Review

Advances in the Application of Nanocatalysts in Photocatalytic Processes for the Treatment of Food Dyes: A Review

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Abstract: The use of food additives (such as dyes, which improve the appearance of the products) has become more prominent, due to the rapid population growth and the increase in demand for beverages and processed foods. The dyes are usually found in effluents that are discharged into the environment without previous treatment; this promotes mass contamination and alters the aquatic environment. In recent years, advanced oxidation processes (AOPs) have proven to be effective technologies used for wastewater treatment through the destruction of the total organic content of toxic contaminants, including food dyes. Studies have shown that the introduction of catalysts in AOPs improve treatment efficiency (i.e., complete decomposition without secondary contamination). The present review offers a quick reference for researchers, regarding the treatment of wastewater containing food dyes and the different types of AOPs, with different catalyst and nanocatalyst materials obtained from traditional and green chemical syntheses.

Keywords: advanced oxidation processes; food dyes; nanomaterials; nanocatalyst; green nanocatalyst

1. Introduction

Food dyes are used worldwide and are classified as natural, semi-synthetic, and synthetic [1]. Owing to their stability, performance, and low cost—synthetic and semi-synthetic dyes are increasingly being used in the food industry [2]. However, these compounds are not eco-friendly as they are resistant to biological oxidation (complex molecular structure) and are, therefore, environmental concerns [3]. There are more than 100,000 commercial dyes available, of which approximately 10–15% of the dyes usually enter the environment through industrial effluents without prior treatment [4]. When these effluents are mixed with clean water they can unbalance the recommended level of organic and inorganic parameters in water bodies [5], impeding the processes of self-purification and biological degradation due to limited light transmittance [6]. Moreover, due to their recalcitrant nature, xenobiotic, highly toxic, and carcinogenic properties, they end up affecting the health of people and ecosystems in general [1,2,7–10].

AOPs are being studied to reduce the presence of dyes in wastewater, to promote the degradation of the pollutant until its mineralization, and finally, to obtain a regenerated effluent [11]. Within the AOPs, homogeneous processes are researched, which are widely used in the elimination of dyes. However, their reactions take place in the same phase, making it difficult to separate them from the reaction medium, in addition to presenting drawbacks, such as high operational costs and energy requirements, acidification of the effluent (Fenton process), and the need to combine with other oxidative processes to improve their degradation yield. Alternatives to improve the degradation performance have been proposed, and, in turn, the operating conditions of the process, such as the use of heterogeneous processes with the application of catalysts (e.g., metal oxides, titanium dioxide, etc.); however, there is a risk of contamination due to the precipitation of toxic metals during the treatment. Therefore, to counteract these potential problems, the use of catalysts (in nanometer sizes) has emerged as a viable alternative due to their unique properties (e.g., large surface area and low concentration) [12].

Nanomaterials are attracting attention due to the degradation of toxic organic pollutants. Unlike conventional catalysts, nanocatalysts can achieve complete mineralization of contaminants in an advanced oxidation process, owing to their chemical reactivities, catalytic activities, electron flow, and other complementary properties, such as physico-chemical and thermal stability and large surface areas [13–15]. In addition, they decrease operating costs since they can be reused during several reaction cycles [16]. This paper aims to perform a literature review on the degradation of food dyes by applying AOPs, focusing on catalysts and nanocatalysts.

2. Dyes in the Food Industry

Dyes are complex unsaturated aromatic compounds that exhibit good color, intensity, and solubility [17]. For their part, food dyes differ in origin (vegetable, mineral, or insect sources, to those chemically synthesized [18]), and have been established as necessary for the processing of various foods. Scotter [19] classified food dyes into six groups according to their chemical structures: Azo (monoazo, diazo, and triazo), triarylmethane, azopyrazolone, xanthene, quinoline, and indigoid. Out of the above, only 45–47% are biodegradable [17,20]. Table 1 presents a series of synthetic dyes, grouped according to Scotter [19] and approved by the European Union (E Number) and the United States of America. Dyes that have the greatest applications in the food industry, as well as the adverse effects they could cause if surpassing the average daily intake (ADI) limit specified by the European Food Safety Authority (EFSA), are listed as references.

Table 1. Synthetic food dyes classification.

Type of Structure	Name	2D Chemical Structure	Color	E-Number	* ADI	FD&C	Application	Hazardous Effect	Ref.
Azo	Brown FK		Orange-brown	E 154	0.14	None	Single-use for coloring herring	Azo dyes, in general, can produce hyperactivity, restlessness, and sleep disturbances in children; when the dyes are combined with other food additives, such as sodium benzoate, it could lead to possible neurodevelopmental impairment and carcinogenic properties.	[21,22]
Monoazo	Sunset Yellow FCF		Yellow-orange	E 110	4	Yellow No. 6	Soft drinks, confectionery, jams, jellies, cookies, ice cream, potato chips, instant pasta, food decorations, and coatings.	Non-alcoholic flavored beverages. Confectionery products. Yogurt.	[22,23]
	Carmoisine		Red to maroon	E 122	4	None	Non-alcoholic flavored beverages. Confectionery products. Yogurt.		
	Amaranth		Red	E 123	0.15	None	Alcoholic beverages. Candied fruits.		
	Ponceau 4R		Red-orange	E 124	0.7	None	Soft drinks. Confectionery. Coatings. Cookies and ice cream. Fine bakery products.		

Table 1. Cont.

Type of Structure	Name	2D Chemical Structure	Color	E-Number	* ADI	FD&C	Application	Hazardous Effect	Ref.
	Allura Red AC		Yellowish red	E 129	7	Red No. 40	Potato chips. Cookies, candies, ice cream, and jams. Non-alcoholic flavored drinks and yogurts.		[22,26]
	Lithol Rubine BK		Red-blue	E 180	1.5	Prohibited	Cheese surface coating.		[27]
	Citrus red		Scarlet red	Prohibited	-	Red No. 2	Orange peel coating.		[28]
Diazo	Brilliant Black BN		Blue-black	E 151	1–5	None	Confectionery products. Decorations and coatings. Fine bakery products. Ice cream and desserts.	Clastogenic and/or aneugenic properties, damage DNA.	[22,29]
	Brown HT		Dark brown	E 155	1.5	None	Soft drinks, spirits, fruit wines, cider, among others.	Possible reduction of body weight and cholesterol (HDL). Increase of hepatic enzymes in the blood. Negative effects on hypothalamic-pituitary-testicular axis function.	[22,30,31]

Table 1. Cont.

Type of Structure	Name	2D Chemical Structure	Color	E-Number	* ADI	FD&C	Application	Hazardous Effect	Ref.
Triarylmethane	Patent Blue V		Violet blue	E 131	5	None	Surface coating of cheese and edible casings. Alcoholic beverages, soft drinks, and other food products.	Anaphylactic reactions. Alterations of hematological parameters.	[32]
	Brilliant Blue FCF		Greenish blue	E 133	6	Blue No.1	Dairy products. Sweets. Beverages.	Carcinogenic effect.	[33,34]
	Green S		Blue-green	E 142	5	None	Confectionery products. Decorations and coatings. Ice cream, desserts. Processed peas.	Mild anemia (transient), increased cecal weight, thyroid degeneration, enlarged lymph nodes in the intestinal wall.	[35]
	Fast Green FCF		Blue-green	Prohibited	-	Green No. 3	Peas. Vegetables. Fish. Desserts, dry bakery mixes, and sauces.	Chromosomal aberration. Inhibition of neurotransmitter release.	[34,36]
Azopyrazolone	Tartrazine		Yellow	E 102	7.5	Yellow No.5	Desserts and sweets. French fries. Muesli, corn flakes, noodles, tartar sauce, mustard, and bouillon cubes. Energy drinks	Migraine. Asthma. Skin conditions (urticaria).	[22,36]

Table 1. Cont.

Type of Structure	Name	2D Chemical Structure	Color	E-Number	* ADI	FD&C	Application	Hazardous Effect	Ref.
Xanthene	Erythrosine		Blue pink	E 127	0.1	Red No.3	Cocktails. Candied cherries.	Oncogenic effect on the rat thyroid gland.	[28,37]
Quinoline	Quinoline Yellow WS		Yellow-green	E 104	0.5	None	Sports and energy drinks. Ice cream and confectionery.	Urticaria. Rhinitis.	[36,38]
Indigo	Indigo carmine		Deep blue	E 132	5	Blue No. 2	Tablets and capsules. Coatings. Ice cream, cookies, candies. Confectionery.	Asthma.	[39]

* ADI, average daily intake (mg/kg body weight/day); FD&C, Federal Food, Drug, and Cosmetic Act.

From the aforementioned groups of dyes, azo dyes (azo, monoazo, and diazo) are considered high pollutants, and they represent 70% of all dyes. In addition, their biological degradations are difficult, with consequent inhibition of microbial activity and cell death when found in high concentrations in the effluent [40]. These are toxic and refractory to conventional treatment processes due to their resistance to physicochemical destruction and they lack biodegradability [41].

In addition, certain parameters of the receiving medium are altered, such as the color, pH, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), and suspended solids, among others. On the other hand, the interruption of the photosynthetic process in marine plants causes eutrophication and, thus, the uncontrolled release of mineral elements, such as nitrates, nitrites, and phosphates [41]. Therefore, in the search for new alternatives for the degradation of this type of dye, Wang and Xu [42] proposed the combination of AOPs (as pretreatment) to be subsequently treated with conventional biological processes (aerobic process), being considered efficient for their degradation. This is how AOPs have been introduced in recent years for the remediation of problems related to the contamination of the aquatic environment by dyes, allowing the removal of a wide range of organic and inorganic pollutants from water and wastewater [43].

3. Advanced Oxidation Process

The AOP is one of the most widely used processes for water treatment involving the formation of radicals, e.g., highly reactive hydroxyl radicals ($\bullet\text{OH}$), superoxide anion radical (O_2^-), hydroperoxyl radical ($\text{HO}_2\bullet$) or alkoxy radical ($\text{RO}\bullet$), with the $\text{HO}\bullet$ radicals attracting the most attention in this area, as they allow the degradation of organic matter for its complete mineralization to CO_2 and H_2O [44,45].

Specifically, the hydroxyl radical ($\bullet\text{OH}$) has an oxidation potential between 2.8 V (pH 0) and 1.95 V (pH 14) vs. saturated calomel electrode (SCE), the most commonly used reference electrode. Being non-selective, it reacts rapidly with various species of the order $10^6\text{--}10^{10} \text{ M}^{-1} \text{ s}^{-1}$ [46–48]. The mechanism of $\bullet\text{OH}$ radicals involves the reaction with organic contaminants via four basic pathways: (i) radical addition; (ii) hydrogen abstraction; (iii) electron transfer; and (iv) radical combination. In addition, carbon radicals ($\text{R}\cdot$ or $\text{R-OH}\cdot$) are generated and can be transformed into organic peroxy radicals ($\text{ROO}\cdot$) with O_2 [47,49]. Additionally, there is the combination of a strong oxidizing agent, such as ozone (O_3), transition metal ion catalysts, or photocatalysts, such as Fe^{2+} or TiO_2 , the latter being the most commonly used [50]. AOPs were first suggested in 1980 for drinking water treatment. However, in later years, their applicability mostly focused on oxidative treatments to different wastewaters. Thus, it was considered a new alternative for the elimination of organic pollutants classified as bio-recalcitrant and for the inactivation of pathogenic microorganisms that were difficult to treat by conventional methods [51–55].

Table 2 shows the source of hydroxyl radicals of homogeneous and heterogeneous AOPs, each process has its advantages and disadvantages, providing a thorough evaluation to select the most efficient process for each dye to be treated.

Table 2. Source of hydroxyl radicals of AOPs.

AOP	Source of Radicals OH	Advantages	Disadvantages
Electrochemical oxidation	Electricity, 2–20 A (water electrolysis)	High-energy efficiency. Operates at ambient conditions.	Selective
Sonochemical oxidation	Ultrasound 20 kHz–2 MHz (water sonolysis)	Does not require the addition of chemicals or catalysts. Operates at ambient conditions.	It needs to be combined with other oxidative processes to improve its degradation performance.

Table 2. Cont.

AOP	Source of Radicals OH	Advantages	Disadvantages
Sonoelectrochemistry	Electricity, 2–20A Ultrasound 20 kHz–2 MHz	Fast reaction rate. Guaranteed degradation efficiency. Increased energy efficiency.	Greater emphasis on the frequency and acoustic power, electrode positioning, cell geometry, and the distance between the tip of the ultrasonic horn and the electrode.
Ozonation	O ₃ O ₃ /UV O ₃ /H ₂ O ₂ O ₃ /H ₂ O ₂ /UV	Increased color removal. No sludge or hazardous by-products. In situ O ₃ generation. Effective at basic pH (lower environmental risk). Treatment is more efficient when combined with H ₂ O ₂ and/or UV. Complete mineralization with O ₃ /H ₂ O ₂ system.	High equipment cost and energy requirements. Mass transfer is the limiting factor of the process. Formation of bromate (carcinogen) by effluents with an excess of bromide ions.
Processes based on H ₂ O ₂	H ₂ O ₂ /UV	The formation of bromate is suppressed.	Lower mineralization rate.
	H ₂ O ₂ /Fe ²⁺ (Fenton)	Lower energy consumption than ozone. Does not form bromates.	Expensive. The strict control of H ₂ O ₂ /Fe ²⁺ ratio. Continuous addition of ferrous ions (high sludge deposition). An acidic medium is required for OH radical generation. pH 3 is the optimum operating pH (Fe(OH) ₃ precipitation at pH > 4).
	H ₂ O ₂ /Fe ³⁺ (Fenton-like) H ₂ O ₂ /Fe ³⁺ /UV (photo-Fenton-like)	Using Fe ³⁺ ion is less expensive than Fe ²⁺ .	Slow process. Limited applicability in environmental technologies.
	H ₂ O ₂ /Fe ²⁺ /UV (photo-Fenton)	The addition of UV light improves the degradation rate. Cyclic process, regeneration of ferrous ion (Fe ²⁺) by photo-reduction of ferric ions. Sludge formation is minimal.	The efficiency of the reaction depends on the lamp power and type of light.
	Electro-Fenton Photoelectro-Fenton	Efficient for complete degradation and mineralization of synthetic and real wastewaters.	Optimal at strong acid pH (pH 2.8–3.5). Non-recyclability of the catalyst used.
Heterogeneous photocatalysis	TiO ₂ /ZnO/CdS + UV TiO ₂ /UV/H ₂ O ₂ O ₃ /catalyst/light (Photocatalytic Ozonation)	More efficient than homogeneous catalysis. High availability and cost-effectiveness of photocatalysts. Non-selective catalytic activity. Applicability to a wide pH range. Does not generate sludge or toxic compounds.	Limited application to inorganic environments. Low chemical stability of the photocatalyst. High-energy consumption. Rapid deactivation/poisoning of the surface-active site. Spent photocatalyst is disposed of as secondary solid waste.

In general, homogeneous processes, such as electrochemical and Fenton, require less energy than heterogeneous processes. However, these processes are selective and lead to the formation of contaminants, such as iron oxides. Another limitation of homogeneous processes, such as in chemical and photochemical systems (H₂O₂/Fe³⁺ and H₂O₂/Fe³⁺/UV), involves their restricted applicability in environmental technologies. In addition, they have high operating costs. In terms of efficiency, the heterogeneous processes are more efficient,

and in some cases can reach the mineralization of the pollutant, which, in many cases, is not achieved in homogeneous processes, mainly in ozonation and $\text{H}_2\text{O}_2/\text{UV}$.

Homogeneous oxidative processes are highly dependent on the dosage of the reagent to the medium. Those using H_2O_2 will have to dose a specific amount to achieve a higher percentage of removal, which will depend on the initial concentration of the dye and the response to oxidation. Often, the amount to be dosed is measured by the theoretical stoichiometric ratio between H_2O_2 and COD. However, it is necessary to analyze the negative effects of the treatment before establishing the dilution ratio. In addition, the excessive use of H_2O_2 generates a residual that contributes to the increase in COD or acts as a scavenger of the hydroxyl radicals generated, slowing down the oxidation rate [6]. Additionally, it affects the microbial activity [56] and, consequently, the efficiency of the biological degradation that is usually employed as a post-treatment to the selected AOP. For all these reasons, the H_2O_2 dosage must be previously adjusted through laboratory-scale studies, so that the exact amount to be used in the effluent in question is obtained [57]. Homogeneous catalysts in AOPs are easy to apply, low cost, do not require complex processes to obtain them, present low resistance to mass transfer, facilitate the treatment of contaminants [58,59], and achieve complete degradation and partial mineralization of the dyes [60–63]. However, as they are in the same phase, their separation from the reaction medium with conventional processes is difficult and the cost per treatment rises [64].

Given the problems presented by homogeneous AOPs, the interest in heterogeneous processes is increasing (due to the generation and use of efficient catalysts) [65,66]. Based on this, a search was performed in the Scopus database (the year 2010–2021) on the application of heterogeneous AOPs for the degradation of food dyes (Figure 1). A total of 10,886 manuscripts were reported, where approximately 34% (3691 documents) corresponded to photocatalytic processes without the use of nanocatalysts. This shows an increase (66% of the total) in the use of nano-photocatalysts in the last decade.

Figure 1. The number of publications of photocatalytic processes using catalysts and nanocatalysts. (Source: Scopus database with keywords search: photocatalysis, nano, degradation, and food dyes).

These heterogeneous photocatalytic processes are already recognized as ‘green’ and viable strategies for environmental problems [11,67–69]. The basic mechanism of this process is the adsorption of light photons and the formation of electron holes [70]. To achieve the degradation of dyes, these processes generally involve three active agents—

hydroxyl radicals ($^{\circ}\text{OH}$), superoxide anion radicals ($^{\circ}\text{O}_2^-$), and holes (h^+) that participate in a series of photocatalytic reactions, described as follows:

However, most of the information presented in the specialized literature is focused on the degradation of dyes and their derivatives in the textile industry using these oxidative processes, without placing greater emphasis on food dyes, which are also part of the organic pollutants that cause alterations to the ecosystem, in general, when present in untreated effluents of these industries [43].

4. Advances in Heterogeneous Advanced Oxidation Processes

4.1. Catalysts in AOPs for Removal of Food Dyes

The use of catalysts for the degradation of organic and inorganic pollutants in effluents has increased in recent years. Catalysts have been widely studied for the degradation of dyes, so a wide range of them have been developed, such as metals, metal oxides, semiconductors, metal–organic frameworks (MOF), even quantum dots, dendrimer with magnetic core, among others [20]. Several strategies were developed for their syntheses, such as coprecipitation, flame hydrolysis, microwave radiation, impregnation, sol–gel method, hydrothermal and solvothermal techniques, chemical vapor deposition method, electrodeposition method, and electrospinning, among others [71,72].

Titanium dioxide (TiO_2) is one of the most widely applied semiconductor materials in the field of photocatalysis. As such, it is the catalyst of choice for decomposing dyes into mineralized products due to its abundance, chemical stability, good optical transparency, high refractive index, non-toxic nature, and low cost [20]. Khiabani et al. [73] analyzed the photodegradation process of Brilliant Blue FCF by adding TiO_2 under UV irradiation. It was shown that, the most important oxidative pathways for the degradation of this dye involved the loss of para-methylbenzenesulfonic acid (MBSA) groups and ethyl groups adjacent to amines. On the other hand, the effectiveness of TiO_2 can be improved when combined with other compounds, as performed by Rajamanickam et al. [74], who synthesized a $\text{SeO}_2/\text{TiO}_2$ composite photocatalyst to degrade the dye Sunset Yellow under solar irradiation in an aqueous solution. The results indicated that this combination was very effective, achieving a degradation of the dye in the order of 96.6%.

However, studies that are more recent show that TiO_2 has properties that are harmful to human health, leading researchers to reflect on the use of this in AOPs, and to search for new catalyst alternatives. In addition, it was shown that relatively high doses are required to achieve complete degradation—and even to achieve mineralization of the simplest compounds in a shorter time [75].

Other semiconductors, such as ZnO , CdS , WO_3 , and Fe_2O_3 , have been used in different studies, but to a lesser extent since they are heavy metals that can generate secondary contaminants [76]. Sudrajat et al. [77] developed and characterized an N-doped ZrO_2 pho-

photocatalyst (N-ZrO₂) by thermal decomposition of the zirconium hydroxide-urea complex. This photocatalyst presented better catalytic activity (with a decolorization of 67.2%) than pure ZrO₂ in degrading amaranth dye under visible or UV light irradiation at neutral pH, initial dye concentration of 10 mg L⁻¹, and catalyst concentration of 1 g L⁻¹. However, as an alternative, it was demonstrated that, with the use of UVA light at 3.5 W m⁻² at similar conditions, the complete elimination of the amaranth dye was achieved, without generating toxic derivatives.

Despite the advantages of applying semiconductor catalysts to increase the effectiveness of treatments, the routes to obtain them are complex. Moreover, they require UV irradiation to photo-decompose a dye due to their large band gaps, which increase the cost of the treatment [20,78]. For this reason, several studies have opted for doping with transition metals to extend the adsorption band to the visible region, as they exhibit excellent photocatalytic activity [79–81]. On the other hand, there is an emerging trend of the combination of photocatalytic processes with electrochemical advanced oxidation processes (EAOPs), to improve the degradation performance by the adequate formation of reactive species. For this purpose, photoactive electrodes (also called photoanodes or bifunctional anodes) are applied, which serve as electrodes as well as photocatalysts. However, technology based on electro-oxidation, in some specific cases, has the disadvantage of the possible formation of recalcitrant by-products.

In general terms, this process can cause poisoning effects due to the release of metal particles or microparticles and produce by-products that are more toxic than the primary pollutant, so it is necessary to evaluate the toxicity of the treated effluent, especially if the objective is not to achieve complete mineralization. Therefore, it is necessary to use new economical and environmentally-friendly methods, such as the so-called green catalysts [82–85], or nanocatalysts that improve the efficiency of advanced oxidative processes in the form of nanoparticles, nanotubes, nanospheres, and nanofibers [75,86,87].

4.1.1. Nanocatalysts

In recent decades, nanometer-sized materials (<100 nm) have been considered promising options, as they can adopt different shapes, sizes, structures, and improve the catalytic activity, selectivity, and stability of the system [14,88,89]. Unlike catalysts, nanocatalysts, due to their nanosize, have a larger surface area, allowing for better surface–volume interaction and, therefore, greater diffusion of reagents and products. Sometimes, to improve the efficiency of the process, these nanoparticles are dispersed on support, coupled, or doped with metals or adsorbents that have high porosity. The most commonly used support materials are zeolites, activated carbon, silicon film, and hybrid materials [15,90]. To ensure the stability of the nanoparticles, their synthesis must be carried out correctly since, when they bind ineffectively to the support, they are easily detached after the first use and washing [91].

The characteristics that make them potential tools for the removal of food dyes in wastewater include low sludge production and the possibility of reuse through regeneration methods integrated into the system [92].

Figure 2 shows several of the different known structures of nanocatalysts that can mediate the degradation of food dyes.

A wide variety of nanocatalysts exist for the degradation of pollutants in aqueous solutions from electrocatalysts, Fenton-based catalysts, and catalysts with antimicrobial characteristics [89]. For example, Thakur et al. [93] synthesized gelatin-Zr (IV) phosphate nanocomposites (GT/ZPNC) to degrade Fast Green dye, achieving a photocatalytic degradation performance of 89.91% over 5 h; even the antimicrobial activity of GT/ZPNC against *E. coli* was measured. Karkmaz et al. [94] synthesized cadmium vanadate nanostructures using the sol–gel auto-combustion process, achieving a 67% photocatalytic degradation of the dye erythrosine. On the other hand, Orooji et al. [95] obtained, for the first time, Gd₂ZnMnO₆/ZnO nanocomposites (GZMO/ZnO NC) by employing the sol–gel auto combustion technique using grape or coffee syrup as green fuel, obtaining a product with

a forbidden band of about 3.27 eV and efficient for dye removal. This study showed a superior photocatalytic activity towards the degradation of anionic dyes (erythrosine) by 73.8%.

Figure 2. Schematic representation of the main nanomaterials used as nanocatalysts in AOPs for the degradation of food dyes.

Each nano-photocatalyst exhibits particle sizes and shapes that affect its reaction performance and, at the same time, the morphology of nanoparticles depends on their chemical environment or presence of adsorbates [96]. Controlling morphology allows selective exposure of a larger portion of reactive facets to enrich and fine-tune catalytically active sites [97]. Nanocatalysts with complex morphologies, such as cubes, prisms, stars, or rods exhibit enhanced adsorption due to their multipolar resonances compared to the spherical nanostructures [98]. Bahadori et al. [99] obtained pure and Fe-doped methyl-imogolite nanotubes by ion-exchange catalyzing photo-Fenton reaction in the aqueous phase for the degradation of tartrazine. Tests showed that both nanotubes effectively photodegrade the food dye, even tartrazine mineralization is achieved with nanotubes containing 0.70 wt.% Fe. Darwish et al. [100] synthesized PVP-coated cadmium sulfide (CdS) nanoparticles using polyol with ethylene glycol, a facile method guaranteed to decrease photo corrosion. CdS exhibits better photocatalytic functions than TiO_2 by rapidly generating electron-hole pairs by photoexcitation. The researchers evaluated the efficiency of nanoparticles obtained by photocatalytic degradation of tartrazine under UVC irradiation and visible light, achieving complete removal of the dye after 90 min. Nazim et al. [101] synthesized porous CuO nanosheets using NaBH_4 as a reducing agent in an aqueous medium at room temperature. It achieved an optical energy band gap of 21.92 eV, resulting in excellent photocatalytic efficiency to degrade the food dye Allura Red AC by 96.99% in 6 min. Table 3 provides a summary of several studies performed for the degradation of food dyes from the use of different types of nanocatalysts in AOPs.

Table 3. Nanomaterials as catalysts in AOPS applied to degrade food dyes.

Process	Dye	C ₀	Catalyst	Dose C	Preparation Method	t	R	D	R.C	Ref.
Fenton photooxidation	Tartrazine	50	Cu-(BiOCl) with H ₂ O ₂	0.5	Coprecipitation	90	Visible	91	1	[87]
Photocatalytic	Tartrazine	10	8% TiO ₂ (PVDF-TrFE)	0.9	-	300	Solar	78	2	[91]
Photocatalytic	Tartrazine	50	MRG-TiO ₂ 50	0.2	Coprecipitation	180	Visible	95	4	[3]
Photocatalytic	Tartrazine	10	Cu- (mpg-C ₃ N ₄)	0.5	Pyrolysis	240	UV	81.9	-	[102]
Adsorption-photocatalysis	Tartrazine	10	Ze-nanZnO	0.1	Chemical precipitation	900	UV	87	-	[103]
Photocatalytic	Tartrazine	16	CuO	0.25	Chemical precipitation	240	Visible	90	-	[104]
Photo-Fenton	Tartrazine	40	MeIMO-Fe 0.025 with H ₂ O ₂	0.25	Ion exchange	240	UV	100	5	[99]
Photocatalytic	Tartrazine	25	ZnO doped Ni	0.1	Hydrothermal	60	UV	98.2	-	[105]
Photocatalytic	Tartrazine	10	C ₃ N ₄ /ZnO @ α -Fe ₂ O ₃	0.5	Direct pyrolysis and sol-gel	35	UV	99.34	3	[106]
Photocatalytic	Tartrazine	20	Benzoic acid/TiO ₂	0.1	-	70	Visible	99.08	4	[107]
Photocatalytic	Tartrazine	50	CuCr ₂ O ₄	0.5	Auto-combustion	120	Visible	99.6	4	[108]
Catalytic	Tartrazine	100	AgNP	60 ug	Green route	15	-	26	6	[109]
Photocatalytic	Tartrazine	25	semiconductors in Ag ₃ PO ₄	0.025	-	120	Visible	100	-	[110]
Catalytic	Tartrazine	50	Co(II) impregnated Al(III)-	0.1	Incipient wetness (CoAP)	10	-	100	-	[111]
Heterogeneous Fenton	Tartrazine	50	Fe ₂ O ₃ · SiO ₂	0.5	Easy impregnation	80	-	98.5	5	[112]
Catalytic	Tartrazine	25	Al ₂ O ₃ doped with cobalt	0.025	Electrolytic oxidation by plasma	180	-	100	5	[113]
Photocatalytic	Tartrazine	10	TPt-GO	0.2	Chemical-thermal	180	Solar	96.23	-	[67]
Photocatalyst	Tartrazine	50mg/L	NP TiO ₂	0.5	Sol-gel	220	UV	83	1	[114]
Photocatalyst	Tartrazine	10	Fe ₂ O ₃ (MF)	0.6		25	Visible	97.1	5	[115]
Photocatalytic	Erythrosine	5	DyVO ₄	0.05	Sonochemical	180	UV	88	5	[116]
Photocatalytic	Erythrosine	20	Graphene -NCs	0.02	Ecological sol-gel	50	UV	88.2	-	[70]
Photocatalytic	Erythrosine	1.2	Nd ₂ Sn ₂ O ₇ · Nd ₂ O ₃	0.05	Ecological synthesis	60	UV	96	8	[117]
Photocatalytic	Erythrosine	45	Cd ₂ V ₂ O ₇	0.05	Sol-gel	90	UV	67	3	[118]
Photocatalytic	Erythrosine	10	AuNPs @ ZnO	0.05	Chemical hydrothermal	30	UV	100	6	[119]
Photocatalytic	Erythrosine	20	NiS ₂	0.02	Hydrothermal	75	UV	57	-	[120]
Photocatalytic	Erythrosine	1.5	Dy ₂ Ce ₂ O ₇	0.06	Green Synthesis	70	Visible	91.4	8	[121]

Table 3. Cont.

Process	Dye	C ₀	Catalyst	Dose C	Preparation Method	t	R	D	R.C	Ref.
Photocatalytic	Erythrosine	8	NiFe ₂ O ₄ /SiO ₂ /Au (NiFe/Si/Au)	0.2	Sonochemical	80	UV and Visible	97.3	7	[122]
Sonocatalytic	Brilliant blue	396	WO ₃ -ZnO	0.2	Sonochemical	40	Ultrasonic	90	6	[123]
Catalytic	Brilliant blue	100	AgNP Viburnum opulus L.	60 ug	Green Synthesis	15	-	97	6	[109]
Photocatalytic	Brilliant blue	25	Chlorine	0.6 Mm	-	30	UV	95	-	[124]
Photocatalytic	Brilliant blue	25	semiconductors in Ag ₃ PO ₄	2.5	-	120	Visible	90	-	[110]
Photocatalytic	Amaranth	25	ZnO: Ag 1%	0.16	Electrospinning	600	Visible	98.4	-	[68]
Photocatalytic	Amaranth	25	ZnO: Ag 1%	0.16	Electrospinning	600	UV	95.9	-	[68]
Photocatalytic	Amaranth	30	superporous TiO ₂	0.3	Sol-gel	15	UV	95.6	5	[125]
Photocatalytic	Amaranth	25	ZnO decorated with Ag ₂ O	1	Ultrasound-assisted precipitation	30	Visible	94	-	[126]
Photocatalytic	Amaranth	12	TPt-GO	0.2	Chemical-thermal	180	Solar	99.56	-	[67]
Photocatalytic	Amaranth	50	MIL-N catalyzed oxone	0.1	-	30	Visible	100	-	[127]
Catalytic	Amaranth	50	(MOF) Fe, MIL-101	0.2	Schiff's base reaction	60	-	100	5	[128]
Catalytic	Amaranth	50	Bimetallic Co/Fe nanoparticles	0.05	Carbonization	20	-	100	5	[129]
Photocatalytic	Sunset Yellow	9	TPt-GO	0.2	Chemical-thermal	180	Solar	99.15	-	[67]
Photocatalytic	Sunset Yellow	10	P90/CN at 40%	0.1	Sonication	5	Solar simulated	98.8	-	[130]
Sonochemical oxidation	Sunset Yellow	50	peroxymonosulfate/CuFe ₂ O ₄	0.025	Coprecipitation	30	Ultrasonic	95.8	-	[131]
Photocatalytic	Sunset Yellow	10	Ailanthus excelsa Roxb	7	Green synthesis	30	UV	55	-	[132]
Photocatalytic	Sunset Yellow	40	Selenium NP-Drumstick	0.3	Green synthesis	180	Solar	83.8	-	[133]
photoelectron-Fenton	Sunset Yellow	100	NP de Fe ₃ O ₄	0.25	Coprecipitation	90	Solar	100	8	[134]
Photocatalytic	Fast green	10	Pp-16 @ Ag/AgCl(1:40)	0.1	In situ polymerization	20	UV	99	4	[135]
Photocatalytic	Fast green	30	OG/La ₂ O ₃ /ZrO ₂	0.05	Coprecipitation	90	Visible	89	5	[136]
Photocatalytic	Brilliant green	50	NiO/CuO	0.09	-	60	Solar	82	3	[137]
Photocatalytic	Malachite green	0.73	AlBc @La/Cu/Zr	0.1	Microwave	240	Solar	94	5	[138]
Photocatalytic	Indigo carmine	10	CdO nanowires	-	Mild chemical pathway	270	Visible	30	-	[139]
Photocatalytic	Indigo carmine	20	Pd-ZnS/rGO	1.0	Co-precipitation	210	UV	100	-	[140]

Table 3. Cont.

Process	Dye	C ₀	Catalyst	Dose C	Preparation Method	t	R	D	R.C	Ref.
Photocatalytic	Ponceau 4R	10	CdO nanowires	-	Mild chemical pathway	270	Visible	39	-	[139]
Photoelectrochemical	Ponceau 4R	15	ZnCo ₂ O ₄ /ZnO	0.125	Sol-gel	220	Solar	70	-	[141]
photo-Fenton	Ponceau 4R	50	Floating catalyst ZVI supported with LDPE	0.072	Thermal fixation	15	UV	100	4	[142]
Catalytic	Carmoisine	100	(AgNP)	60 ug	Green route	15	-	48	6	[109]
Electrochemical	Carmoisine	5	electro-oxidation-plasma electrode BBD	0.045 Mm	Electro-oxidation-plasma	60	-	100	-	[143]

C₀, initial dye concentration (ppm); Dose C, dose catalyst (g/L); t, time of maximum color degradation (min); R, radiation source; D, dye degradation (%); R.C, nanocatalyst regeneration cycles.

4.1.2. Metal Oxide-Based Nanocatalysts

Oxide-based nanocatalysts are inorganic types that are generally prepared with metals and non-metals, which are widely used for the removal of hazardous pollutants from wastewater. They are characterized by high BET surface area, lower solubility, minimal environmental impact, and absence of secondary pollutants [89]. Metal oxide nanocatalysts are classified into three generations: the first generation is composed of a single component, such as TiO_2 and ZnO ; the second generation is composed of multiple components in a suspension, e.g., $\text{WO}_3/\text{NiWO}_4$, $\text{BiOI}/\text{ZnTiO}_2$; and finally, the third generation is formed by photocatalysts immobilized on solid substrates ($\text{FTO}/\text{WO}_3\text{-ZnO}$, $\text{Steel}/\text{TiO}_2\text{-WO}_3$) [20]. There are three natural states of the nanocatalyst, anatase, rutile, and brookite, with anatase being the best nano-photocatalyst, and is usually effective in degrading anionic, cationic dyes, and dyes with different chromophore groups [20,144].

Naik et al. [125] developed super porous TiO_2 nanostructures using the sol-gel method to improve the surface area and robust structural porosity of TiO_2 anatase, and achieve higher photocatalytic performance without using support material. This research obtained excellent results, exceeding the efficiency of other TiO_2 catalysts, in 15 min, degrading 95.6% of Amaranth dye (30 ppm) using 0.3 g of the catalyst under UV irradiation. Moreover, this nanocatalyst also showed good stability for 5 cycles. Lamba et al. [126] synthesized and characterized heterostructures of Ag_2O -decorated ZnO nanowires using the ultrasound-assisted precipitation method. Through photocatalytic reaction processes (Figure 3), photogenerated electron/hole pairs actively participate in the chemical reactions for the generation of superoxide radicals and oxidizing agents, which are responsible for the degradation of the dye. Photocatalytic activity was measured by the degradation of Amaranth dye under visible light irradiation, exhibiting a photocatalytic yield of about 94% in 30 min. These heterostructures showed a synergistic effect, which was more efficient than pure ZnO nanowires and Ag_2O NPs separately, even presenting higher activity under visible light irradiation than commercially available photocatalysts, such as Merck ZnO , $\text{TiO}_2\text{-P25}$, and $\text{TiO}_2\text{-PC-50}$.

Figure 3. The plausible photocatalytic mechanism for photocatalytic degradation of amaranth dye in the presence of Ag_2O nanoparticles decorated ZnO nanorod heterostructures under visible light irradiation. [126].

Aoudjit et al. [91], on the other hand, evaluated the feasibility of using PVDF-TrFE photocatalytic membranes with immobilized TiO_2 nanoparticles as a catalyst to degrade tartrazine in a solar photoreactor. A high removal performance of the pollutant (78%) was obtained, but a second use under the same experimental conditions showed a reduction of approximately 10% in degradation efficiency. The causes of loss of efficiency are mainly because not all of the nanoparticles are effectively bound to the polymeric matrix and tend

to detach from the first use and subsequent washing and/or to a large amount of dye retained on the membrane surface, and the consequent decrease of specific photocatalytic active sites, both reasons becoming the main limitations of immobilized systems.

Alcantara-Cobos et al. [103] prepared zeolite–ZnO nanoparticles (Ze–nanZnO) for a coupled adsorption–photocatalysis process. This coupling obtained lower degradation percentages than pure ZnO nanoparticles; however, although the degradation of nanZnO is higher, the separation process of nanZnO is more complex, making the process more expensive. Finally, Türkyılmaz et al. [105] synthesized and analyzed ZnO nanostructures doped with Ni, Mn, Fe, and Ag, to degrade tartrazine. A maximum degradation of 98.2% was achieved with Ni/ZnO in 60 min. However, applying Mn and Fe did not achieve good results, which may be due to the coexistence of multivalent ions in the ZnO host and their highly agglomerated structures on the ZnO surface.

4.1.3. Graphene-Based Nanocatalysts

Graphene-based nano-photocatalysts have covered a wide field for the photodegradation of dyes in water in two phases. The first one consists of adsorption of the dye on the catalyst surface and the second phase corresponds to the ring scission of the dye ring [145]. Graphene-based nano-photocatalysts exhibit high physicochemical stability, large surface area to volume ratio, high mechanical strength, low optical absorbance and density, good thermal conductivity, and excellent electrical conductivity. [145,146]. However, it is advisable to perform the removal of organic contaminants from the aqueous medium through adsorption separations on graphene-based magnetic nanocomposites [145] and optimize the photothermal performance of graphene-based nanocomposites for enhanced photocatalytic activity [147].

On the other hand, combining graphene with metal oxide semiconductors and other particles favors stability and photocatalytic activity. For this, graphene acts as a photostabilizer of the semiconductors and as an electron sink by greatly reducing the recombination rate of the electron–hole pair, resulting in higher conductivity, low cost, higher absorptivity, tunable optical behavior, longevity, and biocompatibility [148].

A good representation of the series of reactions involved in the use of graphene is represented in Figure 4, in which $\text{OH}^\bullet$ radicals are produced by holes, and photogenerated electrons produce $\text{O}_2^{\bullet-}$, which play an important role in the degradation of the dye.

Figure 4. Mechanism of photogenerated charges in reduced graphene oxide.

Under the mechanism depicted in Figure 4, Nada et al. [3] proposed the degradation of tartrazine by using nano-titania-magnetic reduced graphene oxide (MRGT). The synthesis of nanocomposites was performed by loading reduced graphene oxide (RG) with two nanoparticle components consisting of TiO_2 and magnetite (Fe_3O_4) in varying amounts, achieving a dye degradation of more than 95%.

On the other hand, Agorku et al. [140] achieved Pd-decorated zinc sulfide/reduced graphene oxide nanocomposites (Pd-ZnS/rGO) by the co-precipitation method under UV irradiation. A 100% photocatalytic degradation of indigo Carmine was achieved by employing a 1.0% Pd amount in ZnS, evidencing a good photocatalytic performance due to the synergistic effect of Pd with rGO, small crystal size (crystallite), light absorption intensity, and narrow forbidden band energy (2.0 eV). On the other hand, Rosu et al. [67] developed TiO_2 -Pt (TPt), TiO_2 -Pt/graphene oxide (TPt-GO), and TiO_2 -Pt/reduced graphene oxide (TPt-rGO) NPs by a combined chemical–thermal method to photodegrade different azo dye structures (amaranth, sunset yellow, and tartrazine) in the presence of UV irradiation and natural sunlight. The efficiency of the photocatalytic activities of the synthesized compounds was demonstrated, especially that of TPt-GO with higher photoactivity under sunlight, removing 99.56% of amaranth, 99.15% of sunset yellow, and 96.23% of tartrazine.

4.1.4. Metal–Organic Frameworks Synthesized with Nanocomposites

Metal–organic frameworks (MOFs) are defined as compounds formed by the interaction of metal ions or metal clusters (secondary building units, SBUs) and organic ligands ideal (usually carboxylic acid or nitrogen-containing ligands) for the fabrication of high-performance multifunctional compounds. They are characterized by very good surface areas and high porosity (porous crystalline materials), as well as by dispersed active sites and functionalities within this porosity.

Their ability to act as chemical nanoreactors allows them to synthesize and stabilize, within their channels, a catalytically active species that would otherwise be difficult to access [149–151].

In photodegradation processes, there is a difference between the energies of the light and the band gap. In this case, the mechanism followed by MOFs is that, when the energy promoted by the light source is higher than the energy of the forbidden band, the electrons in the valence band gain energy and are excited to the conduction band, generating a positive hole in the valence band, to then generate reduction and oxidation half-reactions with electrons and holes, respectively. Figure 5 shows the photocatalytic degradation mechanism, where MOFs behaving as semiconductors could be considered potential photocatalysts [152].

Figure 5. Proposed mechanism for the photocatalytic degradation of organic pollutants by $\text{Co}_3\text{O}_4/\text{TiO}_2/\text{GO}$ nanocomposite [153].

In heterogeneous Fenton reactions, iron-containing metal–organic frameworks (Fe-MOF) have been heavily used because of their high capacity to generate hydroxyl radicals $\bullet\text{OH}$ and lower photocatalytic activity resulting from the high recombination of electron–hole pairs generated [151].

The Materials Institute Lavoisier (MIL) group of metal–organic structures is presented as an efficient alternative in the field of environmental remediation [151]. For example, iron-

based MOF materials, such as MIL-88A, MIL-53, and MIL-68, are presented as potential photocatalytic tools for dye degradation [154]. Zhang et al. [128] developed for the first time an Fe nanoparticle-containing MOF, MIL-101 as a support to immobilize ferrocene (Fc) chemically. It was employed to degrade amaranth by oxone activated by Fc-MIL. The Fc-MIL presented good catalytic activity and lower activation energy, an advantage that makes it an excellent catalyst to eliminate the toxic amaranth. Since few studies have been conducted for the employment of Fe-based catalysts to activate Oxone in amaranth degradation, Zhang et al. [127] presented a study of Fe nanoparticle-based MOF using oxone catalyzed by MIL-101-NH₂, which achieves complete degradation of amaranth dye in 30 min under visible light irradiation. The catalyst dosage was 100 mg, to degrade 50 ppm of amaranth dye. This catalyst could eliminate amaranth dye even in the presence of surfactants. In addition, since it is a heterogeneous catalyst, reusability was tested, showing that it can be used for multiple cycles within 60 min of degradation.

Meanwhile, Zhang et al. [155] synthesized a catalyst consisting of TiO₂/mag-MIL-101 (Cr) nanoparticles under visible light. The magnetic property of this TiO₂-loaded composite enhanced its adsorption and photocatalytic efficiency with a forbidden band value of 1.61 eV. In addition to being easy to recover, it increases its reusability [129]. In this study, it was used to adsorb and degrade refractory organic compounds, bisphenol-F (BPF) and acid red 1 (AR1), from water, resulting in efficient removal of both compounds. Thus, it is presented as a new tool of semiconductor-MOF nanocatalysts to degrade organic pollutant compounds in water under the use of visible light. However, the microporosity of the MOFs hinders the complete adsorption of larger dye molecules, since the passage to the internal pores or voids of the MOFs is impeded. In addition, there would be a selectivity of charge and/or size, since the MOF-based nanocatalysts would be designed for a specific type of dye, characteristics that are considered unfavorable for the treatment of wastewater by this method containing complex mixtures of dyes [156].

4.1.5. Nanomaterials Synthesized by Green Technologies

Although the above processes are considered efficient systems, not all of them are economical and environmentally friendly, because nanomaterials synthesized by conventional routes can involve hazardous and volatile chemicals that even lead to the formation of secondary pollutants [152]. Therefore, it becomes indispensable to find new and efficient methods, with high catalytic activity, are easy to handle, and eco-friendly [14]. In this context, in eras of sustainable development and the application of green chemistry concepts, the most recent alternative studies are nanomaterials synthesized by green technologies or biogenic nanoparticles (BNPs), which are presented as a new tool for photocatalytic water remediation (Figure 6). These BNPs are characterized by possessing a large specific surface area, ion exchange mechanism, unique morphology, high biocatalytic reactivity, efficient regeneration, and colloidal properties, so that they can be separated from effluents by gravity filtration, sedimentation, or the coagulation–flocculation technique [157].

Studies demonstrate the efficiency of these processes for the removal of various food contaminants. Deepika et al. [132] identified the bioactive compounds of *Ailanthus excelsa* Roxb bark and leaf extract for the removal of synthetic food dyes. The researchers observed that *A. excelsa* bark extract exhibits better photocatalytic activity by degrading 55% of sunset yellow dye FCF. Likewise, Hassanien et al. [133] synthesized selenium BNPs from *Drumstick* leaf extracts to evaluate their photocatalytic activity towards the decolorization of sunset yellow dye FCF under solar and UV irradiation, achieving a dye degradation rate of 83.8 and 76.6%, respectively. David and Moldovan [109] proposed the synthesis of silver BNPs (AgNP) by using *Viburnum opulus* L. fruit extract, which acts as a reducing agent of silver ions and stabilizer of the obtained nanoparticles. The size, morphology, crystalline nature, catalytic ability, and stability of AgNPs were evaluated to remove toxic food dyes, such as tartrazine, carmoisine, and Brilliant Blue FCF in the presence of NaBH₄. These spherical-shaped nanoparticles, with an average diameter of 16 nm, achieved 48,

26, and 97% degradation of carmoisine, tartrazine, and Brilliant Blue FCF, respectively, in only 15 min.

Figure 6. (a) General representation of the green synthesis of BNPs and (b) reaction mechanism of AgNPs for the degradation of dyes [158].

Although studies on the use of BNPs have increased in recent years (Figure 7a), they are not sufficient in terms of the total number of articles per year. In addition, there is a large difference between the production of BNPs and conventional nanocatalysts. This opens up new lines of research in which the properties of BNPs are not only used in the field of environmental sciences, but also other less-studied areas (Figure 7b).

Figure 7. Number of publications on the use of BNP compared to classical nanocatalysts per year (a), BNPs application area (b) (source: Scopus database with searching keywords: “green AND nanocatalysts AND dyes,” July 2021).

5. Discussion

The degradation of food dyes through photocatalytic AOPs in wastewater treatment processes has become a promising alternative to conventional methods. This is due to the high degradation yields that this type of technology can achieve. There are several reports in the literature on the oxidation of these compounds using various catalysts and nanocatalysts, based on metal oxides, graphene, MOFs, and biogenic nanoparticles. However, all of these materials—to achieve maximum operating performance—depend on various parameters, such as the amount and type of catalyst and oxidizing agent, reaction temperature, preparation method, surface area, chemical composition, particle size, morphology, etc. For this reason, semiconductor-mediated photocatalysis of metal oxides (TiO_2 , ZnO , WO_3 , CdS) presents several limitations, including the difficulty to immobilize powder nanoparticles on substrates acting as support, rapid recombination of photoinduced electron-hole pairs, low activity in the visible region, toxicity to biological systems, and difficulty in enhancing photocatalytic activity (e.g., in degradation processes the efficiency of TiO_2 is less than 10% due to its limited sensitivity to ultraviolet light). For this purpose, evaluations are available that enhance the characteristics of semiconductors through non-metallic doping (N, F, C, S), deposition of noble and transition metals (Pt, Pd, Ag, Au, Ru, Fe, Ni), or even coupling with other semiconductors that allow the development of nanocomposites with improvements in their porous structure, surface area, and photocatalytic activity. In addition, the properties of these metals have allowed approaching the coupling of other AOPs, such as photo-assisted electrochemical methods (e.g., photoelectrocatalysis and photoelectro-Fenton), which improve the generation and action of the reactive agents involved in the degradation. However, degradation is much slower, which strongly increases the energy consumption required for total mineralization.

On the other hand, metal oxides can be enhanced in performance when combined with MOFs, increasing degradation performance. However, most research on MOFs has focused on their efficiency as adsorbents for organic pollutants in wastewater; therefore, the number of food dye degradation evaluations using MOFs as nanophotocatalysts is limited. Engineered metal–organic frameworks based on heterogeneous Fe-containing materials (MIL-N) are considered as promising methods to completely degrade food dyes over several reaction cycles. Moreover, the construction of bimetallic or polymetallic MOFs improves the catalytic performance of monometallic MOFs. However, for large scale applications, these structures present drawbacks in their inherent properties and their production, for which several important aspects must be considered, such as availability and cost of materials, treatment procedures, synthesis, activation, yields, decrease in the amount of solvent use, as well as the stability that can be affected by their geometry, metal-ligand bonds, etc.

Another nanomaterial studied is graphene and its derivatives, which show better results when working together with other compounds. Being used as a semiconductor in its pure form, it has disadvantages due to its hydrophobic nature. Graphene oxide (GO) exhibits a higher electrical insulating character due to the presence of functional groups, and it is necessary to reduce the oxygen content by increasing the carbon content in a C:O ratio from 2:1 to 246:1. The compound obtained is called reduced graphene oxide (rGO), and allows the interaction of π -bonds of organic molecules with the π -systems of rGO, a property that facilitates the efficient removal of contaminants from water. To reduce electron-hole recombination, graphene oxide is shown as a compound capable of acting as an electron acceptor when combined with metal oxide semiconductors and transition metals (electron traps). One advantage that graphene oxide possesses is the ability to combine oxygen functional groups on its surface with the metal centers of a MOF, so that mass and electron transfer is greatly favored. Although graphene-based nanocomposites can be considered as materials with promising characteristics, the intermediates that may be generated tend to be more toxic than the original organic compounds. Therefore, toxicity testing of remediated waters from AOPs that include graphene-based composites is considered essential.

Although nanocatalysts are essential for the degradation of pollutants through AOP, one issue of environmental interest has arisen regarding the separation, recovery, and reuse of these nanocomposites once the treatment is complete. Therefore, these materials are generally synthesized with magnetic properties that facilitate the separation and recovery of the material from the aqueous medium, quickly and efficiently. The high magnetic permeability of the particles or compounds with structures formed with Fe allows them to be eliminated and recycled through the application of a suitable magnetic field without causing major effects on the environment. In general terms, before final disposal, it must be ensured that they comply with environmental legislation based on leachability [159]. For this reason, there is currently a trend in the formation of MOFs and their derivatives, since they do not show apparent changes before and after the catalytic reaction, which indicates excellent stability. In addition, the leaching concentration of Fe ions after the reaction is generally in the range of 0.03–2 mg L^{−1} for MOF catalysts based on Fe NPs, which is below the EU environmental standard (2 mg L^{−1}). On the other hand, modified MOF-based catalysts can reduce the leaching of metal ions by protecting the carbon matrix and core-layer structure [160].

Despite all the advantages manifested by metal oxide, graphene, and MOF NP-based nanocatalysts, they have two limitations in common: (i) the need to couple to other compounds to present improvements in their structures and photocatalytic properties, along with high costs and toxicity synthesis methods, mostly operating at high temperatures and pressures. (ii) The generation of constant secondary contamination, by either the detachment of nanoparticles from the system in function (which end up being incorporated into the aqueous medium) and/or the production of compounds more toxic than the primary contaminant material.

Finally, the scientific community is showing interest in the emergence of the synthesis of nanostructures from biological media, since they are energy efficient, low in cost, easy to scale-up, and do not cause additional damage to the environment, since renewable reagents are used instead of chemical compounds (which act as toxic reduction and dispersing agents). Additionally, they have long reuse cycles without loss of degradation efficiency, and they can be used as stabilizing agents for nanoparticles.

Fungi are favorable options to meet these requirements and, in turn, are the best candidates to synthesize nanocomposites at an industrial level, as they are easily manipulated, secrete large amounts of enzymes necessary for reduction, and present high filamentary tolerance to metals. The use of green synthesis methods may be a way to minimize the risks of secondary contamination.

6. Conclusions and Prospects

The present review demonstrates that AOPs have great potential in the degradation treatment of food dyes since AOP chemistry offers several mechanisms for the treatment of this type of organic pollutant. Recent articles demonstrate the growing interest in the application of AOPs for the treatment of these pollutants, for the protection of the environment and human health. Traditionally, most methods used are based on the application of photocatalytic AOPs with micro-sized photocatalysts. However, the current trend is based on the application of nanotechnology for wastewater treatment. The application of nanomaterials has become an opportunity to revolutionize wastewater treatment concepts, favored by their unique properties, which have created a convergence with AOPs. The use of nanomaterials in AOPs has been advantageous because they act catalytically and provide a higher surface area, which significantly increases degradation efficiency. The nanomaterials explored in this review are divided into transition metal, carbon-based, molecular organic (MOF), and are synthesized from green sources (phyto-nanocatalysts). This last classification is the least explored, which emphasizes intensifying this line of materials for their photocatalytic properties and the possibility of fusing with transition metals.

However, despite the excellent properties possessed by these materials, future research should also focus on:

- (i) Reducing the evaluation of the degradation of food dyes from synthetic waters, with more attention on the analysis of real water systems, since several factors that are not considered notably affect the ecosystem or the application of the nanocatalyst in real remediation systems;
- (ii) Measuring the toxicity mechanisms of the intermediates generated by the use of nanomaterials for the removal of pollutants in wastewater, and to couple such procedures to advanced analytical chromatography techniques to control the various reaction steps given during the treatment; and
- (iii) Controlling and evaluating the type of surface coating, particle size, and residues also generated by the nanomaterials in their different reaction cycles, since even low concentrations of nanoparticles could alter natural processes or cycles and, consequently, affect the microbial community and the ecosystem in general.

In general, it was demonstrated that, whatever the type of nanocatalyst applied, the degradation potential is highly efficient. Studies have been extensively carried out at laboratory or pilot scales. The high costs of production and large-scale availability have become obstacles to large-scale applications. Therefore, the ultimate objective is to develop efficient mechanisms to obtain highly efficient, health-safe, and biodegradable nanomaterials to apply in everyday (and industrial) life.

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