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Utilization of Tin Oxide Nanoparticles Synthesized Through Plant-Mediated Methods and Their Application in Photocatalysis: A Brief Review

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Abstract

This article emphasized sustainable methods using natural resources in producing SnO_2 NPs and how it is employed as an inexpensive and reasonable photocatalyst to degrade dyes pollutant. Green synthesis of SnO_2 NPs using plant extracts is known to lower the consumption of hazardous chemicals or energy. Phytochemicals from plants serve as the capping and reducing agents during the synthesis process, and subsequent heating treatment continued the procedure to furnish SnO_2 NPs. It was discovered that pure SnO_2 NPs synthesized from different plants possess a distinctive band gap value, that react differently for the kinetic reaction. However, all of the photocatalyst are successfully involved in the irradiation of light to this semiconductor, which leads to photo-oxidation activity generating electrons and holes. This resulted in the redox reaction which serves for the degradation of dye molecules, whereby all of them gave different degradation percentages in agreeable values.

Keywords: Tin oxide nanoparticles, Green synthesis, Band gap value, Photocatalytic

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1. Introduction

The superiority of nanotechnology applications is acknowledged by humans owing to nanomaterials' excellent physicochemical properties. They are known to have excellent stability and porosity, which is proven to exceed their traditional material [1]. According to Singh et. al (2019), nanomaterials are broadly developed for many valuable technologies, for instance, electronics, optics, catalysis, space, energy, and biomedical science [2]. At this juncture, a kind of nanomaterial identified as nano metal oxides was found to have significant potential applications, because of its high specific surface area to volume ratio and its distinctive structural characteristics. Among these nanomaterials, tin oxide nanoparticles (SnO_2 NPs) has gained a lot of attention due to their unique electrical and optical feature [3]. This n-type semiconductor (SnO_2 NPs) serves a wide band gap of 3.6 eV, besides having good chemical stability, hydrogen production, low resistivity, and high transparency [4].

To date, the advancement of SnO_2 NPs has been embedded in the application of solar cells, gas sensors, lithium-ion batteries, electrodes, catalysts, medical, energy storage and coatings [5-12]. Sagadaven (2021), Satinder (2015), and Rana et. al (2020) described that the plant extracts had been utilized for the synthesis of SnO_2 NPs under green synthesis protocol, either from leaves, fruits, flowers, seeds, or barks section. Notably, three criteria play a critical role in the green synthesis approach: a non-toxic solvent, an excellent reducing agent, and a non-hazardous material for stabilization. The interesting part of this method is that it offers wide availability of raw materials, is safer, biocompatible, sustainable, requires less energy, is rapid, easy to handle, and affordable. Moreover, raw material contains various phytochemicals employable as reducing and capping agents. The phytochemicals such as flavonoids, polyphenols, alkaloids, glucose, terpenoids, and tannins are reported to be responsible for reducing Sn^{4+} or Sn^{2+} from precursor salt to Sn^0.

They act as the stabilizing agent in retaining the reactivity of the SnO_2 NPs. Several studies have been performed using a range of plant extracts for example *Vitex altissima (L.), Aspalathus linearis, Ziziphus jujube, Daphne mucronate, Aquilaria malaccensis, Citrus aurantifolia, Annona squamosa, Pandanus amaryllifolius, Tradescantia spathacea, Carica papaya, Cyphomandra betacea, Litsea cubeba, Chromalaena odorata, Pruni spinosae, Psidium guajava, Stevia rebaidiana* and many more [13-29]. Previously, the implementation of conventional methods to synthesize SnO_2 NPs were reported to lead to several shortcomings. This can be seen in the use of toxic chemicals and solvents, the requirement of expensive instruments, and high energy, as well as generating ecological contamination. These unfavorable methods are known as sputtering, chemical vaporization, ultrasonication, thermal decomposition, spray pyrolysis, laser irradiation, and electrolysis [30-32]. Thus, by having green synthesis as an alternative method, the production of SnO_2 NPs is in line with the implementation of the Sustainable Development Goal agenda. The unique properties own by SnO_2 NPs make it as the favorable photocatalyst for the green and eco-friendly treatment of dye-polluted water under photocatalytic activity [33]. The photocatalytic process is proven to be efficient and practical under mild conditions, and more significantly, it serves a lot of catalytically active sites [34].

Regarding the issue of dye-polluted water, the aquatic system currently contains many disposal products containing dyes, paper, plastics, food and drinks, textiles, and such. These materials later cause the accidental release of dye molecules after exposure to the water system. Since dyes may accumulate in specific tissues and organs and lead to various illnesses even at low concentrations, dye-polluted water is recognized as a significant concern worldwide since it also disrupts aquatic ecosystems [35]. For instance, dye molecules will block sunlight and then reduce the rate of the photosynthetic process in aquatic plants. The expected growth of aquatic animals will face irregularity by the decreasing oxygen capacity of water. Some disorders such as gastrointestinal, anemia, and skin and bladder irritation are common diseases caused by the uptake of dye-polluted water [15]. This scenario involves the presence of highly stable and soluble dye molecules in water, making them one of the non-biodegradable moieties, making it difficult to remove or degrade [36]. Besides, the complex structure of dye molecules consisting of aromatic rings is another reason for the difficulty [37]. Treatments such as ultrafiltration, reverse osmosis filtration, Fenton oxidation and ozonation did not work out very well despite the use of many chemicals, plus generating an unwanted secondary pollutant. During the photocatalysis process, in general, the catalytic mechanism is based on the creation of electron-hole pairs through band gap energy. Light will be absorbed by photocatalysts referring to SnO_2 NPs and transformed it to higher energy, followed by the subsequent transfer to the pollutant molecule [38]. In this article, we tried to describe the recent advances of green synthesized SnO_2 NPs by reviewing the utilization of several plant extracts to mediate the formation of SnO_2 NPs as the photocatalyst. The mechanism that pertinent to kinetics order, and the respective catalytic performance based on of green-synthesized SnO_2 NPs from different plants in degrading the organic dyes were also be discussed. This article could be the supporting document to prove that the green-synthesized SnO_2 NPs possess high potential to be used as the photocatalyst based on the energy potential represented by the energy band gap, as well as its degradation capability from the experimental quantification.

2. Results and Discussions

2.1. Green Synthesis of SNO_2 NPs using plant extracts

Plants are the unique creation in earth, whereby they can easily convert light energy into chemical energy, consumed, accumulated, employed and recycled various minerals by their own capability as explained by Oza et. al (2020) [39]. Apart from this, plants are brilliant resources of renewable and sustainable materials for the green synthesis of nanoparticles. The plant extract's phytochemicals are highly associated with the donation of electron-proton species to mediate green synthesis. The nucleophilicity effect carried by phenolic rings facilitates them in metal-chelation activity, resulting in efficient action in reducing, capping, and stabilizing actions. Phytochemicals were also discovered to be responsible for controlling the nanoparticles' size and morphology [40-41]. Having this advantages attribute, they are strongly recommended as the green tool. Besides using plants extract as the green synthesis tool, other examples are by biological means such as bacteria, fungi and algae but they seem to be less favorable since the process will involve tedious handling and storage in contrast when using plants extracts. The stages involved in the green synthesis of SnO_2 NPs are relatively straightforward. The first step is the addition of the prepared plant extract into precursor salt solution, which is tin chloride, followed by stirring. In general, all these plant samples are fresh before subjected to drying process and showed particular color change before collection. During the extraction process, most of the solution turned to be in green or brownish solution before being added to precursor salt solution. After stirring, the formation of the precipitate or gel will occur, and it will be collected before thermal treatment. The illustration of the process is shown in **Figure 1.**

2.2. Probable mechanism of green synthesis of SnO_2 NPs

During the formation of SnO_2 NPs using plant extract, three crucial phases are involved in the mechanism: activation, growth, and termination. The illustration is displayed in **Figure 2** [42]. The activation phase relates to reducing the cation from the metal precursor resulting in nucleation afterwards. The second phase, which is growing, is an occurrence of the tiny nanoparticles merging voluntarily into bigger particles, which heightened the thermodynamic stability of nanoparticles. This phenomenon is known as Ostwald ripening. The final phase, the termination, involved the finalized tuning of the size and shape, whereby the help of the stabilizing effect from the phytochemicals (Flavonoids, alkaloids, glucose, terpenoids, and tannins etc.) furnished the most energetically preferable orientation and conformation of nanoparticles.

2.3. Mechanism of photocatalytic activity by SnO_2 NPs

The lower energy band, which refers to the conduction band (CB), is filled with electrons for semiconductors material. While the upper band, which is the valence band (VB), is electrons-free.

The electronic characteristic and performance of the particular material originally come from this energy difference between these two bands, known as the energy band gap (E_{bg}). In addition, the band gap value will define the material's applicability as a semiconductor. SnO_2 is considered an ideal material for photocatalysis since it has a large band gap energy of 3.6eV, which relates to photoactivation within the UV range of the electromagnetic spectrum [43]. The employment of metal-semiconductors in photocatalysis signifies the ecologically friendly method, in which the activity is fastened after being introduced to light. Regarding this, metal oxides would generate an electron-hole pair and break down the dye molecules into degraded products under a redox reaction [44]. Throughout the photocatalysis process, the dye degradation rate is associated with the production of the light-dependent free radical on the surface of the photocatalyst. When the SnO_2 NPs surface is irradiated with appropriate light energy or higher than its band gap energy, migration of electrons occurs, generating holes (h^+) in the valence band and an electron (e^-) in the conduction band (**Equation 1**). The redox reaction is performed by the action of the holes (h^+) as an oxidizing agent and oxidize the dye directly or react with H_2O giving hydroxyl radicals (OH^-) (**Equation 2**). Likewise, the reducing agent represents by the electrons (e^-) in the conduction band to reduce the O_2 adsorbed on the SnO_2 surface to give superoxide radical (O_2^-) (**Equation 3**). The key parameter during this light-dependent excitation of SnO_2 NPs is the generation of two species; superoxide ($O_2^{\cdot-}$) ions and hydroxyl (OH^-) radicals, whereby they initiate the photo-oxidation by reacting with the dye molecules as illustrated in **Figure 3** (**Equation 4**) [45-46].

$$SnO_2 + h\nu \rightarrow SnO_2\,(e^-) + SnO_2\,(h^+) \ldots\ldots (1)$$
$$h^+_{VB} + H_2O \rightarrow OH^- \ldots\ldots (2)$$
$$e^-_{CB} + O_2 \rightarrow O_2^- \ldots\ldots (3)$$
$$O_2^- \text{ or } OH^- + \text{Dye molecules} \rightarrow \text{degraded product} \ldots (4)$$

2.4. Degradation of dyes by SnO_2 NPs photocatalyst

Several reports described the degradation efficiency using green synthesized SnO_2 NPs. Here, SnO_2 NPs synthesized using various plant sources have proven to be an efficient photocatalyst for degrading dye pollutants under irradiation of sunlight or UV. A study by Bhosale et al. utilized an aqueous leaves extract of *Calotropis gigantean* mixed with $SnCl_2$ to give a precipitate that was further annealed at 300°C. A rutile structure of irregular shape of SnO_2 NPs with a crystallite size of 35nm was obtained. The energy band gap signified as 3.1eV in degrading 80% of Methyl Orange [47]. *Camellia sinensis* leaves extract and $SnCl_2.2H_2O$ was used by Luque et al. which experienced a thermal bath at 60°C within 12 hours. The calcination for the obtained pellet was carried out at 400°c for 1 hour, giving a tetragonal rutile with a crystal cluster of SnO_2 NPs within 6 to 10 nm. The band gap calculation discovered values of 4.02, 3.95, and 3.79 eV and they are being tested under a photocatalytic reaction to completely degraded Methylene Blue and Rd-Bdye, and 81% of Methyl Orange [48]. A quasi-spherical of SnO_2 NPs having sizes 3 and 6 nm with a tetragonal rutile phase was obtained by Osuntokun et al. after synthesizing leaves extract of *Brassica oleracea L. var. botrytis* with $SnCl_2.2H_2O$. The synthesis process was heated at 60°C for 6 hours, with subsequent annealing at 300°C and 450°C of the precipitate. The pure SnO_2 NPs discovered to possess band gap values of 3.9 to 4.3 eV that degraded 91% of Methylene Blue [49]. According to Fatimah et. al, the synthesis of *Pometia pinnata* leaves extract with $SnCl_2.2H_2O$ produced flower-like SnO_2 NPs. The procedure was held under reflux for an hour. The calcination of the powder was conducted at 500°C for 2 hours to give a tetragonal structure with sizes within 8 to 20nm, signifying a band gap value of 3.5eV which degraded almost 99% of Bromophenol Blue [50]. Leaves extract of *Delonix elata* was mixed with tin chloride solution, as explained by Suresh et al., before being treated to different procedures, which were wet-chemical, sonication, and microwave at a similar calcination temperature of 400°C for 2 hours. A foam shape of SnO_2 NPs was obtained with crystallite sizes of 5 to 7nm, indicating band gap values of 3.80, 3.89, and 3.91 eV to result from photocatalytic degradation of Rhodamine-B at 82%, 85%, and 92% [51]. Wicaksono et al. clarified the synthesis process using leaves extract of *Amaranthus tricolor L.* with $SnCl_4.5H_2O$ at 80°C within 1 hour, followed by calcination of the obtained powder at 400°C for 2 hours. The procedure gave SnO_2 NPs in the polycrystalline phase, favoured to be spherical with a size less than 20 nm with a band gap of 3.52 eV to degrade about 99% of Bromophenol Blue [52]. Green synthesis using *Citrus aurantifolia* fruit extract and $SnCl_2.2H_2O$ was described by Luque et al. at 60°C. Later, the obtained paste was calcined at 400°C for 1 hour to give a tetragonal structure with a quasi-spherical shape of SnO_2 NPs in size between 5nm to 9nm. The band gap value was discovered as 3.02 to 3.44 eV, which degrades about 96% of Methylene Blue [53]. Garrafa-Galvez et al. employed the fruit extract of *Lycopersicon esculentum* with $SnCl_2.2H_2O$ to obtain pellets that calcined at 400°C for 1 hour, giving a quasi-spherical formation with 4 to 5.5nm crystallite size. The calculated band gap was obtained as approximately 3.3 eV, and the synthesized SnO_2 NPs were found to be efficient in removing all Methylene Blue entirely [54]. Another fruit extract, *Actinidia deliciosa* (kiwi) was reported to be mixed with $SnCl_4.5H_2O$, which was heated at 60°C for 2 hours duration as described by Gomathi et. al [55]. The synthesis obtained pellet that was further exposed to air-dried overnight to furnish spherical SnO_2 NPs with rutile phase within size 5 to 10 nm, indicating a band gap value of 3.96 eV. From the photocatalytic testing, SnO_2 NPs were found competent to degrade 89% Methylene Blue, 87% Methyl Orange, and 97% Rhodamine-B accordingly. Production of pure SnO_2 NPs was conducted by using a mixture of *Trigonella foenum-graecum* seed extract and $SnCl_2.2H_2O$ as described by Goyal et al. The synthesis was carried out at 80°C for 4 hours, followed by the calcination of the jelly product at 400°C within 4 hours. The procedure furnished a 10 nm spherical shape of SnO_2 NPs and indicated 2.6 eV as the band gap value, which was photo catalytically tested to degrade 100% Coralene Red [56]. Shamima Begum and Md. Ahmaruzzaman Begum in their work, explained the synthesis of SnO_2 NPs using *Parkia speciosa Hassk* and $SnCl_4.5H_2O$ under microwave irradiation to give precipitate that was finally heated in the oven at 60°C within 12 hours. The band gap was obtained as 4.3eV, facilitating about 98% degradation of Acid Yellow 23 dye [57]. The efficiency evidence of the green synthesized SnO_2 NPs in the degradation process using a variety of dyes is summarized in **Table 1.**

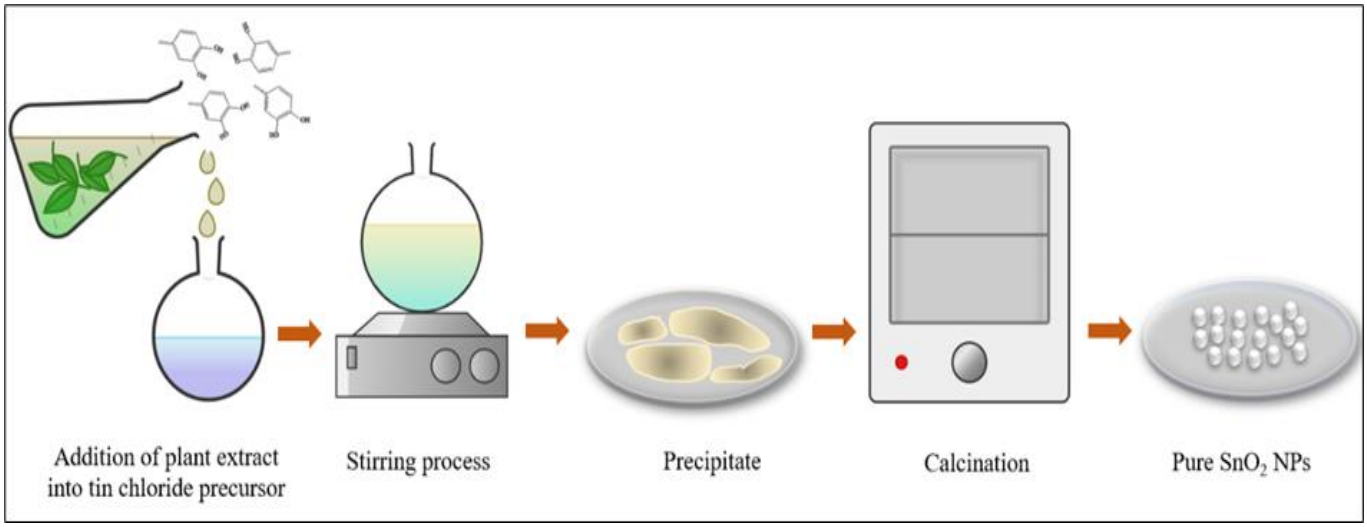

Fig. 1. General procedure for the production of SnO_2 NPs using plant extract [21].

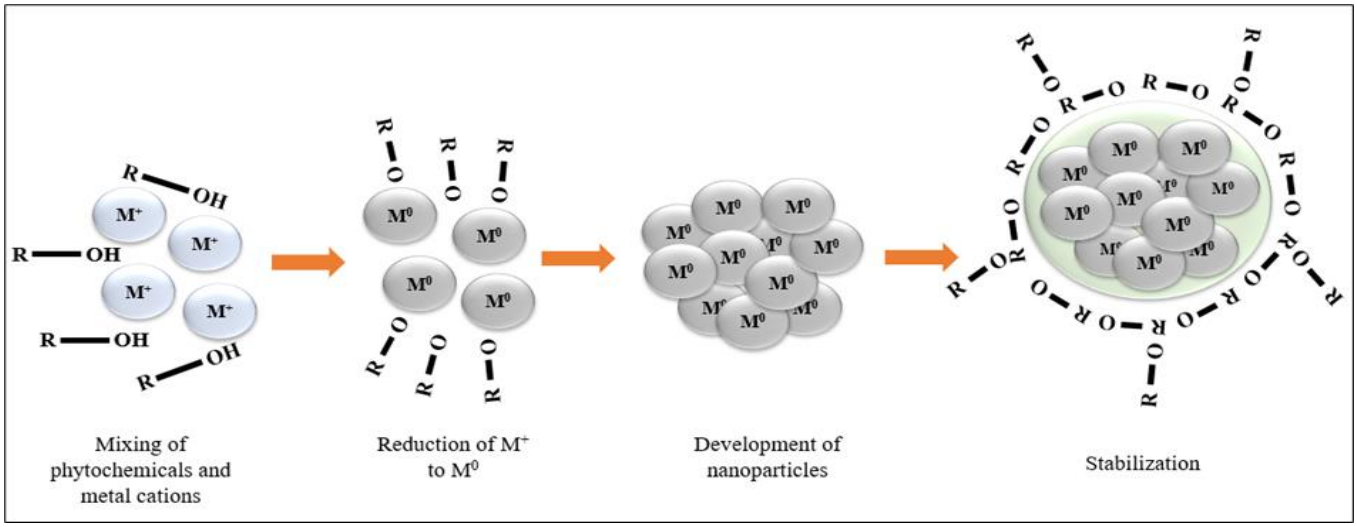

Fig. 2. The synthesis of SnO_2 NPs using phytochemicals from plant extracts [42].

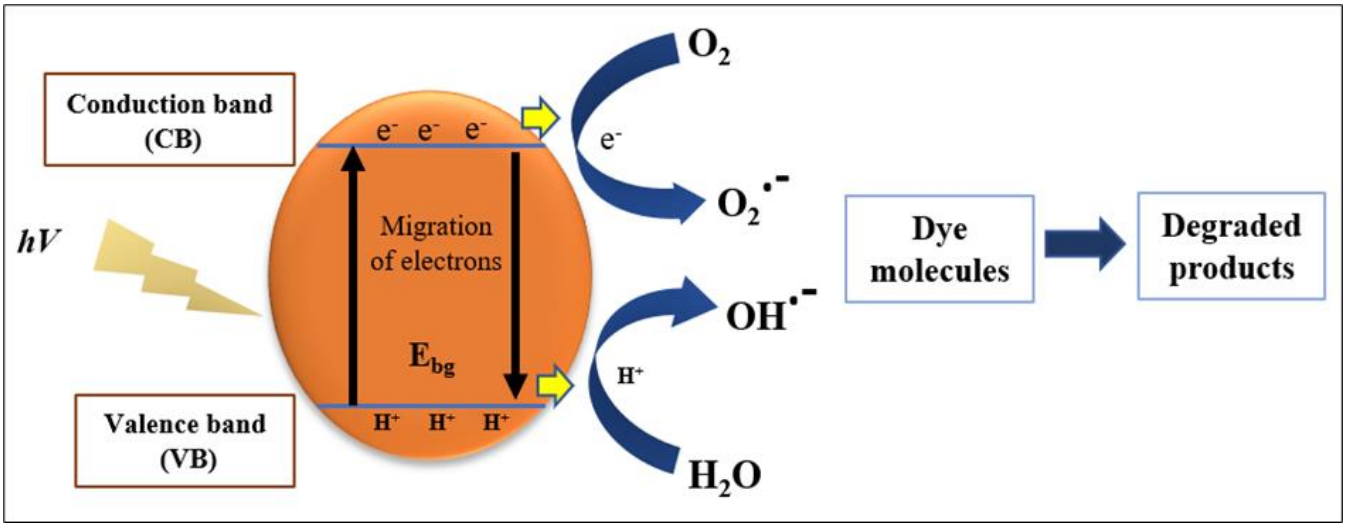

Fig. 3. General reaction mechanism of SnO_2 NPs performing photocatalytic [45-46].

Table 1: The green synthesized SnO_2 NPs using various plants and their performance on dyes degradation.

No.	Plant	Band gap value (eV)	Degradation of dyes	Ref.
1	*Calotropis gigantea*	3.1	80% ; Methyl Orange	[47]
2	*Camellia sinensis*	3.79 to 4.02	100% ; Methylene Blue 81% ; Methyl Orange 100%; Rhodamine-B	[48]
3	*Brassica oleracea L. var. botrytis*	3.9 to 4.3	91% ; Methylene Blue	[49]
4	*Pometia pinnata*	3.5	99% ; Bromophenol Blue	[50]
5	*Delonix elata*	3.80-3.91	82% to 92% ; Rhodamine-B	[51]
6	*Amaranthus tricolor L*	3.52	99% ; Bromophenol Blue	[52]
7	*Citrus aurantifolia*	3.02 to 3.44	96% ; Methylene Blue	[53]
8	*Lycopersicon esculentum*	3.3	100% ; Methylene Blue	[54]
9	*Actinidia deliciosa (Kiwi)*	3.96	89%; Methylene Blue 87 % ; Methyl Orange 97 % ; Rhodamine-B	[55]
10	*Trigonella foenum-graecum*	2.6	100% ; Coralene Red	[56]
11	*Parkia speciosa Hassk*	4.3	98 %; Acid Yellow 23	[57]

3. Conclusions

This article overviews the green synthesis of SnO_2 NPs as safe, low-priced, environmentally friendly, trustworthy, and pertinent for photocatalytic applications. The synthesized SnO_2 NPs mediated by different plants possess different band gap values, thus resulting in different degradation degrees for the dyes. The irradiation of light with appropriate frequency under certain band gap energy would generate superoxide and hydroxyl radicals at the surface of SnO_2 NPs, thus facilitating the redox reaction in breaking down the pollutant molecules, resulting in a bi-product that is harmless to the human and ecological system. Under this photocatalytic mechanism, the green synthesized SnO_2 NPs successfully degraded several types of dye pollutants within the range of 80% up to 100%, which proved to be an efficient and versatile photocatalyst.

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