A review of new developments in the synthesis of CuO nanoparticles via plant extracts for enhancing the photocatalytic activity

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Abstract: Metal and metal oxide nanoparticles are used widely in a variety of fields of science, research organizations, and industry sectors due to recent advancements in nanoscience and nanotechnologies. Due to their exclusive its unique characteristics and uses, copper oxide nanoparticles (CuO NPs) have drawn more attention than further other metal oxides. The expensive components reagents, equipment, and environmental hazards and risks connected to the physical and chemical processes of CuO NPs synthesis have been a major cause for concern. This review main features of a collection of thorough data from the latest advancements in the synthesis, characterization, and applications from prior research studies on the biological method of synthesizing CuO NPs in the sample order to puffer a solution to the given aforementioned techniques by aiming to reduce reducing environmental pollution and producing inexpensive cheaper nanoparticles with effective characteristics. CuO NPs demonstrated astounding photocatalytic efficiency against the degradation of industrial waste dye. For the photocatalytic destruction of organic contaminants, CuO NPs have high prospective applications. This review study provides additional information on the use of CuO photocatalysts, which are low-cost and environmentally acceptable, to efficiently remove hazardous colors from industrial wastewater. This investigation also provides useful and informative knowledge on the instant synthesis of CuO NPs from plant extracts with desired properties.

Keywords: Copper oxide; Green synthesis; Photocatalytic activity.

1. INTRODUCTION

Although there is a greater understanding of the need to prevent environmental contamination, various industries continue to release the excessive accumulation of synthetic or natural toxins, such as dyes, hazardous metals, pharmacologic waste, and modern agricultural waste, such as pesticide residues, into the environment [1-3]. Since dyes are utilized by numerous industries, including those in the polymer, paper, synthetic fibers, rubber, cosmetic, and agricultural companies, they are one of these that contribute the most significant amounts to environmental pollution [4-6]. Because of their multifaceted aromatic structure and difficulty in degradation, dyes can have negative consequences for the aquatic environment, including color change, odor change, nutrient enrichment, under-oxygenation, and bioavailability [7-9]. Therefore, it is extremely essential to remove them from

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industrial effluent in order to obtain concentrations below legally permitted levels for dispersal.

Currently, a number of methods, including adsorption, chemical coagulation, membrane processes, sedimentation, and advanced oxidation process (AOPs), have been explored for the removal of contaminants from industrial wastewater [10-11]. AOPs are becoming increasingly important recently because of their capacity to produce an adequate quantity of highly reactive radicals for efficient water purification. There are many AOPs, but photocatalysis and catalysis using Copper oxide nanoparticles (CuO NPs) have garnered considerable interest as effective methods for the destruction of harmful organic pollutants [12-14, 96]. In the case of photocatalytic degradation, exposure to photons with energies above the band gap of NPs causes the synthesis of electron and hole pairs, which in facilitate and encourage in the production of highly reactive oxygen species (ROS), which eventually take part in the breakdown of hazardous chemicals [15-17]. It is clear that the capacity to generate more ROS is the limiting element for catalytic degradation [18-20]. Thermal breakdown of water occurs in such harsh conditions, producing extremely reactive radical species including $OH^{\bullet}$, $H^{\bullet}$, and $O^{\bullet}$ that can oxidize and destroy biodegradable toxins in wastewater [21-23, 94]. There has been a noticeable movement toward using NPs in emerging green and ecologically sustainable methodologies in synthetic chemistry in line with environmental issues [24-26].

In addition to interfacial interactions, shape, and size-dependent dependent features play a significant role in determining how effectively CuO NPs work in the intended application [27-29, 98]. Therefore, a long-term goal in the effort to create CuO NPs is to create simple methods for controlling the morphology, size, shape, and composition. The chemical and physical characteristics of CuO NPs can be adjusted attributable to this control. The synthesis of CuO NPs was conducted using a variety of techniques, using a greener approach that produced several benefits in the synthesis of CuO NPs, including low energy use, rate efficiency, shortened dispensation times, lower toxicity, use of a green solvent, and an inferior reaction temperature with high productivity as related to other techniques [30-32, 99]. Both external and intracellular methods can be used to create NPs in a socially and environmentally responsible manner. NPs production employing extracts collected through diverse processes is a part of extracellular techniques. Using extracts from a wide variety of herbal species, including remedial plants found all over the world, biogenic synthetic techniques are used to create CuO NPs for use in electrical, magnetic, device solar cell applications, biological, pharmacological, dermatological, energy, and catalytic applications [33-34, 95].

In this review paper, we investigated the potential of greenly synthesized CuO NPs as a catalyst for the degradation of several wastewater dyes as model environmental contaminants. We employed UV-visible light and several energy sources to determine which energy source works best in conjunction with CuO NPs to degrade dyes in a highly efficient manner. It has been specifically determined how pH, catalyst dosage, and initial dye concentration affect the interaction period. In both instances, an effort has been made to preserve the CuO NPs from the reaction combination and reprocess them for use in later procedures.

2. SYNTHESIS OF CuO NPs BY DIFFERENT METHODS

Top-down and bottom-up methods are the two main synthesis techniques for nanomaterials, as illustrated in Figure 1.

a. Size reduction top-down from bulk materials
b. Bottom-up: synthesis of materials starting at the atomic level

Various researchers have reported numerous methods for producing CuO NPs, including hydrothermal, biosynthesis techniques, electron beam lithography, solid-state reactions, sol-gel methods involving surfactants, microwave-assisted protocols, copper acetate decomposition, and sonochemical combination methods [35-38]. Additionally, it has been found that the way CuO NPs are made has an impact on both their morphological characteristics and toxicity behavior. Fig. 1 displays the flow chart illustrating the various processes used to create CuO NPs.

2.1. CuO NPs

Copper has the atomic number 29, an atomic mass of 63.54, and a density of more than 5 g/cm^{-3}. It possesses exceptional features such as excellent mechanical properties, malleability, high thermal and electrical conductivity, exceptional corrosion resistance, low chemical resistivity, and so on. Copper

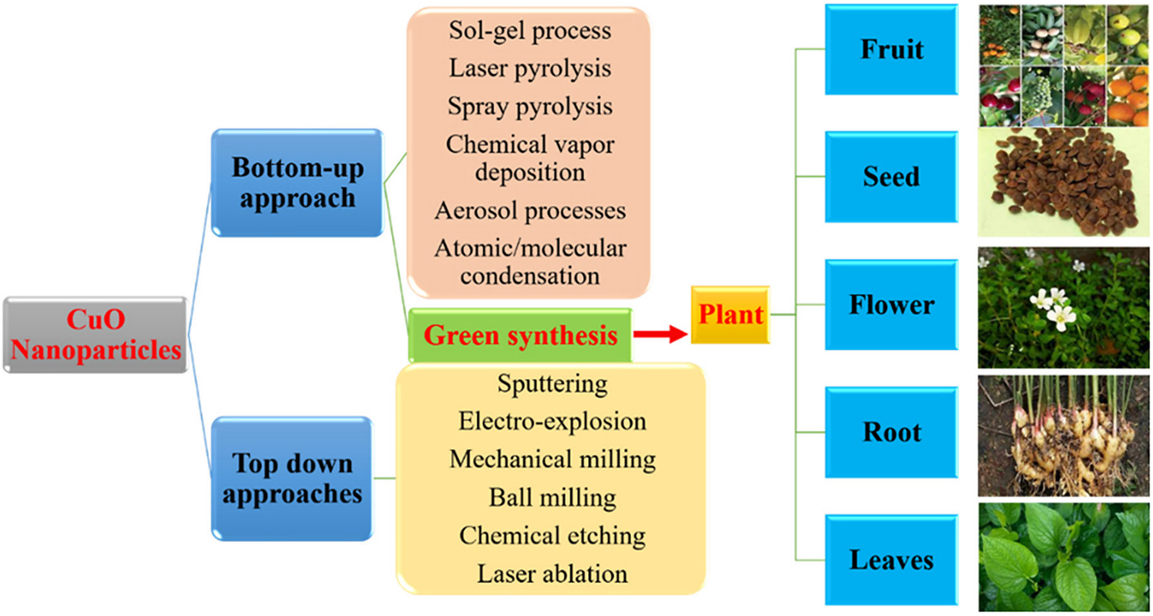

Figure 1. Top-down and bottom-up approaches that use different chemical and physical techniques to produce CuO NPs.

NPs are a potential material in several fields of science due to the qualities listed above [39]. Copper NPs are created in a variety of ways, including physical, biological, and chemical methods. However, the synthesis of copper NPs is far more complicated because copper NPs oxidize and agglomerate when they come into contact with air. To solve the aforementioned issue, copper NPs are synthesized in the presence of inert gas, or polymers, and surfactants are utilized as stabilizing agents during the synthesis of copper nanoparticles [40NPs [20].

2.2. Plants role in the green synthesis of CuO NPs

Plants include a vast variety of physiologically active chemicals; thus, most plants have a demonstrated track record for their antiparasitic, anticancer, anticarcinogenic, antimicrobial, and fungistatic characteristics [41]. CuO NPs are created by simply combining the metal solution with plant extract. NPs are formed in the media as a result of metal ion reduction. The organic hydrocarbon component assists in copper oxidation. Citron's acidic nature also limits copper oxidation because protons in the media affect copper electro-deposition at low pH levels. These biomolecules effectively decrease copper salts while avoiding aggregation. Phenolic substances hydroxyl and ketonic groups interact with metals and behave as catalysts. Flavonoids have the ability to directly scavenge molecular species of active oxygen [42]. The reduction of copper ions to form CuO NPs was linked to the presence of phenolic chemicals in the plant. The extract components are thought to act as both reducing and capping agents in the stability of produced CuO NPs as in Table 1.

2.3. Characterization techniques

Since the applications of CuO NPs are heavily reliant on their characteristics, their characterization is imperative. The particle and pore size, shape, geometric fractal dimensions, crystallinity, and surface area of produced CuO NPs can all be characterized using significant characterization approaches [54-55].

In this article, we go into great detail about the various techniques used to characterize CuO NPs. These methods can be used separately or in combination to study a specific property, depending on the situation. We compare each of these methods, evaluating things like their accessibility, expense, precision, non-destructiveness, ease of use, and affinity for particular compositions or materials. Despite the abundance of techniques presented here, each one is thoroughly studied. The size, morphology, and crystal structure of the nanomaterials can be determined using the optical probe, electron probe, scanning probe, photon spectroscopy, ions-particle

Plant	Precursor	Method	Size (nm), morphology	Application	Ref.
Centella Asiatica	Copper chloride di-hydrated	Sol-gel	12-18, Spherical	Photocatalytic and antibacterial activity	[43]
Punica granatum	Copper chloride	Co-precipitation	28-36, Spherical	Photocatalytic activity	[44]
Tinospora cordifolia	Copper nitrate trihydrate	Colloid-thermal	6-8, Sponge	Photocatalytic and antioxidant activity	[45]
Turmeric	Copper nitrate	Co-precipitation	30-80, Spherical & narrow	Photocatalytic and antiviral activity	[46]
Ferulago angulata (schlecht) boiss	Copper acetate	Microwave	44, Spherical & crystalline	Photocatalytic activity	[47]
Oak Fruit Hull	Copper acetate	Sol-gel	34, Quasi-spherical	Photocatalytic activity	[48]
Azadirachta indica	Copper nitrate trihydrate	Thermal decomposition	28-35, Spherical	Photocatalytic and antibacterial activity	[49]
Bauhinia tomentosa	Copper (II) sulfate	Electrochemical	22-40, Clustered & spherica	Photocatalytic and antibacterial activity	[50]
Euphorbia Chamaesyce	Copper chloride	Microwave	36-40, Spherical	cytotoxic activity	[51]
Rheum palmatum	Copper chloride	Colloid-thermal	10-20, Spherical	antioxidant activity	[52]
Gloriosa superba	Copper nitrate	Thermal decomposition	5-10, Spherical	Photocatalytic and antibacterial activity	[53]

Table 1. Several plant extracts employed in the synthesis of CuO NPs and their applications.

probe, and thermodynamics probe characterization techniques. Other methods, like magnetic methods, are tailored for particular classes of materials. Numerous other methods offer additional details about the structure, elemental make-up, optical characteristics, and other general and more focused physical characteristics of the nanoparticle samples. These methods include X-ray, spectroscopy, and scattering techniques, as examples.

This review is divided into sections that present a wide variety of unique characterization methods for NPs in relation to the properties examined (Figure 2). The sections are divided into the various technique groups as previously mentioned.

3. PHOTOCATALYTIC ACTIVITY

3.1. Classification of dyes

Dyes come in two main categories: (I) natural dyes and (II) synthetic dyes. Natural dyes are typically derived from herbal plant sources (root, stem, leaves, bark, and flowers), as well as from minerals and animal sources [56]. In addition, synthetic dyes can be divided into three groups: anionic dyes (which are water-soluble, and include acid, direct, and reactive dyes), cationic dyes (basic dyes), and nonionic dyes (dispersing, pigment, and solvent dyes) are shown in Figure 3 [57]. Most of cationic dyes are hazardous, including both humans and aquatic ecosystems, making them more poisonous than anionic dyes [58].

3.2. Removal of dyes from wastewater

The p-type semiconductor CuO NPs have several benefits, such as high UV-Vis light exposure with an energy band gap of 1.2-1.5 eV, improved quantum efficiency, inexpensive, nonhazardous, excellent optical absorption capabilities, and wide availability [59]. In addition, when CuO NPs is used as a photocatalyst for the breakdown of organic dyes, they primarily degrade or disable through a photocatalytic in UV-Vis light and sunlight process. Table 2 and 3 represents the degradation of various dyes for pure and green synthesized CuO NPs, respectively.

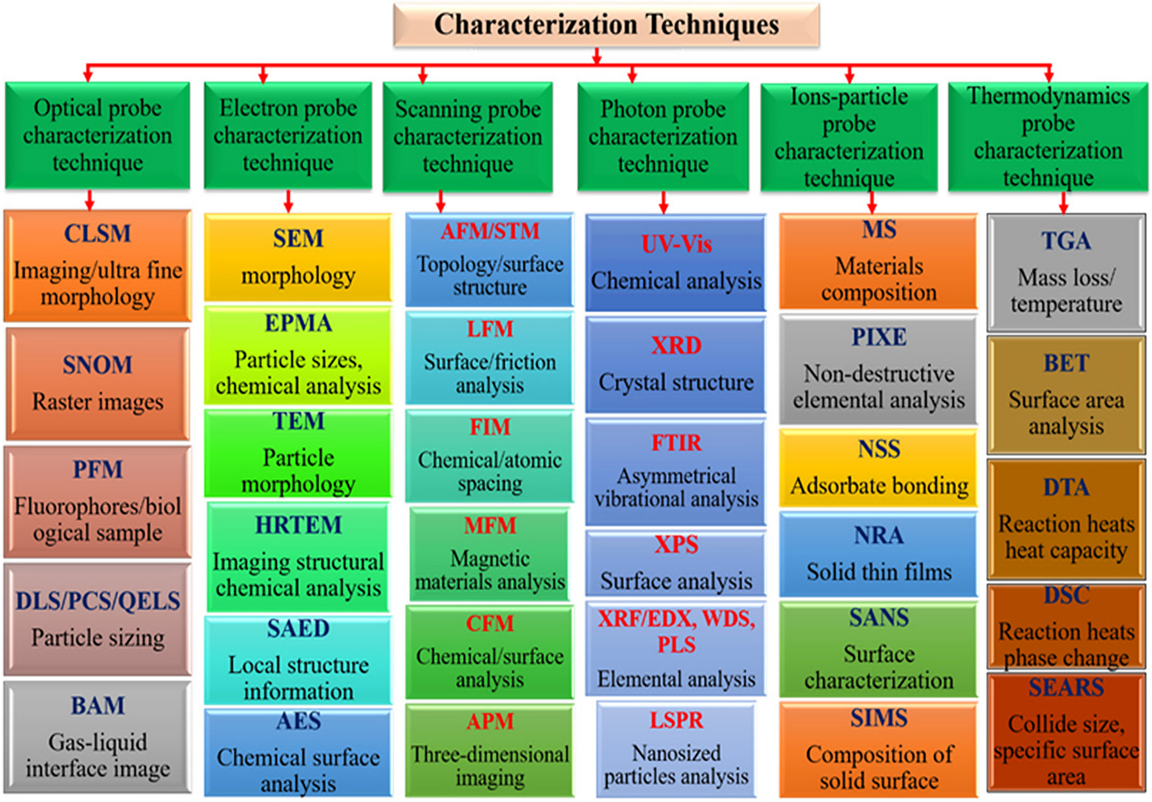

Figure 2. Various characterization techniques for CuO NPs.

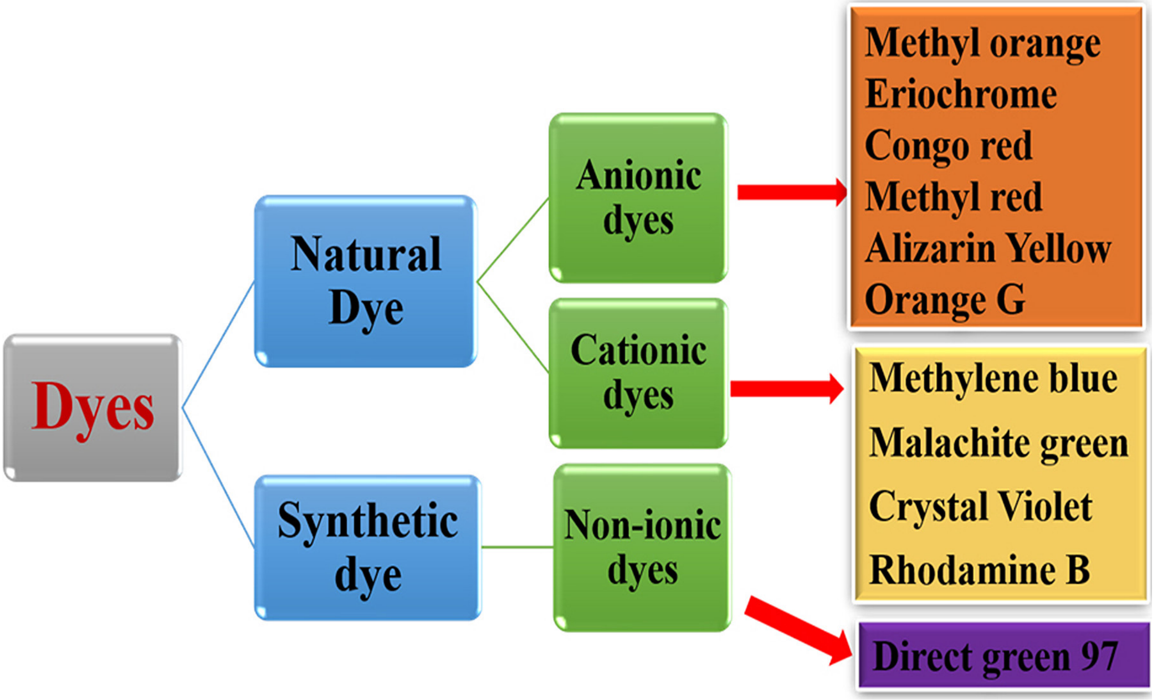

Figure 3. Classification of dyes.

Types of dye	Source condition	Time period (min)	Degradation efficiency (%)	References
Methyl violet	UV radiation	90	89	[60]
Methylene blue	Visible light	120	78	[61]
Eosin-Y	Visible light	180	92	[62]
Coomassie brilliant blue	UV radiation	40	94	[63]
Malachite green	Sunlight	60	78	[64]
Crystal violet	Visible light	80	48	[65]
Congo red	Sunlight	120	66	[66]
Rhodamine B	Visible light	150	93	[67]
Methyl orange	UV radiation	75	99	[68]
Brilliant cresyl blue (BCB)	Visible light	100	97	[69]

Table 2. Photocatalytic activity of pure synthesized CuO NPs.

Plant	Types of dye	Source condition	Time period (min)	Degradation efficiency	References
Rhazya stricta.	Methylene blue	Solar irradiation	120	83	[71]
cedrus deodara	Methylene blue	Sunlight irradiation	160	73/94	[72]
Celastrus paniculatus	Methyl orange, Methylene blue	Visible light	120	90/92	[73]
Capparis decidua	Trypan blue	UV radiation	180	99/89	[74]
Carica papaya L.	Methyl orange	UV radiation	160	66	[75]
Jatropha curcas	Rhodamine B, methylene blue	Visible light	85	75, 82	[76]
Oak Fruit Hull	Crystal violet	UV radiation	90	86	[77]
Caesalpinia bonducella	Methyl orange	Visible light	100	93	[78]
Banana peel	Congo red	Sun light	120	89	[79]
Jatropha curcas	Methylene blue	UV radiation	85	90	[80]
Areva Lanata	methylene blue	Sun radiation	90	79	[81]

Table 3. Photocatalytic activity of green synthesis CuO NPs.

3.3. Factors influencing affecting the photocatalytic degradation of dyes by photocatalysis

3.3.1. pH effect

Due to its numerous many responsibilities, including electrostatic interactions between the photocatalyst external surface, solvent particles molecules, adsorbent, and charged radicals created during the reaction procedure, pH impacts on the dye photodegradation method are particularly important. The charge on the catalyst surface and the charge on the dye molecules typically cause pH to have an impact on photocatalysis, increasing or decreasing the activity. The photodegradation efficiency was therefore 96.26% at pH 2, whereas the MB dye efficiency enhanced to 98.71% at pH 4 [82]. However, the photodegradation activity decreased as the pH level rose higher. The interference between the negatively charged dyes and the positively charged catalyst surfaces (NPs) in the acidic solution (pH ¼ 3), increases removal efficiency. Increased dye adsorption occurred as a result of a cumulative increasing electrostatic interface interaction between the positively charged dye and the negatively charged catalyst when the dye solution became more basic (pH > 7) [83].).

3.3.2. Catalyst load

According to research, the amount of catalyst loading significantly affects photocatalytic activity. When the catalyst (CuO NPs) concentration was increased from 2 to 8 mg/mL, MB degradation rose from 40 to 96%. The photodegradation activity did not increase further when the catalyst concentration was increased from 8 to 12 mg/mL [84]. The particles are small and widely scattered at lower concentrations. This increases the active sites and surface area, which boosts the NPs' ability to photodegrade dye. The catalyst accumulated and dissolved, which caused a rapid decline in photodegradation effectiveness. Additionally, at larger NP catalyst loading, the suspension becomes opaquer and more turbid, which causes light scattering. As a result, less radiation is able to pass through the material (reaction mixture) [85].

3.3.3. Temperature, time and morphology

Temperature, duration, and shape all have a significant impact on photocatalytic performance are used green-synthesized CuO NPs in the temperature range of 25-40 ºC to demonstrate that temperature affects the rate of substituent breakdown. According to the degradation efficacy values, a minor increase in reaction rate is brought on by a rise in temperature. The moderate increase in photocatalytic degradation caused by raising the temperature may be caused by an increase in molecular collision frequency [86]. Higher temperatures may also cause the contaminated molecules to turn into disabled from the catalyst surface, reducing the rate of reaction. The removal of oxygen from the reaction mixture, alternately, is caused by intense temperatures and is essential for the oxidation of contaminants [87].

3.3.4. Mechanismof photocatalytic degradation

The main aspect of photocatalysis is a photogenerated chemical reaction based on the interaction of photons with CuO NPs. A semiconductor photocatalyst appears to have a very straightforward photocatalytic process. The reaction mechanism, though, consists of intricate pathways that can be thought of as a succession of redox reactions. The hypothesized photocatalytic mechanism of a CuO material when exposed to light. It's significant to note that the photoactivity changes depending on the materials utilized in the specific system [88]. Figure 4 illustrates the various steps elaborate in a photocatalytic reaction using a CuO photocatalyst.

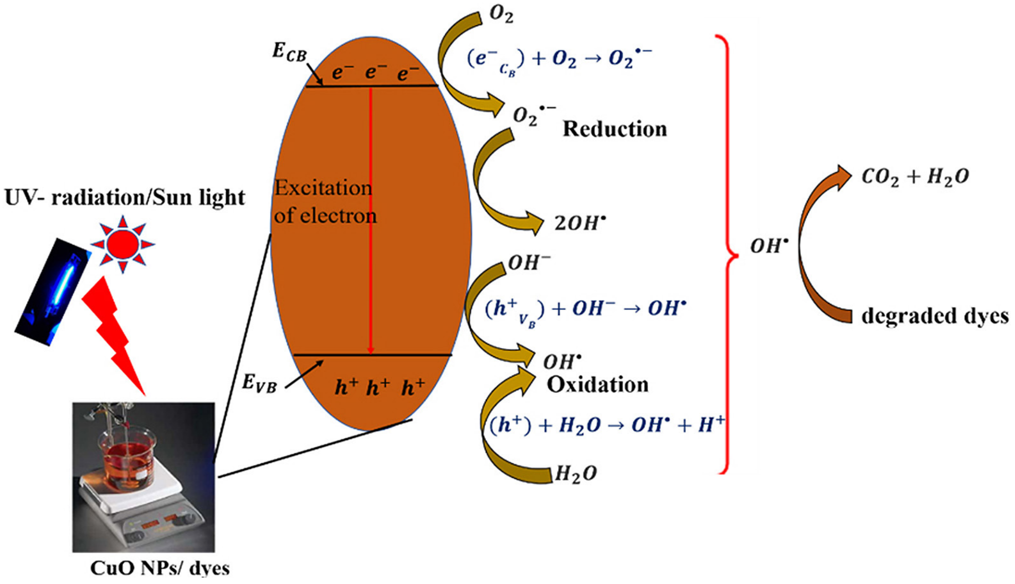

Figure 4. Photocatalytic mechanism for CuO photocatalyst.

The photogenerated electrons are excited from the tied-up valence band to the vacant conduction band when the CuO NPs photocatalyst is exposed to light of the appropriate wavelength. As a result, an electron-hole pair (e^-, h^+) is created, departure a hole in the valence band [89-90].

$$Cu + hv \rightarrow \text{Cu}\ e^-_{CB} + h^+_{VB} \tag{3}$$

wherever, h^+ and e^- are the two main oxidizing and reducing agents. Although they do so via various chemical paths, both the electron and the hole contribute to photodegradation. The degrading reaction sequences for the particle reaction path are as follows:

$$h^+ + OH^- \rightarrow OH^\bullet \tag{4}$$
$$OH + reactant \rightarrow Oxided\ products \tag{5}$$

The hydroxyl ion can interact with a photogenerated hole in an aqueous environment to produce a hydroxyl radical Eq. (4). Hydroxyl radicals function as exceptionally potent oxidants in aqueous settings, according to reports in the literature [91-92]. As a result, the reactant can be openly oxidized by the hydroxyl radicals to produce other safe products Eq. (5). The degrading reaction chains for the electron-driven route can be explained by

$$e^- + O_2 \rightarrow O_2^{\bullet -} \tag{6}$$
$$O_2^{\bullet -} + H^+ \rightarrow HO_2^\bullet \tag{7}$$
$$HO_2^\bullet + HO_2^\bullet \rightarrow H_2O_2 + O_2 \tag{8}$$
$$H_2O_2 + e^-\ CB \rightarrow OH^\bullet + OH^- \tag{9}$$
$$Reactant + O_2 \rightarrow degradation\ products \tag{10}$$

Superoxide O_2^- Eq. (6) is produced when conductance band electrons produced by photons interact with electron acceptors like oxygen. The producing of hydrogen peroxide Eq. (8), predominantly through the reduction of adsorb oxygen, should be possible according to thermodynamic theory given the oxidation-reduction potential of the CuO NPs electron/hole pair [93-95, 97]. By interacting with oxygen and hydroxyl radicals, the reactant is effectively degraded in Eq. (10).

4. CONCLUSION

The various copper precursors for copper NPs were reviewed in this review, and investigations have revealed that different plants, sizes, morphologies, stability, and characteristics are sensitive to diverse conditions. Biological methods are most commonly utilized for the preparation of NPs. Potential applications for plant-mediated NPs include renewable energy, healthcare, cosmetics, medicines, and other essential goods. Future medical and industrial items could make extensive use of metallic NPs synthesized by plants. In this article, CuO NPs that can be used in wastewater treatment are reviewed. Diverse techniques, such as ion exchange, coagulation/flocculation, aerobic degradation, anaerobic degradation, ozonation, and photocatalytic processes, have been utilized to remove organic color contaminants from wastewater. The use of photocatalysis with NPs as an alternative to physical and chemical processes is one of these strategies that have the most potential. Nano catalysts are also reasonably priced, chemically stable, environmentally friendly, and quickly oxidize. Nowadays, it looks highly optimistic that scientists will be able to resolve current environmental, social, and industrial issues based on a better knowledge of the effects of NPs size and shape and their interactions with support materials or stabilizing agent. The combination of Cu metals on green-synthesized photocatalysts was studied in this review as a prospective alternative for better efficient photocatalytic degradation.

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Conflict of Interest

The authors declare no competing financial interest.

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