches such as nitriles (–C≡N) or alkynes (–C≡C). Minor bands between 2137 and 2063 cm^{-1} suggest the presence of isocyanate or analogous heteroatom-containing groups. Weak overtone bands around 2002–1934 cm^{-1} could be ascribed to carbonyl overtones. A sharp band at 1606 cm^{-1} signifies C=C stretching in aromatic rings or N–H bending, indicating the presence of amines or aromatic systems. The pronounced peak at 1085 cm^{-1} is correlated with C–O or C–N stretching, affirming the presence of ethers, esters, or amines. Lower frequency peaks between 700 and 607 cm^{-1} denote aromatic ring deformations, and the peaks at 459–430 cm^{-1} reside within the metal–oxygen or complex ring skeletal vibration realm. The FTIR results are in commendable concordance with the study reported by Jabeen et al. [33], and also bear resemblance to the findings regarding copper oxide nanoparticles synthesized by the conventional method as reported by Keabadile et al., 2020 [34].

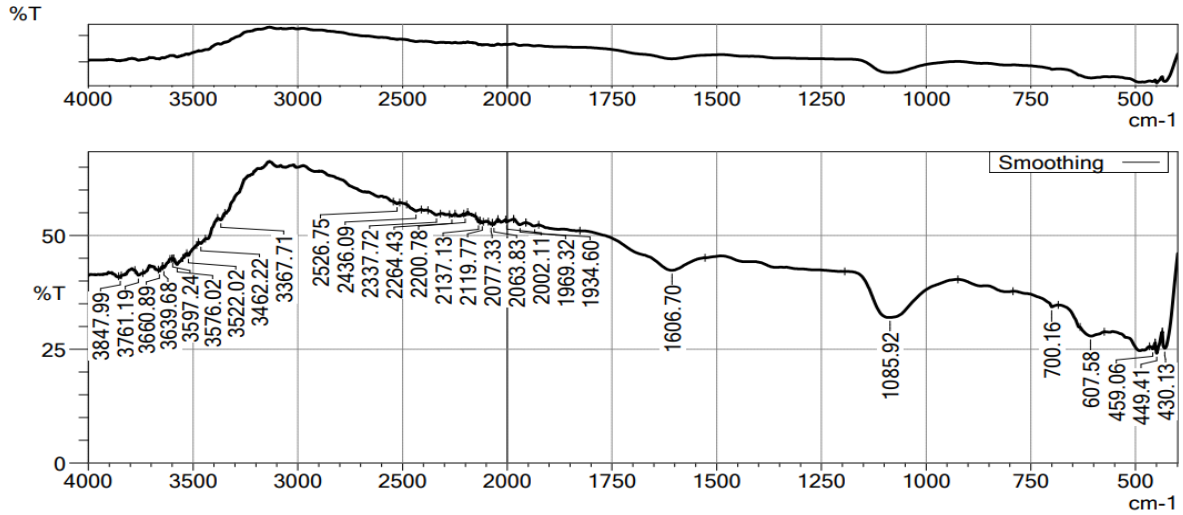

Figure 2: FTIR spectrum of copper nanoparticle

Dynamic light scattering

Dynamic Light Scattering (DLS) analysis elucidated that the copper nanoparticles synthesized utilizing Phyllanthus emblica exhibited a Z-average particle size of 240.0 nm with a singular predominant peak, implying the formation of relatively monodisperse nanoparticles (Figure 3). The intensity-based size distribution signifies that the particles reside within the nanoscale spectrum, albeit slightly exceeding the <100 nm range frequently pursued in biomedical applications. This augmentation in size may be ascribed to surface capping by phytochemicals derived from P. emblica, as similarly observed in other green synthesis protocols. The Zeta potential of the synthesized nanoparticles was determined to be -3.44 mV, denoting moderate colloidal stability. Although a zeta potential magnitude of ±30 mV or superior is generally deemed optimal for electrostatic stability, numerous biosynthesized copper nanoparticles exhibit values ranging from -5 to -25 mV due to neutral or weakly charged capping agents (such as polyphenols, flavonoids, and proteins). The DLS outcome of the current study aligns with the investigation reported by Kumar et al., 2020 [35].

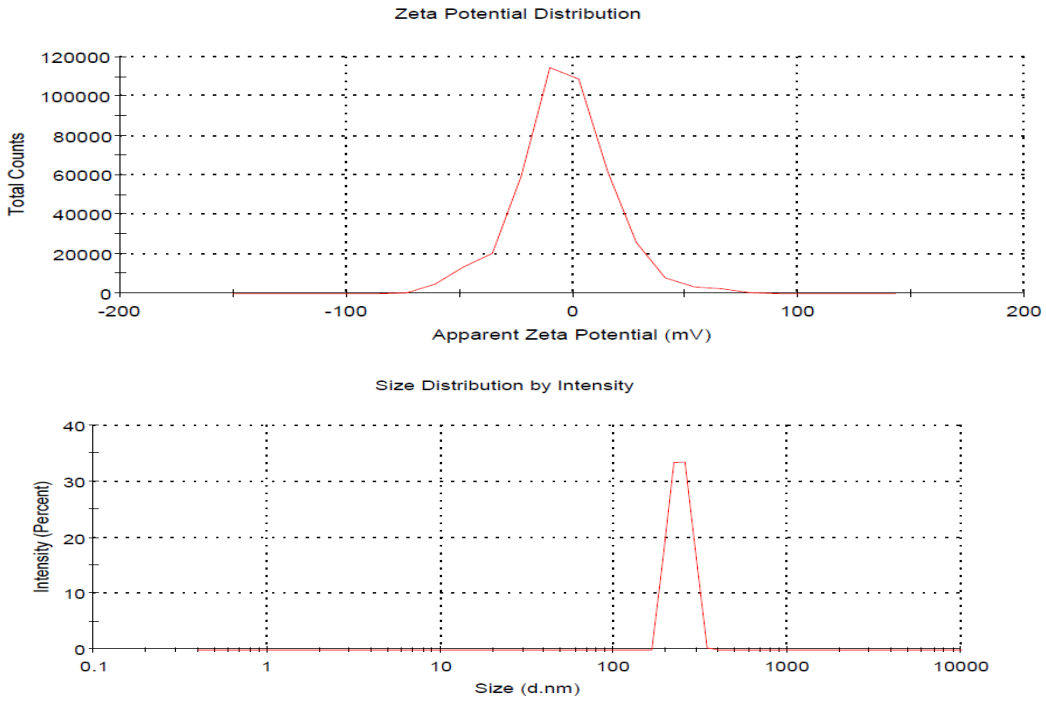

Figure 3: Zeta potential and size distribution of nanoparticle

SEM analysis

The SEM image of the copper nanoparticles fabricated utilizing Phyllanthus emblica demonstrates predominantly spherical to quasi-spherical morphology with discernible aggregation (Figure 4). This observation is in agreement with prior documentation by Kumar et al. [35], wherein Annona squamosa-mediated CuNPs manifested analogous shapes and moderate clustering attributable to bio-organic capping agents. Azizi et al. [36] likewise discerned aggregated spherical CuNPs synthesized through Syzygium aromaticum, with dimensions ranging from 80–200 nm. The morphology observed herein substantiates the successful reduction and stabilization by phytochemicals, which typically culminate in aggregated yet uniform particle distributions. These revelations further corroborate the efficacy of green synthesis in regulating nanoparticle shape and size.

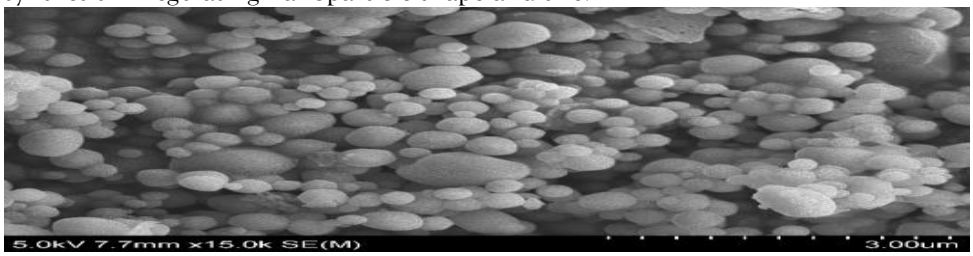

Figure 4: SEM image of copper nanoparticles

XRD analysis

The XRD profile of the copper nanoparticle demonstrates pronounced diffraction peaks at 2θ = 18.96, 22.50, 24.26, 27.23, 31.84, and 37.43 corresponding to the (110), (002), (111), (202), (113), (311), and (004) orientations of monoclinic CuO, in accordance with JCPDS card no. 45-0937 (Figure 5). No ancillary phases were observed, signifying elevated phase purity, akin to preceding reports concerning strictly crystalline CuO nanoparticles. The XRD profile denotes a semi-crystalline or nanocrystalline substance with numerous sharp peaks and moderate peak broadening. The preeminent peak at 24.26° implies a specific crystalline phase, likely denoting the principal component. Peaks in proximity to 27.2°, 31.8°, and 37.4° are frequently observed in silica-based or metal oxide substances. The peak broadening and relative intensities suggest the material possesses fine crystallites, potentially within the nano-range (~51.6 nm). The present study's findings are congruent with the investigation reported by Fatma et al., 2017 [31], delineating the crystalline character of the synthesized copper nanoparticles utilizing Passiflora foetida leaf extract.

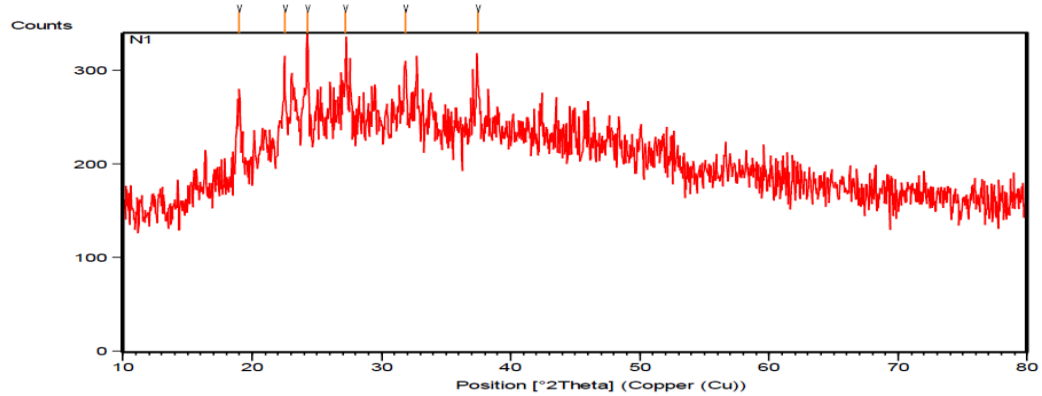

Figure 5: X-ray diffraction pattern of copper nanoparticle

CONCLUSION

The present study successfully demonstrates the green synthesis of copper nanoparticles using Phyllanthus emblica bark extract, which acts as a natural reducing and stabilizing agent. The biosynthesized CuNPs were effectively characterized through UV–Vis, FTIR, DLS, SEM, and XRD techniques, confirming their optical, morphological, and structural properties. The formation of spherical CuNPs with moderate stability and a crystalline nature aligns well with previously reported green synthesis methodologies. This environmentally benign approach offers a promising alternative to conventional synthesis methods, eliminating the use of hazardous chemicals and aligning with the principles of sustainable nanotechnology. The synthesized CuNPs possess potential for applications in diverse sectors including antimicrobial therapy, catalysis, and environmental remediation. Future studies may focus on evaluating their biological efficacy and scalability for industrial applications.

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