**Review**

**The Influence of Synthesis Methods and Experimental Conditions on the Photocatalytic Properties of SnO\(_2\): A Review**

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1. Introduction

For decades, the world has been affected by serious environmental problems, especially water resources that have been polluted over the years. In particular, the disposal of domestic-industrial effluents in water bodies has been a major problem for modern society [1–5]. In this sense, the research community, public agencies, and environmentalists have been very concerned about this issue. Among the contaminants commonly present in water bodies, persistent organic pollutants (POPs) are considered one of the main substances. POPs are heterogeneous synthetic compounds based on carbon from natural or from anthropogenic origins, with a high chemical resistance to degradation in the environment. These substances are characterized by a high level of toxicity and can be disruptive, neurotoxic, or immunosuppressive [6–9]. The excessive presence and accumulation of POPs in food chains has a major impact on human health and well-being. For instance, Mansouri and Reggabi [10] reported that the exposure to endocrine disruptors, and especially POPs, may contribute to the development of type 2 diabetes mellitus. According to these authors,
diabetic subjects had higher plasma concentrations of POPs than non-diabetics, and environmental exposure to some POPs is associated with an increased risk of type 2 diabetes in the studied samples. In addition, authors have stated that the exposure to some other POPs may increase health problem risks [11,12].

The elimination, restriction, and control of POPs took place first in 2001, after the Stockholm Convention [13]. Currently, more than 35 (thirty-five) POPs are reported in literature, such as polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDTs), and hexachlorocyclohexane isomers (HCHs), which are general types of drugs and medicine, pesticides, and synthetic dyes [14–17]. In this context, the search for new methodologies to promote full POP removal and degradation from the environment has attracted too much attention of the scientific community as well as public agencies, but it has also been a challenge to be faced today [18]. Different physical, chemical, and biological methods are used for degrading and removing pollutants present in water, air, and soil, among which adsorption [19,20], electrocoagulation [20], and advanced oxidative processes (AOPs) [21,22].

Among the diversity of water treatment methods, AOPs, particularly, represent a group of techniques characterized by the generation of highly oxidizing agents as free radicals. AOPs stand out due to their high efficiency in the degradation of numerous organic compounds, such as pesticides, surfactants, chlorophenols, and benzene, among others. APOs can promote mineralization of pollutants by transforming them into carbon dioxide, water, and inorganic anions [23–25]. AOPs are divided into homogeneous and heterogeneous, as well as photochemical and non-photochemical, processes [26]. Particularly, the heterogeneous photochemical process known as heterogeneous photocatalysis is one of the most important AOPs techniques used nowadays for being considered a promising technique for the treatment of domestic-industrial effluents and environmental decontamination [23,24,27–29]. In this process, semiconductors are irradiated by a light source (UV and visible light or even sunlight) with energy greater than or equal to its bang gap energy ($E_g$)—the minimum energy required to excite electrons from the lower energy band (valence band, VB) to a higher energy band (conduction band, CB). Thus, the energy absorption results in electronic excitation from VB to the CB, which leads to the formation of photoinduced charge pairs ($e^-/h^+$). Electron–hole pairs present very positive electrochemical potentials, which, when generated, give the semiconductor its redox properties [27,30].

During photocatalysis, photogenerated $e^-/h^+$ can migrate to the surface of the material and interact with an adsorbed species; in addition, they can be captured in intermediate energy states, or undergo recombination. When $e^-$ are in the BC, one of the very important reactions can occur, which is the reduction of the adsorbed O$_2$ on the catalyst surface to superoxide ($^•$O$_2^-$) radicals, avoiding recombination of electrons and the hole. This results in the accumulation of an oxygen radical species that can participate in photocatalytic reactions. On the other hand, when $h^+$ from the VB migrates to the catalyst surface, it can react with adsorbed H$_2$O to generate hydroxyl ($^•$OH) radicals, reacting with the pollutants to be degraded. It is worth mentioning that electronic recombination does not favor photocatalysis, because if the $e^-$ and $h^+$ recombine, the formation of the photogenerated $e^-/h^+$ pairs fails to participate in the oxidation-reduction process and formation of free radicals in the process [30–32]. An illustration of the general mechanism involved in photocatalysis is given in Figure 1, as also reported in [33].

A great variety of semiconductors have been playing an important role in the heterogeneous photocatalysis for energy production from water splitting [34–36] and for environmental remediation toward the degradation of different organic pollutants such as POPs [36–38]. Within the broad range of oxide semiconductors applied in photocatalysis, wide band gap ones, such as SnO$_2$ [39,40], TiO$_2$ [41,42], ZnO [43], NiO [44], and WO$_3$ [45–47], have been extensively explored because they are considered good semiconductors and have showed promising results. Although most studies have been dedicated to titanium oxide (TiO$_2$), tin oxide (SnO$_2$) is also worth mentioning, as indicated by
the number of published research articles listed in Table 1. SnO$_2$ is an n-type semiconductor with a large band gap value between 3.6 and 4.0 eV [47,48]. Its intrinsic characteristics are responsible for the material’s conductivity, good optical and electrical characteristics, and high thermal stability, among other qualities. Furthermore, SnO$_2$ presents a high oxidation potential and chemical inertness, as well as corrosion resistance and non-toxicity [47]. Thus, SnO$_2$ has been successfully used in heterogeneous photocatalysis and showed to be very efficient in the degradation of different organic pollutants, especially dyes [49–51].

![General mechanism of the heterogeneous photocatalysis using a hypothetical semiconductor.](image)

**Figure 1.** General mechanism of the heterogeneous photocatalysis using a hypothetical semiconductor.

In respect to the synthesis of SnO$_2$ that can affect the material properties and applications, the photocatalytic ones can be easily modified according to the synthesis route used to prepare the material. In addition, although most SnO$_2$-based photocatalysts are explored in their powder form, their separation and reuse after the process is difficult, as it needs filtration to recover the catalyst. Thus, other researchers have prepared SnO$_2$-based catalysts in their film form, which is considered an effective approach to overcome the filtration issues after a photocatalytic reaction, allowing the reusability of the catalyst without any material loss. These factors are important to design new and efficient photocatalysts.

Some review articles have dealt with the applications of SnO$_2$ materials in photocatalysis. However, to our knowledge, none of them draw attention to particular characteristics of synthesis methods and experimental conditions used to prepare the different SnO$_2$-based photocatalysts and how these directly impacted the sample characteristics, properties, and photocatalytic efficiency for organic dyes degradation. For instance, in the review published by Al-Hamdi et al. [47], results from studies conducted up to 2017 were shown, which focused on the applications of SnO$_2$ in an advanced oxidative process for the degradation of organic pollutants, such as phenols, phthalates, and other toxins in water. In addition, authors listed relevant investigations on fundamental aspects related to SnO$_2$, such as structure and properties, the charge transfer mechanism involved, and parameters that affect the photodegradation of pollutants in aqueous solutions, such as the catalyst load, concentration of the contaminant, and pH. In a recent review, Sun et al. [48] discussed the importance of employing SnO$_2$-based photocatalysis in environmental science and energy fields. The authors focused several studies dealing with strategies used to enhance the SnO$_2$ properties for photocatalysis. The effect of doping, formation of solid solutions, stoichiometry, particle size and morphology, besides the formation of hierarchical, porous, and heterojunctions structures, were demonstrated. Apart from the applications in water
splitting and organic dyes photodegradation, the authors discussion gave examples of SnO$_2$ applications in Cr(VI) and CO$_2$ reduction. The authors also mentioned the complexities of applying photocatalysts in a large scale to simulate the real scope of the industry since, although significant progress has been achieved in the improvement of the photocatalytic efficiency of SnO$_2$, there are still major challenges to be faced.

Table 1. Metal oxides applied in heterogeneous photocatalysis of dyes over the last 5 years. Data collected from different research literature databases on 20 January 2022.

| Oxide | Publications Attributed to Photocatalysis of POPs | Database | Publications Attributed to Photocatalysis of Dyes | Database |
|-------|-----------------------------------------------|----------|-----------------------------------------------|----------|
| TiO$_2$ | 9.103 | Scopus | 2.132 | Scopus |
|       | 1.369 | Web of Science | 3.155 | Web of Science |
|       | 2.492 | ScienceDirect | 3.933 | ScienceDirect |
|       | 1.364 | ACS Journal Search | 1.679 | ACS Journal Search |
| ZnO | 2.862 | Scopus | 1.251 | Scopus |
|      | 252 | Web of Science | 1.759 | Web of Science |
|      | 4.558 | ScienceDirect | 7.659 | ScienceDirect |
|      | 542 | ACS Journal Search | 8.06 | ACS Journal Search |
| NiO | 346 | Scopus | 93 | Scopus |
|      | 27 | Web of Science | 1.38 | Web of Science |
|      | 1.164 | ScienceDirect | 1.50 | ScienceDirect |
|      | 171 | ACS Journal Search | 1.74 | ACS Journal Search |
| SiO$_2$ | 582 | Scopus | 1.49 | Scopus |
|      | 41 | Web of Science | 1.50 | Web of Science |
|      | 228 | ScienceDirect | 3.43 | ScienceDirect |
|      | 438 | ACS Journal Search | 4.39 | ACS Journal Search |
| WO$_3$ | 951 | Scopus | 1.35 | Scopus |
|      | 10 | Web of Science | 1.91 | Web of Science |
|      | 2.593 | ScienceDirect | 3.41 | ScienceDirect |
|      | 2.570 | ACS Journal Search | 3.12 | ACS Journal Search |
| SnO$_2$ | 425 | Scopus | 1.81 | Scopus |
|      | 21 | Web of Science | 2.39 | Web of Science |
|      | 173 | ScienceDirect | 3.30 | ScienceDirect |
|      | 191 | ACS Journal Search | 2.61 | ACS Journal Search |

Based on what was mentioned, the present work aims to address important aspects about recent and relevant achievements of how the choice of the appropriate synthesis method and experimental parameters may influence the morphology, particle size, bandgap energy, and, consequently, the impact on the photocatalytic properties of SnO$_2$ for the degradation of different organic dyes as targets of persistent organic pollutants (POPs) molecules. Apart from the applications of pure SnO$_2$ toward the degradation of dyes, studies concerning photocatalysis using metal and nonmetal-doped SnO$_2$, as well as SnO$_2$-based composites, are also summarized. We believe that this literature review may provide important aspects for the better development of SnO$_2$-based catalysts, and understanding the limitations, in order to use them in practical devices.

2. Persistent Organic Pollutants (POPs), Dyes, and Photocatalysts

Environmental problems, and especially water resources, have been a major problem for the population, researchers, and environmentalists [1,2]. There are several contaminants that are usually disposed into the environment, and can be classified into different types, depending on their purpose and origin: persistent organic pollutants (POPs), pharmaceuticals and personal care products (PPCPs), endocrine disrupting chemicals (EDCs), and agricultural chemicals (pesticides, herbicides) [51–53]. Particularly, persistent organic pollutants (POPs)—which are organic chemicals that are highly toxic to humans—animals,
and vegetation stand out among these pollutants. The specific effects of POPs can include allergies, cancer, and damage to the central and peripheral nervous system and reproductive, endocrine, and immune systems [13,14,54].

According to the Stockholm Global Conference held in 2001 that aimed to discuss the consequences of the environmental degradation, twelve (12) chemical substances were included into the POPs class. However, thirty-five (35) POP substances are listed as most harmful [13,15,21,55–59]. Amongst the variety of POPs, drugs, pesticides, hormones, dyes, synthetic textile products, artificial sweeteners, some micro-organisms, and algae toxins have extensively been introduced on a large scale into the environment, which can strongly affect aquatic life and human beings [21,53]. Particularly, synthetic organic dyes stand out for being considerably used over the years in several technological sectors, such as textile industries, in medicine, and in the manufacturing process of pens, among others.

Synthetic dyes are classified as POPs because of their difficulty to be degraded and removed from the environment [55]. The disposal of effluents from textile industries is one of the main sources of pollutants responsible for changes in the quality of receiving water. The different dyes used in textile industries generally have a high organic load, and this can be very harmful to the environment if inappropriately discharged. There is an estimate that about 20% of the dyes used are disposed in effluents because of losses during the dyeing process [36,59,60]. In this context, several methods (chemical, physical, and biological) have been used for dealing with the treatment of contaminated effluents with POPs (Figure 2). However, great efforts have been dedicated to studies related to photodegradation and mineralization of organic dyes using photocatalytic materials under UV-visible light, as well as sunlight irradiation.

Figure 2. Published articles referring to the treatment of different persistent organic pollutants. Data collected from Web of Science on 20 January 2022.

As aforementioned, different materials have been applied in heterogeneous photocatalysis, especially for photodegradation of POPs. As this review aimed to discuss important works involving the photodegradation of organic dyes and types of POPs, the article’s search was done considering different databases, as listed in Table 1. It is important to highlight that most of the publications have devoted to the application of TiO₂-based photocatalysts. However, different metal oxides have been explored for such purposes because of their unique and prominent properties, due to their structure, particle size, and morphology [61]. From this search, SnO₂ appears in 6th place among the six (6) oxide-
based photocatalysts used in photocatalysis of organic dyes. It is still important to mention that scientific research based on tin materials has been increased over the last years.

It is important to highlight that the search was done using the following words for articles dealing with photodegradation of POPS, such as for SnO$_2$: “SnO$_2$, photocatalysis and POPs” and “tin oxide, photocatalysis and POPs”; while for works concerning photodegradation of dyes included, “SnO$_2$, photocatalysis and dyes” and “tin oxide, photocatalysis and dyes”. A similar search has been completed for each metal oxide listed in Table 1. Compound words such as “photocatalytic properties”, “photocatalytic efficiency”, and “organic dyes” were not placed in quotes for the search of papers related to photocatalysis of dyes using these oxides.

Considering organic dyes, additional studies dealing with these pollutants are in need. For instance, the emission of effluents from textile industries is one of the main things responsible for alterations in water bodies’ quality. Organic dyes have a high organic load, generating strong impacts on the environment when inappropriately discharged. Depending on the type of dye and the place where it is discarded, they can react with other substances present in the environment, originating mutagenic and carcinogenic by-products [62,63]. In dye molecules, two important organic groups are present: the chromophore one, which is responsible for the color by light absorption, and the auxochrome group that is responsible for dye fixation on fibers and for color intensification. These groups are constituted by conjugated systems with double bonds and functional groups (auxochromes), which are electron donors or acceptors [64]. Such characteristics have made dyes one of the great villains in the process of polluting water. In this context, a great amount of research has explored the photocatalysis of different organic dyes using wide band gap semiconductors, such as SnO$_2$. These works are discussed as follows for SnO$_2$, which has been gaining prominence in this type of application over the years, as shown in Figure 3.

![Figure 3. Number of published articles concerning the photodegradation of organic dyes using SnO$_2$-based photocatalysts. Search performed on 20 January 2022, using different databases. The period of 2017–2021 was considered for this search.](image-url)
3. Tin-Oxide-Synthesis, Structure, Properties, and Applications as Heterogeneous Photocatalysts

Tin dioxide (SnO₂), obtained by combining Sn⁴⁺ and O²⁻, is a ceramic material that has been used in a wide variety of applications, such as gas sensors, photovoltaic energy converters, and photocatalysts [65–67]. The success for multiple applications of SnO₂ is due to its intrinsic characteristics, such as: n-type conductivity, which is responsible for the conductivity of the material, and in addition to optical and electrical characteristics, high thermal stability, and high surface area [36,49,68]. Electrical conductivity can be described in terms of the movement of negatively charged electrons, and this gives rise to an n-type semiconductor [36,49,50,69–71].

As for the crystal structure of SnO₂, at room temperature, it adopts a rutile-type tetragonal structure (cassiterite) with P42/mnm space group, as illustrated in Figure 4. This structure is formed by a tetragonal unit cell defined by three (3) parameters: the a and c lattice parameters and the internal parameter, u, which defines the oxygen position (u, u, and 0). At room temperature, the theoretical lattice parameters for SnO₂ are a = 4.7374 Å, c = 3.1864 Å [72], and u = 0.3056 Å. It has been reported that SnO₂ can also adopt an orthorhombic, CaCl₂-type (Pnnm) structure, besides existing in an orthorhombic α-PbO₂-type (Pbcm), a cubic pyrite-type (P4-3), an orthorhombic ZrO₂-type (Pnma), and a cotunnite-type (Pnma) structure. However, these structures are metastable at ambient conditions, and it is hard to follow these phase transitions through traditional methods under low pressure and temperatures [63,64].

![Figure 4](image_url)

**Figure 4.** (a) A P42/mnm tetragonal crystal structure of SnO₂ showing (b) environment of distorted SnO₆ octahedron and (c) lattice parameters, obtained using VESTA software and structural parameters reported in [73].

The structure of SnO₂ consists of chains of SnO₆ octahedra in which each Sn atom is octahedral, surrounded by six oxygen atoms, while each oxygen is surrounded by three Sn atoms arranged at the corners of an equilateral triangle. The structure’s Sn:O coordination is 6:3, with each octahedron not being regular, showing a slight orthorhombic distortion [72,73].

The literature has reported that SnO₂ can present nano- and micro-structured characteristics, however, most researchers have been dedicated to the synthesis of SnO₂ in the form of nanoparticles. Such characteristics have offered good opportunities to explore new physical and chemical applications. Recently, SnO₂ has been applied to the degradation of antibiotics [74], fuel cell catalysts [73], gas sensors [75], sodium ion batteries [76],
light-emitting diodes [77], heterogeneous photocatalysis, and antimicrobial activity [78,79]. There are several synthesis routes and experimental conditions to obtain SnO$_2$ in powder form, the choice of which may influence the particles’ morphology and texture, besides their structural, optical, and electronic properties, which will consequently impact the efficiency and final application of the material. In relation to SnO$_2$-based thin films, chemical deposition methods have appeared to be the most used [62,80–84].

With respect to the applications in photocatalysis, SnO$_2$ catalysts with a tetragonal, rutile-type structure are the main phases investigated, but a recent work reported that the coexistence of mixed tetragonal–orthorhombic phases affected the photocatalytic efficiency of SnO$_2$ [85]. All these factors are described below.

4. Synthesis Methods and the Influence of Experimental Parameters on the Characteristics and Photocatalytic Properties of SnO$_2$-Based Materials

One of the main aspects this review refers to is the approach of how different synthesis methods and experimental parameters affect the photocatalytic properties of SnO$_2$-based materials.

Researchers have employed different methods to produce new materials with specific properties and applications. The choice of an appropriate condition to prepare a material is crucial to modifying its physicochemical characteristics, such as crystal structure and morphological and texture characteristics of the particles (size, shape, surface area, and surface charge characteristics), as well as the electronic and optical properties. The variation on these characteristics may change the applicability of the material. In this sense, different researchers have devoted to obtaining SnO$_2$-based materials with specific characteristics and efficient photocatalytic behaviors. Among the great variety of methods known in the literature, SnO$_2$-based photocatalysts have been obtained, in their powder form, by a solvothermal reaction [86,87], the microwave-assisted hydrothermal method [88], chemical precipitation [68], the polymeric precursor method [89], and sol-gel [14], all which are mostly used.

In relation to the photocatalytic properties of SnO$_2$ particles, although there is a variety of organic pollutants, organic dyes such as Methylene blue (MB), Rhodamine B (RhB), and Methyl orange (MO) have been the most used as target molecules to evaluate the photocatalytic efficiency of SnO$_2$. However, other organic dyes such as Congo red (CR), Malachite green (MG) and Eriochrome black T (EBT) have also been explored for such purposes. In this context, the photodegradation of these organic dyes is highlighted in this review. Apart from powdered SnO$_2$ materials, SnO$_2$-based films have also been used as photocatalysts for dye photodegradation [90–93]. For the preparation of films, chemical [64,80,84,86,89] and physical-based deposition methods [64] are used. In the following sections, photocatalytic applications of pure and doped SnO$_2$-based catalysts, as well as the composites based on SnO$_2$ in powdered and film forms prepared by different techniques, are also discussed.

4.1. Pure SnO$_2$-Based Photocatalysts

Besides being synthesized in its pure form, SnO$_2$ has been prepared in its doped form with metals or nonmetal ions, as well as in the form of composites with other semiconductor materials, or even impregnated in inert, non-active photocatalytic materials. It is well known that the generation of electrons/holes (e$^–$/h$^+$) pairs by absorption of a photon of equal energy, to or higher than the band gap energy induced by light, is a basic prerequisite for a semiconductor to be used in photocatalysis. Because of its wide band gap of SnO$_2$ (3.6 eV), no absorption response to the visible light would be achieved, and this is the main disadvantage of this material, which restricts its application in practical devices. A wide variety of pure semiconductor materials, particularly SnO$_2$, have been investigated regarding the photocatalytic properties, but only few of them are considered effective photocatalysts.
Besides the value of the forbidden band energy corresponding to absorption in the visible region, it is required that the energy levels of the conduction and valence bands are suited to the redox potential of the water, so as to produce reactive agents to promote breaking of organic pollutants molecules. Therefore, different authors have employed various synthesis methods and varied experimental conditions in order to change particle characteristics, which include size, morphology, and texture, as well as the crystallinity and the presence of defects, in order to design efficient photocatalysts under UV irradiation. These parameters can play an important role during photocatalysis, as they can affect the adsorptive and photo-absorption capacity of the catalyst, besides reducing photogenerated changes of recombination during photoexcitation. In addition, some authors also explored the reaction mechanisms involved in the photodegradation of the dyes and how the reactive species act for the photocatalysis to occur. These points are discussed throughout the review.

For instance, considering pure tin oxide as a photocatalyst, Akram et al. [90] prepared SnO$_2$ nanoparticles by the continuous microwave flow synthesis (CMFS) method in a domestic microwave oven operating at 600 W, using tin chloride pentahydrate (SnCl$_4$·5H$_2$O), sodium hydroxide (NaOH), ethanolic solutions. The solutions were pumped through the microwave with the aid of peristaltic pumps to attain 10 min of retention time inside the device. The resulting suspension was filtered, washed, and dried at 80 °C for 12 h, followed by heating at 200 °C for 2 h to obtain SnO$_2$ samples. The photocatalytic properties of the nanoparticles were investigated toward the photodegradation of Methylene blue (MB) dye under UV irradiation (365 nm). In their study, the authors also investigated the effect of the concentration of the reacting SnCl$_4$·5H$_2$O and NaOH materials on the crystallinity, particle size, and morphology, as well as the photocatalytic behavior of the SnO$_2$ nanoparticles. The authors evidenced that crystalline SnO$_2$ samples with tetragonal, rutile-type structures were obtained only after heating at 200 °C. The increase in the concentration of SnCl$_4$·5H$_2$O (from 0.25 to 0.75 M) and NaOH (from 1 to 3 M) provoked an increase in sample crystallinity and an average particles size from 4.33 to 8.56 nm, with no meaningful change in morphology. This phenomenon was attributed to the increased number of nuclei sites formed by the reacting species, as well as to the microwave irradiation that favors better nucleation and crystal growth. Surprisingly, a decrease of the band gap ($E_g$) values from 3.33 to 3.19 eV was also observed as a function of the precursor’s concentration, and it was associated to the increase of the particle size. In relation to the photocatalytic property of the samples, that sample was prepared using the lower concentration of the reacting species, which presented the lowest degree of crystallinity (63%), smallest particle size (4.43 nm), largest surface area (153.57 m$^2$ g$^{-1}$), and widest band gap (3.33 eV), and this was the most efficient in the photodegradation of MB dye, reaching up to 93% of degradation in 240 min. According to the authors, the higher efficiency observed for that sample was mainly due to the large surface area and higher concentration of defects.

Still considering pure tin oxide photocatalysts, Abdelkader et al. [92] synthesized SnO$_2$ nanoparticles via a sol-gel method and calcined it at different temperatures (80, 450, and 650 °C) for 4 h, in order to achieve different crystallinity and particle morphology. The authors investigated the photocatalytic efficiency of the synthesized samples toward Congo red (CR) dye degradation under UVA irradiation. For the synthesis of the nanoparticles, tin chloride (SnCl$_2$·2H$_2$O) was dissolved in 250 mL of deionized water to obtain a white suspension of a 0.4 M Sn (II) concentration. The suspension was stirred for 1 h at room temperature and oxalic acid was added dropwise to the aqueous solution as a chelating agent with a molar ration of 1:1 (oxalic acid: tin cations). The obtained suspension was centrifuged, filtered, and washed several times to eliminate chloride ions. The washed precipitate was dried at 80 °C/24 h and calcined at 450 to 650 °C for 4 h. According to the results, the sample obtained after drying at 80° for 24 h (SnO$_2$-80) crystallizes in pure tin oxalate (SnC$_2$O$_4$) with a monoclinic structure. On the other hand, pure SnO$_2$ with a $P4_2/mnm$ tetragonal, rutile-type structure was obtained after calcinations at 450 (SnO$_2$-450) and 650 °C (SnO$_2$-650). The authors draw attention to the use of oxalic acid as a chelating agent, and it affected the particle characteristics for controlling the nucleation and crystal
As the photostability and reusability of the photocatalyst are important aspects, the authors evidenced the complete mineralization of dye led to CO\(_2\) evolution during the photocatalysis, which was analyzed by GC analysis. The authors evidenced that samples calcined at higher temperatures are more aggregated with a foamed aspect because of the smallest particle size of the powders. The samples calcined at different temperatures also presented a specific surface area that varied from 66.41 to 37.54 m\(^2\) g\(^{-1}\), and with band gap values from 3.35 to 3.49 eV for the SnO\(_2\)-450 and SnO\(_2\)-650 samples, respectively.

Curiously, regarding the photocatalytic behavior of the samples prepared by Abdelkader et al. [92], the highest efficiency in the degradation of CR dye (catalyst/CR dye concentration of 0.5 g L\(^{-1}\)) was achieved using SnO\(_2\)-650 that presented a lower surface area. This sample presented a dye photodegradation efficiency of 61.53% after 100 min under irradiation. The authors pointed out that the highest efficiency observed for this sample is especially due to its greater crystallinity that gives less surface defects as well as particle aggregation. The lower density of surface defects (Sn\(^{2+}\) and oxygen vacancies) could reduce recombination of the electron–hole (e\(^{-}\)/h\(^{+}\)) pairs. Because of this fact, the authors proposed the photocatalytic mechanism involved in the CR photodegradation. The reactions mechanism can be represented by Equations (1)–(6). According to the authors, under UVA light, electrons are excited from the VB to the CB of SnO\(_2\) (Equation (1)) and, simultaneously, holes are created in the VB. The photoinduced e\(^{-}\) in the CB can directly reduce Sn\(^{4+}\) to Sn\(^{2+}\) (Equation (2)). However, as Sn\(^{4+}\) can act as a scavenger of e\(^{-}\), Sn\(^{2+}\) can influence the photo-reactivity by altering a e\(^{-}\)/h\(^{+}\) recombination (Equation (3)). The h\(^{+}\) in the VB is captured by H\(_2\)O generating hydroxyl \(\cdot\)OH radicals (Equation (4)). In addition, according to the band energy position, the authors stated that the h\(^{+}\) in the VB of SnO\(_2\) (+3.50 eV/NHE) is more positive than that of the H\(_2\)O/\(\cdot\)OH couple (+1.9 eV/NHE), which is required for organic pollutant decomposition R/R\(^{**}\) (+1 V/NHE), indicating that the photoinduced holes in the VB can oxidize the adsorbed CR dye and H\(_2\)O molecules on the SnO\(_2\) surface. Thus, the formation of organic cation-radicals (R\(^{**}\)) is formed (Equation (5)). As a result, all the O\(_2^{**}\), \(\cdot\)OH, and R\(^{**}\) radicals participate in redox reactions responsible for decomposing CR dye (Equation (6)).

\[
\text{SnO}_2 + \text{UVA}(365 \text{ nm}) \rightarrow \text{SnO}_2 (\text{h}^+_{\text{VB}} + e^-_{\text{CB}}) \\
\text{Sn}^{4+} + e^-_{\text{CB}} \rightarrow \text{Sn}^{2+} \tag{1}
\]

\[
\text{Sn}^{2+} + 2\text{O}_2 \rightarrow 2\text{O}_2^{**} + \text{Sn}^{4+} \tag{2}
\]

\[
\text{H}_2\text{O} + \text{h}^+_{\text{VB}} \rightarrow \text{\(\cdot\)OH} \tag{3}
\]

\[
\text{R} + \text{h}^+_{\text{VB}} \rightarrow \text{R}^{**} \tag{4}
\]

\[
(\text{O}_2^{**}, \text{\(\cdot\)OH, R}^{**}) + \text{CR dye} \rightarrow \text{intermediate products} \rightarrow \text{Dye degradation} \tag{5}
\]

Although most studies concerning photocatalytic degradation of dyes using pure SnO\(_2\) have been performed under UV light, some authors investigated the photocatalytic properties of SnO\(_2\) under visible light and sunlight. For instance, Kumar et al. [94] reported the use of SnO\(_2\) particles prepared by a simple, eco-friendly, and low-cost biosynthesis process using guava (Psidium guajava) leaf extract in the photodegradation of Reactive yellow 186 (RY186) dye under sunlight. The authors prepared the samples by mixing a 2.1 M SnCl\(_4\) solution with the extracts in a ratio of 1:1, and kept under stirring at 60 °C for 4 h, followed by calcination at 400 °C for 4 h to obtain SnO\(_2\) nanoparticles. The authors evidenced that the SnO\(_2\) single-phase nanoparticles with a size of 8–10 nm showed a high photocatalytic efficiency, degrading 90% of RY186 dye in 180 min. For the photocatalysis, a concentration of 1 g L\(^{-1}\) of SnO\(_2\) sample was used. According to the authors, superoxide (O\(_2^{**}\)) and hydroxyl (\(\cdot\)OH) radicals are responsible for the photodegradation of dye. The efficiency of the photocatalyst in the degradation and mineralization of RY186 was confirmed by CO\(_2\) evolution during the photocatalysis, which was analyzed by GC analysis.
evaluated them by performing five consecutive photocatalytic cycles. The photoactivity of the catalyst remained about constant even up to five experiments, although gradual losses in activity were expected. It is important to highlight that the use of powdered samples in photocatalysis might be disadvantageous due to the loss in the amount of the catalyst after centrifugation and filtration processes, which leads to a loss in photoactivity. However, the authors reinforced that the synthesized catalyst is easily separable from the solution.

Like Kumar et al. [94], Haq et al. [78] synthesized tin dioxide (SnO\(_2\)) nanoparticles by an eco-friendly process using leaves extracts. However, in the study conducted by Haq et al. [78] *Daphne mucronata* leaf extract was used as a capping and reducing agent in order to control particle size and morphology. The authors evaluated the photocatalytic performance of the samples toward Rhodamine 6G (R6G) dye degradation using 0.4 g L\(^{-1}\) of the catalyst. For the materials synthesis, a mixture of a 0.003M SnCl\(_4\)·5H\(_2\)O solution with 20mL of the leaf extract was kept under 500 rpm and stirring at 55 \(^{\circ}\)C. A greenish gel was obtained after 40 min and aged for 24 h, which was subsequently washed with hot water, filtered, washed with ethanol, and finally dried at 100 \(^{\circ}\)C for 6 h to obtain a fine, colorless powder. The authors obtained nanoparticle samples with an average particle size of 64 nm and specific surface area around 147 m\(^2\) g\(^{-1}\). A maximum of 99.70\% of the dye degradation was observed after 390 min under simulated sunlight. The charge generation and photodegradation mechanism reported by Haq et al. [78] is the same as the one described by Kumar et al. [94] for RY186 dye [94].

Kumar et al. [94] synthesized SnO\(_2\) nanocrystals by a solution-phase growth technique, and the structural, optical, and photocatalytic properties of the nanostructures were investigated. The effects of reaction temperature (180 and 200 \(^{\circ}\)C), time (24 and 30 h), the use of CTAB (CetylTrimethylAmmonium Bromide) surfactant on the particle size, morphology, and band gap energy were evaluated. According to the authors, the SEM images showed that samples synthesized at 180 \(^{\circ}\)C for 24 (Sample 1) and 30 h without the surfactant (Sample 2) presented particles with a rod-like morphology and crystallite size of 17.12 and 26.51 nm, respectively. When the sample was synthesized at 200 \(^{\circ}\)C for 24 h without surfactant (Sample 3), SnO\(_2\) nanostructures with a nanoflower-like morphology, with crystallite size at 28.25 nm were obtained. On the other hand, the samples prepared at 180 (Sample 4) and 200 \(^{\circ}\)C for 24 h with the addition of CTAB (Sample 5) showed particles with a nanosphere-type morphology with sizes of 23.80 and 32.14 nm, respectively. The band gap (\(E_g\)) values estimated for sample 1 (nanorods), Sample 2 (nanorods), Sample 3 (nanoflowers), Sample 4 (nanospheres), and sample 5 (nanospheres) were 4.05, 3.88, 3.84, 3.95, and 3.76 eV, respectively. The authors associated the variation of the \(E_g\) values mainly to the temperature and time conditions of which the samples were prepared, that directly affected the particle size and morphology and, therefore, impacted photocatalytic activity of the samples. Despite the wide band gap of the samples, the authors evaluated their photocatalytic activity using 0.5 g L\(^{-1}\) of the catalysts in the degradation of Rhodamine B (RB) dye under direct sunlight irradiation. Surprisingly, the authors observed a high photodegradation of dye under sunlight, and the efficiency of the SnO\(_2\) nanostructures was strongly related to the particles’ morphology. According to the authors, even presenting a \(E_g = 3.76\) eV, sunlight was enough to promote photoexcitation in Sample 5 to degrade 91.7\% of the dye after 2 h. As expected, Sample 1 (SnO\(_2\) nanorods with \(E_g = 4.05\) eV) displayed the lowest dye degradation efficiency in 2 h (76\% of the dye is degraded). The authors established that size, morphology, specific surface area, and dispersion of the catalysts played key role in the photodegradation of the dye.

Assis et al. [58] used a polymeric precursors method to prepare SnO\(_2\) particles at different temperatures (700, 800, and 900 \(^{\circ}\)C). After being prepared, the powders were impregnated in polystyrene foams in order to increase surface area due to the porous characteristic of polystyrene, besides a favoring for the recovery of the material after use. The photocatalytic property of the samples was investigated in the degradation of RB dye under UV irradiation with a catalyst/dye concentration of 0.4 g L\(^{-1}\). The authors observed, using high-resolution transmission electron microscopy (HRTEM), that the SnO\(_2\) samples
present nanoparticles with sizes ranging between 20 and 80 nm. Moreover, the formation of agglomerates was observed in the samples calcined at higher temperatures (800 and 900 °C). The oxide obtained at lower temperatures presented a smaller particle size and a larger surface area, which resulted in a greater photocatalytic activity, degrading 98.2% of degradation of the rhodamine RhB after 70 min.

A very conventional synthesis procedure for obtaining oxide-based catalysts is the so-called sol-gel method. Thus, Najjar et al. [93] synthesized SnO$_2$ nanoparticles by a green sol-gel method, using chitosan as a polymerizing agent, and calcined it at different temperatures (500, 700, 800, and 1000 °C). The authors also draw attention to the use of chitosan that may increase particle stability, prevent particles aggregation, and reduce the particles’ toxicity. According to the TGA-DTA analysis, the temperature of 700 °C (namely, SnO$_2$-NPs at 700 °C) proved to be more adequate to prepare the desirable SnO$_2$ catalyst. The material calcined at this temperature presented a spherical particle morphology with an average size of 10 nm, as observed by TEM analyses. The authors evaluated the photocatalytic properties of SnO$_2$-NPs at 700 °C toward the photodegradation of Eriochrome black T (EBT), an azo-type anionic dye. The photocatalytic tests were carried out by adding 21.1 mg L$^{-1}$ of the catalyst in 100 mL of EBT dye solution ($10^{-5}$ M) and kept at a constant stirring and UV irradiation (Hg vapor lamp, 500 W) for 270 min. Regarding the photocatalytic activity of the prepared SnO$_2$-NPs, a photodegradation efficiency of 77% was obtained after 270 min. In order to investigate the best conditions for optimum dye degradation using SnO$_2$-NPs, Najjar et al. [93] also investigated the influence of the catalyst concentration (8.7, 21.1, and 43.2 mg L$^{-1}$) and the solution pH (3.5, 5, 7, and 9). The authors observed an increase of the photodegradation rate by increasing the concentration of the photocatalyst from 8.7 to 21.1 mg L$^{-1}$, decreasing afterwards. According to the authors, this decrease in photocatalytic efficiency of SnO$_2$-NPs in a higher concentration is due to the fact of accumulation of nanoparticles that lead to a decrease in the generation of reactive radicals, such as hydroxyls (•OH). With respect to variation of the solution pH, the highest photodegradation efficiency (77%) was attained at the isoelectric point of SnO$_2$-NPs at pH 3.5 (Zeta potential = 0 eV). Curiously, the photodegradation of the anionic EBT dye at positive Zeta potential values (at pH 2) was not as expressive as that observed at the isoelectric point of the catalyst.

According to Najjar et al. [93] the general mechanism involved in the dye photodegradation using SnO$_2$-NPs is summarized by Equations (7)–(12), that are similar to those reactions displayed in Figure 1 for a hypothetical catalyst.

\[
\begin{align*}
\text{SnO}_2 + h\nu &\rightarrow \text{SnO}_2 + (h^+_{CB} + e^-_{VB}) \quad (7) \\
\text{SnO}_2 (h^+_{CB}) + \text{H}_2\text{O} &\rightarrow \text{SnO}_2 + \text{H}^+ + \cdot\text{OH} \quad (8) \\
\text{SnO}_2 (e^-_{CB}) + \text{O}_2 &\rightarrow \text{SnO}_2 + \text{O}_2^- \quad (9) \\
\text{O}_2^- + \text{H}^+ &\rightarrow \text{HO}_2^* \quad (10) \\
\text{HO}_2^* + \text{H}_2\text{O} &\rightarrow \text{H}_2\text{O}_2 + \cdot\text{OH} \quad (11) \\
\text{Dye} + \cdot\text{OH} &\rightarrow \text{degradation dye} \quad (12)
\end{align*}
\]

As the photostability of the catalyst is an important factor for its reuse in consecutive photocatalytic tests, cyclic experiments of EBT photodegradation were carried out for the SnO$_2$-NPs under the optimal conditions established in the work. Thus, Najjar et al. [93] evidenced that the degradation rate of EBT remained over 74% after five cycles. In addition, using FTIR, XRD, TEM, and FESEM analysis, the authors showed that no visible changes were observed in the samples after the fifth cycle, which confirms the high photostability of the synthesized catalyst.

Recently, Luque et al. [39] synthesized SnO$_2$ nanoparticles (SnO$_2$ NPs) by a green synthesis using Citrus x paradisi extract as a stabilizing capping agent. There were different concentrations of the extract (1, 2, and 4% in relation to the aqueous medium—SnO$_2$
NPs-1, 2, and 4%). It is important to highlight that a heating treatment at 400 °C for 1 h was completed to obtain crystallized SnO$_2$ NPs. The authors obtained crystalline SnO$_2$ nanoparticles with average sizes of 9.1, 5.1, and 4.7 nm when 1, 2, and 4% of the capping agent was used in the synthesis, respectively. These samples also presented band gap values ($E_g$) of 3.28, 2.77, and 2.69 eV, respectively, which were smaller than those $E_g$ values reported by Mahmood et al. [95]. This confirmed the role of this capping agent in controlling the particles’ size, as well as in the modification of optical band gap properties of SnO$_2$ NPs. The photocatalytic properties of the samples were then investigated under both solar and UV irradiation using a SnO$_2$ NPs/dye concentration of 1.0 g L$^{-1}$. Furthermore, Methyl orange (MO), Methylene blue (MB) and Rhodamine B (RhB) were used as target dyes. Regarding the photocatalytic efficiency of the SnO$_2$ NPs, SnO$_2$ NPs-4% presented the highest efficiency in the degradation of the dyes, degrading 100% of MO after 180 and 20 min under solar and UV irradiation, respectively. In relation to other dyes, SnO$_2$ NPs-4% degraded 100% of MB and RhB after 60 min under UV irradiation. The efficiency of this photocatalyst in degrading MO, MB, and RhB dyes under these conditions was confirmed by Turnover number (TON) and Turnover frequency (TOF) analysis. The authors associated the superior photocatalytic efficiency of SnO$_2$-NPs-4% to the smaller particle size, larger surface area, and the increased number of active sites present on the surface when compared to the other samples. Finally, the authors investigated the involvement in the degradation of dyes, and they evidenced that •OH radicals are the main species responsible for degradation.

Apart from the above-mentioned SnO$_2$-based photocatalysts prepared by various synthesis methods and the experimental conditions, the search for different photocatalytic materials with the desired efficiency is still a challenge. In this context, different authors have prepared SnO$_2$ catalysts, owing to the flexibility of applications, including photocatalysis. Compared to powdered materials, the use of films in photocatalysis has some advantages, especially for being easily recovered and reused in different batches. For instance, Bezzerouk et al. [80] deposited SnO$_2$ thin films on glass substrates at 450 °C by an ultrasonic spray pyrolysis technique, and desired polycrystalline SnO$_2$ films were obtained with a band gap of 3.80 eV, greater than that for bulk SnO$_2$ ($E_g = 3.6$ eV). The authors evaluated the photocatalytic property of the films toward Methylene blue (MB) degradation under UV-LEDs (340–400 nm, 7 W) and ultrasound (US) transducer (40 KHz). Different degradation processes were investigated, such as: photolysis (UV), photocatalysis (SnO$_2$ + UV), the sonolysis process (US), and sonocatalysis (SnO$_2$ + US) as well as sonophotolysis (US + UV) and sono-photocatalysis (SnO$_2$ + UV + US). Curiously, SnO$_2$ film did not show meaningful activity in the degradation of MB dye under UV irradiation (photocatalysis). However, when US was employed, a pronounced increase in the dye degradation was observed, reaching to 88.33%, 94.31%, 97.28%, and 98.25% of efficiency when sono-photocatalysis (SnO$_2$ + UV + US), sonolysis (US), sonophotolysis (US + UV) and sonocatalysis (SnO$_2$ + US) processes were used, respectively. The authors associated the highest efficiency of dye degradation using sonocatalysis to the production of acoustic cavitation in the water that can favor the dissociation of water and the formation of an important quantity of •OH radicals that participate in the degradation of dye. Finally, the authors associated the lower efficiency of the processes under UV irradiation to the rapid recombination of the electron–holes during SnO$_2$ photoexcitation. Therefore, authors showed different processes used to improve dye degradation using SnO$_2$-based material.

As one could see, several methods and experimental conditions were employed to synthesize undoped SnO$_2$ materials (in powder and film forms) with different characteristics and properties. It has been shown that these characteristics can directly impact the photocatalytic activity toward the degradation of dyes under UV-visible light, as well as under direct or simulated sunlight or even coupled with ultrasound irradiation. A summary of some important works concerning the photocatalytic applications of different undoped SnO$_2$ materials obtained by different methods is listed in Table 2.
Table 2. Summary of works related to the synthesis of pure SnO$_2$ materials by different methods and their application as photocatalysts for organic dye degradation.

| Catalyst Type         | Synthesis Method                                           | Synthesis Conditions (Temperature/Time of Calcination) | Dye Solution Concentration (mg L$^{-1}$) | Photocatalyst Concentration (g L$^{-1}$) | Pollutant * | Irradiation | Efficiency/Time |
|-----------------------|------------------------------------------------------------|--------------------------------------------------------|----------------------------------------|------------------------------------------|-------------|-------------|----------------|
| SnO$_2$ NPs [94]      | Green solution synthesis                                   | 400 °C/4 h                                             | 40                                      | 1.0                                      | RY186       | Sunlight    | 90%/3 h        |
| SnO$_2$ NPs [90]      | Continuous microwave flow synthesis (CMFS)                | 200 °C/2 h                                             | 50                                      | -                                        | MB          | UV          | 93%/4 h        |
| SnO$_2$ NPs [92]      | Sol-gel                                                   | 650 °C/4 h                                             | 20                                      | 0.5                                      | CR          | UVA         | 61.53%/1.66 h  |
| SnO$_2$ NPs [40]      | Green solution synthesis                                   | 400 °C/1 h                                             | 0.75                                    | 1.0                                      | MO          | MB          | (MO) 100%/0.33 h |
| SnO$_2$ NPs [93]      | Green solution synthesis using Tinospora Cordifolia extracts | 700 °C/2 h                                             | 4.61                                    | 0.21                                     | RhB         | EBT         | 77%/4.5 h       |
| SnO$_2$ NPs [91]      | Green solution synthesis                                  | 400 °C/2 h                                             | 20                                      | 2.0                                      | RhB         | UV          | 99.9%/0.75 h   |
| SnO$_2$ NPs [78]      | Green solution synthesis                                   | 100 °C/6 h                                            | 15                                      | 0.4                                      | R6G         | Simulated sunlight | 99.7/6.5 h   |
| SnO$_2$ microflowers [95] | One-pot hydrothermal                                     | 190 °C/24 h                                           | 10                                      | 1.0                                      | RbB        | UV          | 99%/2 h        |
| SnO$_2$ nanocrystals [96] | Solution phase growth technique                          | 200 °C/24 h                                           | 5                                       | 0.5                                      | RbB         | UV-visible  | 97.7%/2 h       |
| SnO$_2$ NPs [97]      | Electrospinning by precursor solution                     | 120 °C/48 h                                            | 5                                       | 1.33                                     | MO          | MB          | (MO) 92%/4 h    |
| SnO$_2$ nanorods [85] | Chemical                                                  | 550 °C/4 h                                             | 20                                      | 0.6                                      | MO          | UV          | 52%/1 h        |
| SnO$_2$ multilayered films [93] | Microwave hydrothermal                                  | 800 °C/2 h                                            | 10                                      | -                                        | RhB         | UV          | 100%/4 h       |
| SnO$_2$ thin films [88] | SPD using microscopy                                     | 500 °C/6 h                                             | 3.99                                    | -                                        | MB          | MO          | (MB) 42%/6 h    |
| SnO$_2$ thin films [80] | Ultrasonic spray pyrolysis technique                     | 450 °C                                                 | -                                       | -                                        | MB          | UV          | 98%/0.66 h     |

* Pollutants: RY186—Reactive yellow 186; MB—Methylene blue; MO—Methyl orange; MG—Malachite green; MR—Methylene red; CR—Congo red; RhB—Rhodamine B; R6G—Rhodamine 6G; EBT—Eriochrome Black T.
It is known that, to design an efficient system, photocatalysts usually need to meet some requirements, such as appropriate band gaps for light absorption, effective charge of carriers’ separation, and appropriate VB and CB edge potentials. However, it is difficult for pure SnO$_2$-based photocatalysts to satisfy all of them. In this sense, doping SnO$_2$ with different cations and the formation of SnO$_2$-based composites with other materials have attracted interest, as they drive other possibilities of photocatalytic studies. Therefore, discussion of different works concerning the photocatalytic properties of doped SnO$_2$ and SnO$_2$-based composite is given in the following sections.

4.2. Doped SnO$_2$ Photocatalysts

Although pure SnO$_2$ nanoparticles with a different morphology have shown efficiency in the degradation of dyes under irradiation, different authors have developed strategies to overcome the low photoactivity of SnO$_2$ under visible light exposure. For instance, doping SnO$_2$ with different foreign ions has shown to be an efficient way to shorten its band gap and enhance its photocatalytic activity.

Based on this fact, N. Mala et al. [98] synthesized SnO$_2$ nanoparticles doped with Mg$^{2+}$ + Co$^{3+}$ cations by a low-cost chemical solution method and investigated the antibacterial activity and photocatalytic efficiency toward the degradation of Methylene blue (MB) and Malachite green (MG) dyes. The authors revealed that the samples presented a tetragonal crystalline phase, with an average crystallite size of 24 and 25 nm for pure SnO$_2$ and SnO$_2$-Mg:Co, respectively. The authors suggested that this slight increase of the crystallite size after doping was due to local distortions in the SnO$_2$ lattice induced by the presence of dopants. A nanorod-like morphology was confirmed through SEM images, with a reduction in the crystal length and in the average diameter after doping. Surprisingly, an increase in the band gap energy estimated for SnO$_2$ (3.52 eV) and SnO$_2$-Mg:Co (4.22 eV) was observed. This behavior is attributed to the quantum confinement effect that normally happens when the nanoparticle size decreases. However, no meaningful variation was observed in the particle size for pure and doped SnO$_2$ samples. Regarding the photocatalytic activity of SnO$_2$ and SnO$_2$:Mg:Co nanoparticles, the authors observed that SnO$_2$ presented an efficiency of 82 and 86%, while SnO$_2$-Mg:Co displayed 89 and 92% efficiency toward MB and MG dyes degradation, respectively, under visible light after 60 min. The authors explained that three factors are responsible for the increase in the photocatalytic efficiency of doped SnO$_2$, which are: prevention of the recombination of electron–hole pairs photogenerated by surface defects, generation of greater number of oxidative species ($\cdot$OH, O$_2^-$, and H$_2$O$_2$), and particle size reduction.

Chu et al. [99] synthesized Bi$^{3+}$-doped SnO$_2$ by the hydrothermal method at 180$^\circ$C for 24 h, with a variation of bismuth molar content (3, 5 and 7%). The Rhodamine B (RhB) and Ciprofloxacin hydrochloride (CIP) were used as target molecules to evaluate the photocatalytic activity of the synthesized materials under simulated sunlight. XRD analysis confirmed the cassiterite tetragonal phase for all the samples, with no secondary phases, confirming that Bi$^{3+}$ is dissolved into the oxide crystal lattice by replacing Sn$^{4+}$ during the synthesis. The Bi$^{3+}$/Sn$^{4+}$ replacement in SnO$_2$ was confirmed by HR-TEM, UV–vis DRS, and XPS measurements. The average crystallite sizes decreased as a function of doping from 5.3 nm in SnO$_2$ to 3.3 nm in Bi-SnO$_2$(7%). In addition, the band gap ($E_g$) values showed a subtle variation of 3.72, 3.75, and 3.78 eV for Bi-SnO$_2$(3%), Bi-SnO$_2$(5%), and Bi-SnO$_2$(7%) samples, against 3.86 eV for the pure one. The authors explained that the introduction of new levels in the band gap of materials can act as a trap center for electron and hole, reducing charge recombination, which is beneficial to improve photocatalytic activity. By using PL spectroscopy, authors confirmed the lower recombination charge rate in the Bi-SnO$_2$(5%) sample for presenting the lowest PL emission among all samples. Regarding the photocatalytic activity, Bi-SnO$_2$(5%) showed an efficiency of 98.28% of RhB dye degradation after 100 min and 92.13% of CIP degradation after 90 min under irradiation. The excellent photodegradation efficiency of the doped samples was due to the increase in light absorption, as well as the effective separation and migration of photogenerated
charge carriers. All the results also indicated that there is an ideal amount of Bi$^{3+}$ doping to optimize the mentioned characteristics in order to enhance the SnO$_2$ material functionality.

Although doped SnO$_2$ is most prepared in powder, thin films based on doped SnO$_2$ have also been studied in photocatalysis. For instance, S. Vadivel and G. Rajaraja et al. [84] prepared magnesium-doped SnO$_2$ films by the chemical bath deposition method, varying Mg$^{2+}$ molar concentrations (1, 5, and 10%). The films were deposited on glass, and after deposition they were annealed at 500 $^\circ$C for 5 h in air to promote crystallization. From XRD analysis, the tetragonal rutile phase was confirmed in all films. Atomic force microscopy (AFM) images revealed that the surface roughness decreases with increasing dopant concentration. The optical band gap energy for pure SnO$_2$ was 3.63 eV, decreasing to 3.42 eV for the film doped with 10% Mg. The photocatalytic activities of the films were evaluated by the degradation of Methylene blue (MB) and Rhodamine B (RhB) dyes under UV irradiation. The maximum photodegradation of the dyes was reached for 10% Mg-doped SnO$_2$ film, degrading 80% of MB and 90% of RhB after 120 min. Fast electron transfer and high efficiency in electron–hole pairs separation led to a significant improvement of photocatalytic activity in the doped sample.

In the work conducted by Haya et al. [82] films of pure SnO$_2$ and doped with 2, 4, 6, and 8% of Sr$^{2+}$ were prepared by a chemical solution deposition method using the sol-gel method to deposit the solution coating on a glass substrate. The effect of doping on the structural, optical, morphological, and photocatalytic properties of the films were studied. According to the results, the increase in Sr$^{2+}$ doping promotes a decrease in crystallite size and an increase in the lattice distortion. These effects generate a greater number of defects, such as grain boundaries, micro-stresses, and displacements in the thin film lattice. The average crystallite size decreased from 7.61 nm for undoped SnO$_2$ to 3.80 nm for 8% Sr-SnO$_2$. It was also observed by UV-visible analysis that the presence of dopants introduced new intermediate levels in the semiconductor band gap ($E_g$), decreasing $E_g$ from 3.86 eV for pure SnO$_2$ film to 3.76 eV for Sr-richer SnO$_2$ film. Additionally, the morphology of the films was analyzed by AFM, and a smaller grain size was observed for 8% Sr-SnO$_2$ (4.96 nm). Consequently, it showed the lower surface roughness when compared to the other films. Concerning the photocatalytic activity, the greatest efficiency in the degradation of MB dye under irradiation was attained for 8% Sr-SnO$_2$ film, which was attributed to smaller grain sizes and surface roughness, as well as the introduction of new energy levels below the conduction band of the pure material, resulting from the Sr doping.

Using a non-conventional method to prepare thin films, Loyola Poul Raj et al. [83] prepared SnO$_2$ thin films doped with 3 and 6 mol% of Tb$^{3+}$ on a glass substrate by the spray nebulized pyrolysis (NSP) method and calcined them at 400 $^\circ$C to crystalize the materials. The authors investigated the photocatalytic property of the films in the degradation of MB dye under UV irradiation. According to the authors, doping SnO$_2$ films with up to 6% of Tb$^{3+}$ cations induces a decrease in the grain size from 80 to 56 nm, and the band gap from 3.51 to 3.36 eV, which directly impacts photocatalysis, as reported by other authors. Indeed, a maximum of 85% of dye degradation was observed after 120 min under UV irradiation using SnO$_2$ film doped with the 6 mol% Tb$^{3+}$. Using PL spectroscopy, the authors reveal that Tb doping leads to the creation of more defects that act as reactive sites for catalyzed reactions. As a consequence of the study, the authors concluded that Tb doping favored photocatalytic reactions by reducing particle size, and therefore increasing the surface area and the number of reactive sites on the surface, which allows the dye adsorption. In addition, Tb doping induces a decrease in the band gap of the materials, which favors photo-absorption, aiming to potentialize charge carriers to participate in photocatalytic reactions.

Other studies based on the synthesis of doped SnO$_2$ catalysts and their applications in the photodegradation of different organic dyes are listed in Table 3.
Table 3. Summary of the studies for doped SnO$_2$-based photocatalysts applied in the photodegradation of different organic dyes.

| Catalyst Type | Synthesis Method | Synthesis Conditions (Temperature/Time of Calcination) | Dye Solution Concentration (mg L$^{-1}$) | Photocatalyst Concentration (g L$^{-1}$) | Pollutant * | Irradiation | Efficiency/Time |
|---------------|------------------|--------------------------------------------------------|------------------------------------------|-------------------------------------------|-------------|-------------|----------------|
| SnO$_2$:Mg$^{2+}$:Co$^{3+}$ NPs [84] | Wet chemical method | 550 °C/2 h | - | 0.5 | MB | MG | (MB) 89%/1 h (MG) 92%/1 h |
| SnO$_2$:Sr$^{2+}$ thin films [82] | Sol-gel method using a dip-coating technique | 500 °C/2 h | 5 | - | MB | UV | 37.90%/2 h |
| SnO$_2$:Tb$^{3+}$ thin films [83] | Nebulized spray pyrolysis (NSP) technique | 400 °C | 3.2 | - | MB | UV | 85%/2.08 h |
| SnO$_2$:Bi$^{3+}$ quantum dots [99] | One-step hydrothermal | 180 °C/24 h | 20 | 1.0 | RhB | UV | 98.58%/1.66 h |
| SnO$_2$:Mg$^{2+}$:Co$^{3+}$ thin films [84] | Chemical bath deposition | 500 °C/2 h | 15 | - | MB | UV | (MB) 80%/2 h |
| SnO$_2$:Co$^{3+}$ [100] | Chemical solution | 400 °C/2 h | 60 | 0.2 | MB | UV-visible | 95.38%/2 h |
| SnO$_2$:Ni$^{2+}$ NPs [101] | Chemical precipitation | 410 °C/2 h | 5 | 0.15 | BG | UV | 97.54%/1.75 h |
| SnO$_2$:Zn$^{2+}$ NPs [102] | Combustion | 600 °C/8 h | 10 | 1.0 | MB | UV | 9.6%/1 h |
| SnO$_2$:Zr$^{4+}$ NPs [103] | Co-precipitation | 200 °C/24 h | 20 | - | MO | UV | 89.6%/3 h |
| SnO$_2$:Mn$^{2+}$ nanowires [104] | Co-precipitation | 400 °C/4 h | 61 | 1.0 | NBB | Solar irradiation | 99%/3 h |

* Pollutant: BG—Brilliant green; NBB—Naphthol blue black; MB—Methylene blue; MO—Methyl orange; MG—Malachite green; CR—Congo red; RhB—Rhodamine B.
4.3. SnO$_2$-Based Composite Photocatalysts

As one can see from studies discussed in the sections above, pure and doped SnO$_2$ particles and films have been well explored. However, SnO$_2$ has also been combined with other different semiconductors in order to reduce recombination of the photoinduced charge carriers, to therefore improve photocatalytic activity.

Considering this fact, Abdel-Messih et al. [105] synthesized SnO$_2$/TiO$_2$ nanoparticles with a spherical mesoporous morphology synthesized by the sol-gel process, using polymethylmethacrylate as a template. The amount of SnO$_2$ (0–25%) in relation to the mass of pure TiO$_2$ was varied to obtain composites with different compositions. The samples were calcined at 800 °C for 3 h to ensure complete organic polymer decomposition. In relation to the photocatalytic property of the materials, photodegradation of Rhodamine B (RhB) dye was performed under UV irradiation using a catalyst/dye concentration of 1 g L$^{-1}$. The photodegradation efficiency of the composites increased with the increase of tin oxide content up to 10% (about 92% of the dye degraded after 3 h). However, the sample with 25 mol% of SnO$_2$ showed the lowest efficiency, which was attributed to the loss of the titanium anatase phase. The authors concluded that there is an optimal amount of SnO$_2$ to achieve the maximum efficiency. In addition, the remarkable reduction in particle size by the existence of SnO$_2$ in the composites enhanced the oxidizing power and extended the photoinduced charge separation, and these were the main reasons for the increase in the catalytic activity of the samples.

Das et al. [36] prepared Sn/SnO$_2$ nanocomposites by the precipitation method, followed by carbothermal reduction and calcination at 800 °C for 2 h. The authors investigated the photocatalytic property of Sn/SnO$_2$ composites in the degradation of methylene blue under UV irradiation using a catalyst concentration of 0.5 g L$^{-1}$. It was found that there was a maximum efficiency of 41% for pure SnO$_2$ after 210 min under irradiation, while the Sn/SnO$_2$ composite showed a higher photocatalytic activity of 99%. The highest efficiency observed for the composite was related to the role of Sn on the surface of SnO$_2$ nanoparticles. As the Fermi energy level of Sn is higher than that observed for SnO$_2$ due to its lower work function, when metallic Sn is bound on the surface of SnO$_2$ nanoparticles, electrons migrate from Sn to SnO$_2$ to reach Fermi-level equilibrium. The effect of the pH solution on the photocatalytic efficiency of the composites was also evaluated. The pH had a direct influence on the photocatalytic process, being the neutral pH favorable for the degradation of MB dye. Finally, the authors investigated the reusability of the composites after three cycles and confirmed that the photocatalyst is stable, but gradual loss in efficiency was observed due to the loss of the material during recovery processes.

Li et al. [85] synthesized carbon-coated, mixed-phase (tetragonal/orthorhombic) SnO$_2$ (i.e., tetragonal/orthorhombic) nanorods photocatalysts by a combined chemical precipitation and hydrothermal method at 180 °C for 6 h. The SnO$_2$-C composite with a SnO$_2$ tetragonal phase was obtained after calcination of the hydrothermal products at 550 °C for 4 h. The photocatalytic activities of the samples were investigated toward the degradation of Methyl orange (MO) dye under UV irradiation using a catalyst/dye concentration of 0.6 g L$^{-1}$. The as-prepared mixed-phase SnO$_2$ showed a photodegradation activity of 52% against an efficiency of 39% for pure SnO$_2$ with a tetragonal phase after 60 min under irradiation, indicating the influence of different phases on the junction formation to tune photocatalytic activity. Coating mixed-phase SnO$_2$ nanorods with carbon provided a degradation activity of 98%. The tetragonal/orthorhombic-SnO$_2$ material exhibits very high stability after three cycles, remaining about constant without apparent deactivation. Photocatalytic activity was not primarily attributed to the narrower band gap or visible light absorption tail. By demonstrating that the transfer and separation of photogenerated electron–hole pairs are improved by the introduction of a carbon layer in interparticle space. To understand the photocatalytic mechanism, different scavengers were used in the study—triethanolamine (TEOA), tert-butyl alcohol (TBA), and benzoquinone (BQ). The results indicated that the species h$^+$, O$_2^-$, and HO$^*$ played important roles in the degradation of MO.
Constantino et al. [97] synthesized a porous composite based on SnO$_2$/cellulose acetate with the electrospinning method and calcined it at 120 °C for 48 h. The photocatalytic activity of the nanocomposites was studied toward the degradation of MO and MB dyes under UV irradiation using a catalyst/dye concentration of 1.3 g L$^{-1}$. The photocatalytic efficiency of SnO$_2$/cellulose was approximately 92% of MO degradation after 210 min and 95% of MB degradation after 240 min. It is worth mentioning that the photodegradation process did not alter the average diameter and morphology of the fibers as well as their surface chemistry. From TOC analysis, the authors evidenced that only 54% and 79% of MO and MB dye are mineralized after the photocatalytic process. However, presence of other compounds as by-product of dye degradation was confirmed by LC–MS.

Silva et al. [37] synthesized spherical nanoparticles and microrods of Ag$_3$PO$_4$/SnO$_2$ composites, by the in situ coprecipitation method, with various molar ratios of 5, 10, 15, and 20% of SnO$_2$ in relation to the mass of pure Ag$_3$PO$_4$, followed by calcination at 350 °C for 2 h. The photocatalytic performance of the samples was investigated by the degradation of Rhodamine B (RhB) dye under visible light irradiation using a catalyst/dye concentration of 0.6 g L$^{-1}$. The authors observed superior photocatalytic activity for all the composites when compared to pure Ag$_3$PO$_4$. The authors evidenced that the excess of SnO$_2$ damaged the interfacial contact between the Ag$_3$PO$_4$ and SnO$_2$, which was due to the high degree of particle agglomeration. The photocatalytic mechanism involved in the photodegradation of the dye was also investigated for pure Ag$_3$PO$_4$ and Ag$_3$PO$_4$/SnO$_2$-15%. It has been confirmed that the photogenerated holes participated in the direct degradation of RhB when Ag$_3$PO$_4$ was a photocatalyst. On the other hand, there was a significant participation of O$_2^*$ radicals when Ag$_3$PO$_4$/SnO$_2$-15% is used. The highest photodegradation efficiency presented by the Ag$_3$PO$_4$/SnO$_2$-15% composite was confirmed by total organic carbon (TOC) analysis. Reusability tests were also performed for Ag$_3$PO$_4$/SnO$_2$-15% and a loss of 43.2% of its photocatalytic efficiency was observed after the third cycle, which was similar to that observed for pure Ag$_3$PO$_4$. Using the X-ray diffraction technique, the presence of Ag in the composition of the samples was observed after the photocatalysts were used in photocatalysis.

Apart from the above-mentioned studies performed using composite particles, SnO$_2$-based composite photocatalysts have also been explored as films. For instance, porous SnO$_2$/TiO$_2$ films were prepared using Ar-assisted, modified thermal evaporation, followed by the atomic layer deposition (ALD) technique at 300 °C in the reaction chamber [62]. To prepare SnO$_2$/TiO$_2$ films, TiO$_2$ layers were deposited on porous SnO$_2$ nanofoam, with those previous deposited on 2 × 2 cm$^2$ Si (100) wafers or ITO substrates, with variation in deposition cycles of TiO$_2$ (10, 25, 50, and 100 cycles) by ALD. The samples were denoted SnO$_2$/TiO$_2$-10, SnO$_2$/TiO$_2$-25, SnO$_2$/TiO$_2$-50, and SnO$_2$/TiO$_2$-100, respectively. After the TiO$_2$ deposition, the material was calcined at 700 °C for 1 h. The photocatalytic properties of the films were evaluated by the degradation of MB at a concentration of 1.2 mg L$^{-1}$ under UV irradiation. The nanofoam heterostructures showed higher photocatalytic activity when compared to the porous SnO$_2$ nanofoam. SnO$_2$/TiO$_2$-50 nanofoam, which exhibited the highest efficiency, reached to 99% of MB degradation after 300 min. The authors correlated this fact to being due to a synergistic effect occurring between SnO$_2$ and TiO$_2$, and due to the sparse deposition of the TiO$_2$ layer on porous SnO$_2$. Separation of charge carriers due to the potential difference between SnO$_2$ and TiO$_2$ increases the lifetime of the charge and improves the interfacial charge transfer to the species adsorbed on the surface. This phenomenon, along with the strong oxidant *OH radicals formed in the VB of the TiO$_2$ layer, improves photocatalytic efficiency of the SnO$_2$/TiO$_2$ heterostructure.

Other important works reporting the activity of SnO$_2$-based composite photocatalysts for the degradation of dyes are summarized in Table 4.
Table 4. SnO$_2$-based composites formed with different semiconductors for photodegradation of organic dyes.

| Catalyst Type | Synthesis Method | Synthesis Conditions (Temperature/Time of Calcination) | Dye Solution Concentration (mg L$^{-1}$) | Photocatalyst Concentration (mg L$^{-1}$) | Pollutant $^*$ | Irradiation | Efficiency/Time |
|---------------|------------------|------------------------------------------------------|------------------------------------------|------------------------------------------|----------------|-------------|-----------------|
| CrO$_4$-SnO$_2$ spherical NPs [106] | Coprecipitation | 150, 300 and 450 $^\circ$C/3 h | $1 \times 10^{-4}$ mol L$^{-1}$ | 1.6 g L$^{-1}$ | TB | Sunlight | 80%/1 h |
| g-C$_3$N$_4$/SnO$_2$ nanosheets decorated with NPs [107] | Microwave-assisted hydrothermal method | 550 $^\circ$C/2 h | 10 mg L$^{-1}$ | 0.5 g L$^{-1}$ | RhB | UV and visible lights | 100%/4 h (UV) and 98.5% (visible light) |
| Zn$_2$SnO$_4$-SnO$_2$ and Zn$_2$SnO$_4$-Sn spherical NPs [108] | Solid state reaction | 800 $^\circ$C/1 h | 20 mg L$^{-1}$ | 0.2 g L$^{-1}$ | MB | UV | 92%/2 h |
| Polyaniline SnO$_2$ nanoneedles and nanograins [109] | Chemical oxidation polymerization method using aniline monomer | 500 $^\circ$C/2 h | 50 mg L$^{-1}$ | 1 g L$^{-1}$ | RY | UV | 96%/1 h |
| ZnO-SnO$_2$ thin films [81] | Sol-gel | 550 $^\circ$C/1 h | 16 mg L$^{-1}$ | - | MeG | UV | 42%/45 min |
| ZnO-SnO$_2$/NPs hexagonal nanopillar [110] | Sol-gel | 600 $^\circ$C/2 h | 10 mg L$^{-1}$ | 0.025 g L$^{-1}$ | MO | UV | 91.78%/3 h (MO) |
| CuCr$_2$O$_4$/SnO$_2$ [111] | Sol-gel/solid state reaction | 900 $^\circ$C/6 h and 600 $^\circ$C | 15 mg L$^{-1}$ | 1 g L$^{-1}$ | CV | Sunlight | 100%/1.5 h |
| SnO$_2$/GO spherical NPs [112] | Sonochemical method | 180 $^\circ$C/6 h and 100 $^\circ$C/6 h | $1 \times 10^{-5}$ mol L$^{-1}$ | 0.5 g L$^{-1}$ | RbB | TDM | 95%/2 h (RbB) |
| SnO$_2$/Zn$_2$SnO$_4$ cube-like NPs [113] | Hydrothermal | 150 $^\circ$C/12 h and 700 $^\circ$C/2 h | $10^{-5}$ mol L$^{-1}$ | 1 g L$^{-1}$ | MB | MO | 97.1%/2.5 h (MB) |
| SnO$_2$-MoS$_2$ spherical NPs [114] | Sonochemical liquid exfoliation method | 80 $^\circ$C/2 h | 100 ppm | 1 mL of SnO$_2$-MoS$_2$ added in 50 mL of dye solution | MR | Visible light | 94.0%/2 h (MR) |
| SnO$_2$-WO$_3$ NPs [115] | Green combustion method | 500 $^\circ$C/1 h | 5 ppm | 0.8 g L$^{-1}$ | MB | Visible light | 70%/3 h |

$^*$ Pollutant: CV—Crystal violet; CR—Congo red; EBT—Eriochrome black T; MB—Methylene blue; MO—Methyl orange; MeG—Methyl green; MR—Methylene red; RbB—Rhodamine B; RY—Remazol yellow; TB—Trypan Blue; TDM—Textile dye wastewater.
Most of the authors reported that the number of materials for the formation of the SnO$_2$ with a different particle size and morphology, besides doped SnO$_2$ with an appropriate amount and type of dopant, and also the formation of the composite with SnO$_2$ plays an important role in improving the photocatalytic activity of the SnO$_2$ material. In relation to composites, the excess of both species can be harmful to the contact surface between the phases, mainly due to the high degree of particle agglomeration. Tests using scavengers, such as p-benzoquinone (BZ, C$_6$H$_4$O$_2$), isopropanol (ISO, (CH$_3$)$_2$CHOH), and ammonium oxalate monohydrate (AO, (NH$_4$)$_2$C$_2$O$_4$·H$_2$O) indicate •OH is the main species in most photocatalytic mechanisms. However, to obtain more insights about the photocatalytic mechanism involved in composite materials, we seek to understand the charge transfer between the phases from the band structures of each individual material. Structural and electronic defects can also generate energy levels between the VB and CB, and, therefore, modify the photocatalytic mechanism of composites. The creation of different interfaces between the phases may reduce charge carriers’ recombination, leading to the formation of a great number of free radicals to improve photocatalysis. In addition, several other parameters can impact the photocatalytic efficiency of composites, such as phase composition, surface area, morphology, particle size, pore structure, electron–hole recombination rate, and band gap energy of the individual components. Some authors showed that the high surface area and the presence of pores are more effective parameters that affect dye degradation since the existence of several active sites, responsible for the adsorption of molecules, is crucial for the photocatalysis to occur.

Based on the findings above, it can be concluded that to design a new photocatalytic material with specific characteristic, one has to consider optimizing type and amount of dopants and interface characteristics between materials, or even the nature of the desired product (powder, film, etc.), besides the microstructure of the material (particle size and morphology), and by a choice of specific synthesis methodology and appropriate experimental conditions.

5. Conclusions and Final Remarks

From this review work, it was possible to evaluate the importance of synthesis methods and experimental parameters in obtaining tin-oxide-based materials with high performance in heterogeneous photocatalysis of persistent organic pollutants, more specifically, organic dyes. The search for new materials and methodologies that provide efficient results for the remediation of such pollutants has been one of the great challenges for the scientific community. Among the studied promising materials, SnO$_2$ has shown excellent results as a catalyst in heterogeneous photocatalysis processes due to its intrinsic characteristics, which have been responsible for the material’s conductivity, optical and electrical properties, and high thermal stability. As a consequence of the choice of the synthesis method and experimental conditions, it was possible to evidence different morphology, particle size, surface area, structural modifications, optical bandgap energy, and surface and bulk defects, and, therefore, obtain excellent results in the application of pure and modified SnO$_2$ toward the degradation of persistent organic pollutants (POPs). In general, SnO$_2$-based photocatalysts have shown promising efficiency for degrading a series of different organic dyes.

Considering pure SnO$_2$ catalysts, synthesis conditions may especially influence particle size and morphology, specific surface area, crystallinity, and the presence of electronic defects on surface and bulk of the materials, which are important parameters to change photocatalytic efficiency under both UV and visible irradiation. With respect to doped SnO$_2$, the type and number of dopants may introduce different intermediate levels within the gap, decreasing band gap energy to improve the photo-absorption in visible light. Finally, it has been demonstrated that the composite formed with SnO$_2$ is responsible for band structure alignment and improvement of charge separation that led to an increased photocatalytic activity when compared to the individual components. It is still important to highlight that the study of the reaction mechanism involved in the dye degradation is an
important aspect, which allows the design of new and efficient SnO$_2$-based photocatalysts, and an understanding of their laminations in order to use them in practical devices.

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