

Review

Revolutionizing Wastewater Treatment: Harnessing Metal–Organic Frameworks for Exceptional Photocatalytic Degradation of Azo-Type Dyes

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Abstract: Due to the high stability of azo-type dyes, conventional treatment processes such as adsorption, flocculation, and activated sludge are not efficient for decolorizing wastewater effluents. An alternative to traditional wastewater treatment is photocatalysis, which has gained significant interest because research has shown it to be a viable and cost-effective process that uses sunlight as an inexhaustible energy source. In heterogeneous photocatalysis, a photocatalyst is required, such as TiO₂, ZnO, composite materials, and, more recently, metal–organic frameworks (MOFs). MOFs, also known as “coordination polymers”, exhibit photocatalytic properties and have been proven to be promising materials in the photocatalytic degradation of dyes. This study presents recent advances in using MOFs as photocatalysts to degrade recalcitrant contaminants like azo-type dyes. Recent advancements in developing photocatalysts based on MOFs are focused on two strategies. Firstly, the development of new MOFs composed of complex ligands or a mixed ligand system, and secondly, the synthesis of composite materials based on MOFs and metal oxides, metals, sulfides, nitrides, etc. Both strategies have significantly contributed to the search for new semiconductors to degrade some recalcitrant contaminants in wastewater.

Keywords: MOFs; composite-based MOFs; colorants; dyes; basic dyes; acid dyes; reactive dyes; photocatalytic degradation of dyes



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1. Introduction

1.1. Colorants

Color has a long history with humans and is valuable and essential in our daily lives. Colorants are classified into dyes and pigments and are crucial to adding color to various materials (Figure 1) [1]. The key distinction between both is that dyes are soluble in the substrate, dispersing at a molecular level, while pigments are insoluble and are applied as particles [2]. There are two main types of dyes: synthetic and natural [3]. Synthetic dyes are derived from petroleum compounds, while natural dyes are obtained from plants, animals, and minerals [4]. Synthetic dyes consist of organic molecules with three essential components: the chromophore, the auxochrome, and the matrix [2,3]. The chromophore is the part of the molecule responsible for absorbing specific wavelengths of light and giving the colorant its color. The common chromophores include the azo (–N=N–) and non-azo (nitro (–NO₂), nitroso (–N=O), carbonyl (–C=O), and thiocarbonyl (–C=S)) group. On the

other hand, an auxochrome is a group of atoms or ions that can alter or enhance the color of compounds when attached to a chromophore group. They are often responsible for the different colors we observe in organic compounds, such as dyes. Common auxochromes include the hydroxyl (OH) group, amino (NH_2), aldehyde (CHO), and methyl mercaptan (SCH_3). Finally, the remaining atoms in the molecule make up the matrix, the dye's third component. The matrix includes the solubilizing groups, stabilizing groups, and counterions. Azo dyes are mainly used by textile, paper, plastics, and food industries and are thus estimated to account for approximately 70% of the world's dye production [5,6].

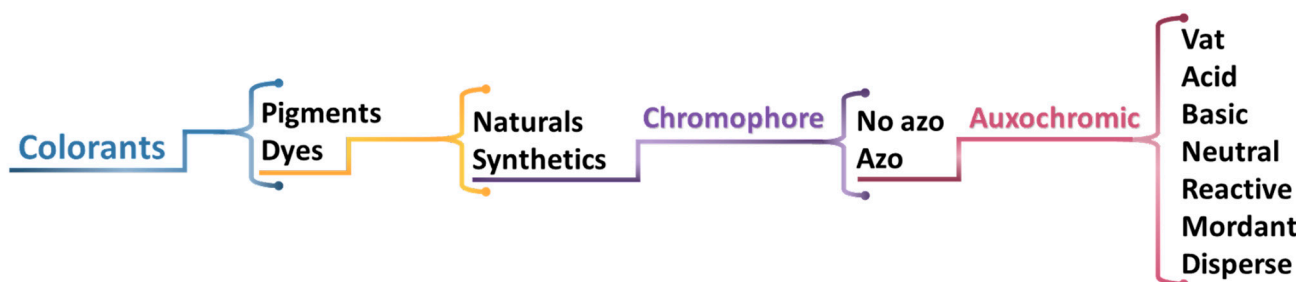


Figure 1. Structures of organic dyes that will be studied in this work.

Azo-type dyes find various industrial applications and can be categorized into several classes, including acid, basic, neutral, reactive, vat, mordant, and disperse, as shown in Figure 1 [4]. Table 1 shows the chemical structure of any azo-type organic dyes according to their classification. Acid dyes are anionic compounds that are generally water-soluble. They are used for dyeing protein fibers under acidic conditions [7]. A distinctive characteristic of acid dyes for protein and polyamide fibers is the presence of one or more sulfonate ($-\text{SO}_3^-$) groups, usually in sodium salt ($-\text{Na}^+$). These groups serve a dual purpose: ensuring solubility in water and imparting a negative charge to the dyes.

On the other hand, basic dyes are cationic compounds carrying a positive charge, typically originating from the ammonium cation. They adhere well to negatively charged fibers and are employed to dye materials like acrylic, nylon, and other synthetic fibers. Basic dyes also have applications in paper and ink dyeing [8]. Direct or substantive dyes constitute a significant proportion of anionic azo dyes and are used to dye natural or regenerated cellulose [9]. The essential requirement for the classification of a dye in this group is its substantivity, i.e., its absorption from an aqueous salt-containing solution onto cellulosic materials.

Table 1. Structure of any azo-type organic dyes according to their classification.

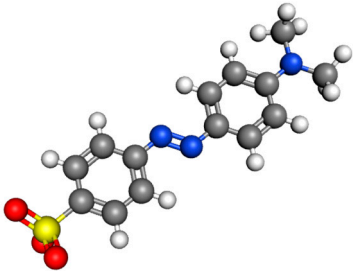
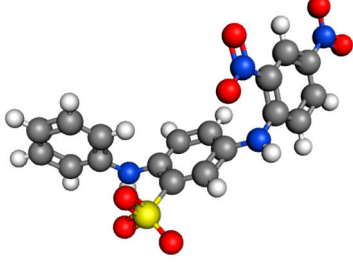
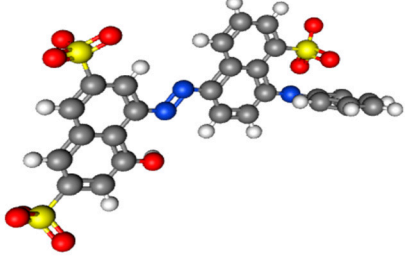
Acid Dye		
MO, Methyl Orange, C.I. Acid Orange 52	Acid Orange, C.I. Acid Orange 7	AB92, Acid Blue, C.I. Acid Blue 92
		

Table 1. Cont.

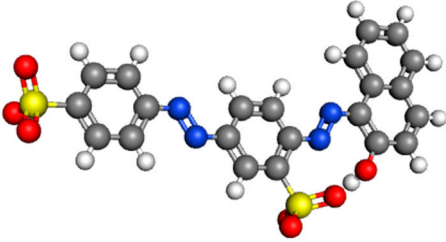
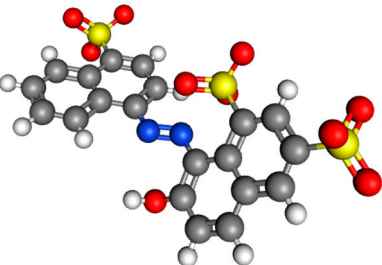
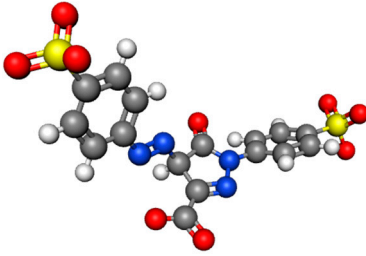
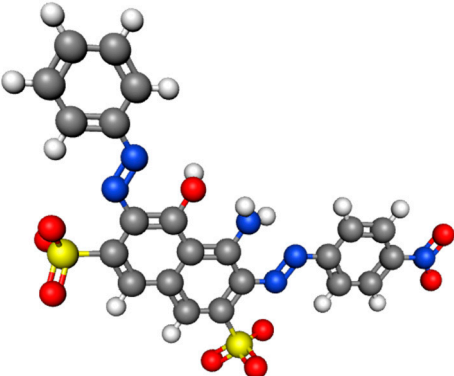
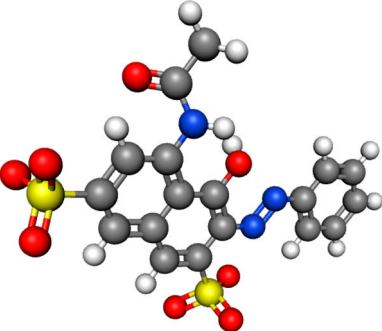
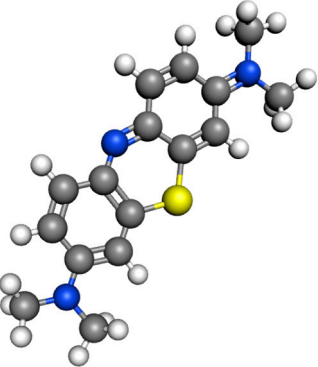
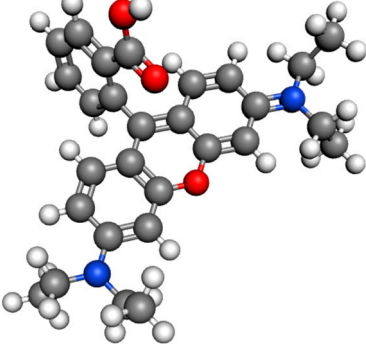
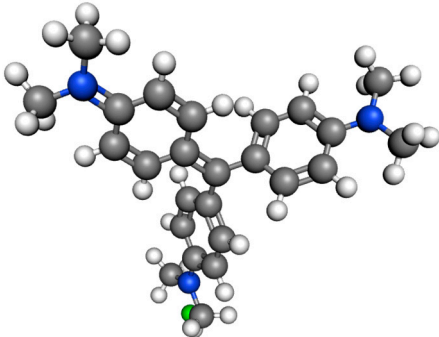
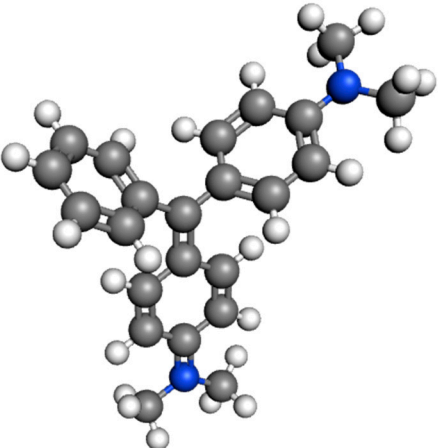
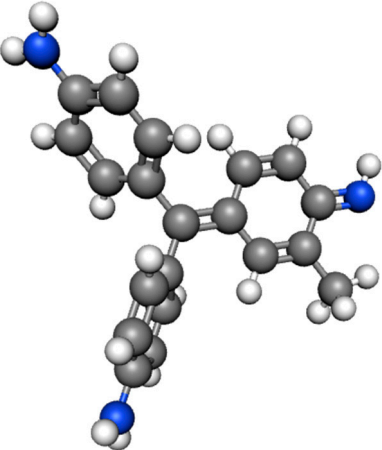
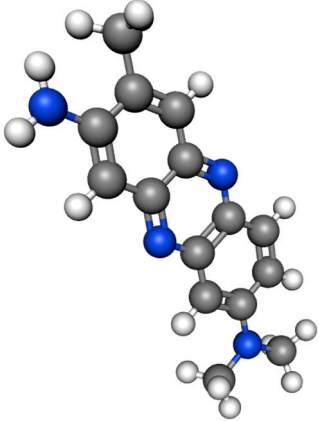
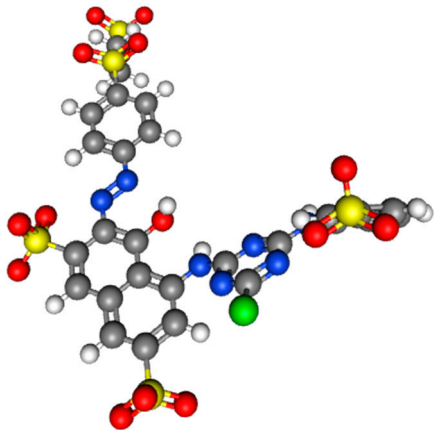
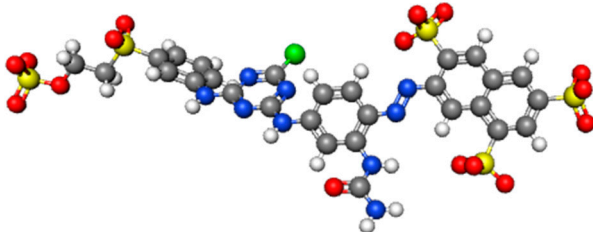
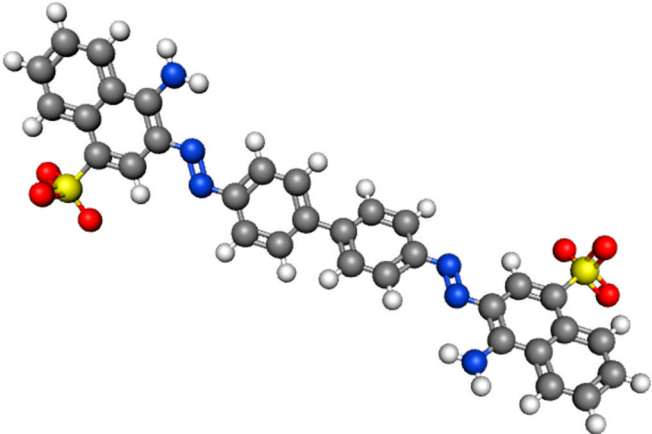
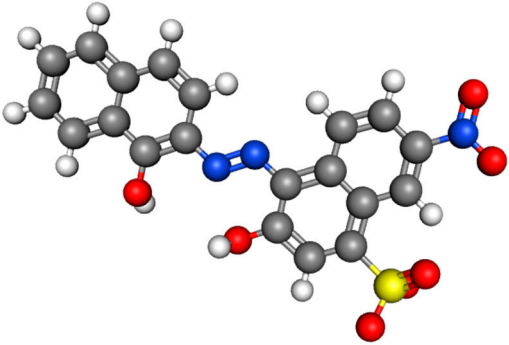
Ponceau BS, C.I. Acid Red 66	New Coccine, NC, C.I. Acid Red 18 = 1	Tartrazine, C.I. Acid Yellow 23
		
Acid Black, C.I. Acid Black 1	Acid Red 1, C.I. Acid Red 1	
		
Basic dye		
MB, Methylene Blue, C.I. Basic Blue 9	Rhodamine B, RhB, C.I. Basic Violet 10	Methyl violet, MV, CV, C.I. Basic Violet 3
		
Malachite Green (MG), Green Basic, C.I. Basic Green 4	Basic Violet (BV14), C.I. Basic Violet 14	Neutral Red, NR, C.I. Basic Red 5
		

Table 1. Cont.

Reactive dye	
Reactive Red (RR)198, C.I. Reactive Red 198	Reactive Yellow (RY), C.I. Reactive Yellow 145
	
Direct dye	Mordant dye
Congo Red, CR, C.I. Direct Red 28	Eriochrome Black T, C.I. Mordant Black
	

Blue = Nitrogen, Red = Oxygen, Yellow = Sulfur, White = Hydrogen, Gray = Carbon, Green = Chloro.

1.2. Main Degradation Methods of Dyes in Wastewater

The advent of the Industrial Revolution marked a profound transformation in human life as machines were harnessed for mass production, creating a substantial demand for various dyes to color large quantities of fabrics. During this era, the available dyes were primarily of plant and animal origin, which proved impractical in meeting the growing demand. Consequently, humans began synthesizing various dyes without considering these compounds' potential degradation and toxicity. As time passed, these compounds were recklessly disposed of into water bodies and sewers, leading to severe environmental issues [10].

Since most azo-type dyes are of anthropogenic origin, their natural degradation is minimal, if not virtually non-existent. As a result, they persist in the environment for extended periods [11]. For this reason, in recent years, significant efforts have been made to improve conventional treatments for the degradation of azo-type dyes, such as adsorption, oxidation methods, biological methods, and photocatalysis [12]. All these dye treatment methods are depicted in Figure 2.

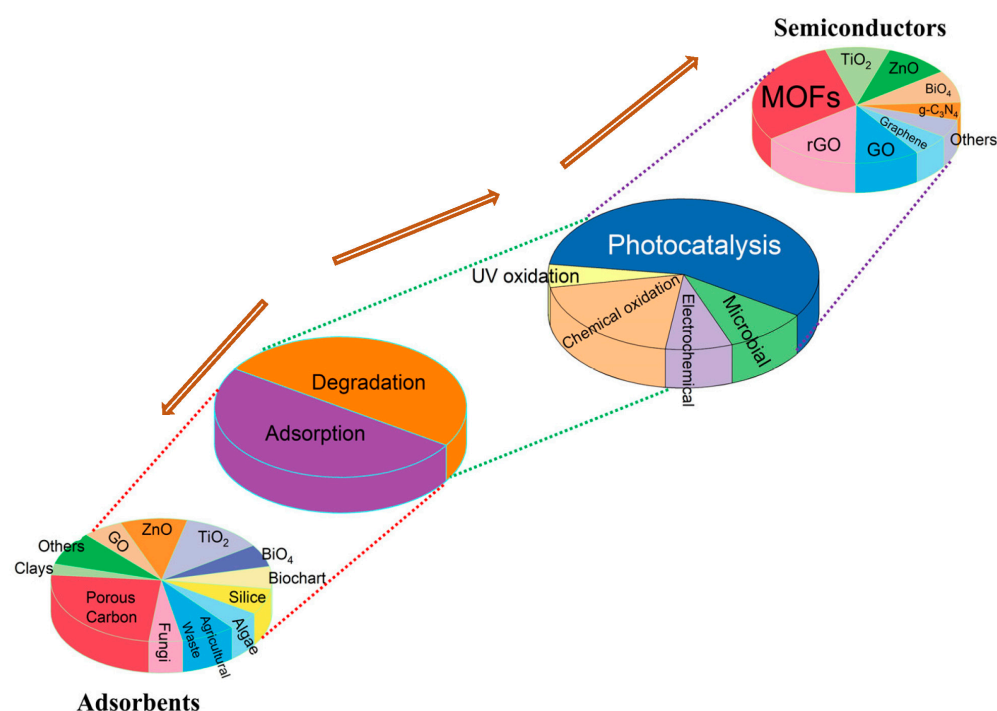


Figure 2. Main treatments of dyes.

Table 2 shows the advantages and disadvantages of any of the convectional and advanced treatments used for the degradation of azo-type dyes in wastewater.

Table 2. Advantages and disadvantages of any of the methods used for the elimination/degradation of azo-type dyes in wastewater [13–18].

Treatment	Advantage	Disadvantage
Adsorption	-Able to adsorb all the concentration of the dye. -One step to recover the absorbent with the dye.	The dye only changes the medium; the environmental problem persisted throughout the time using this treatment.
Chemical oxidation	-Oxidation can be achieved in most of the dyes. -One or more oxidant substances can be used to oxidize the dye.	-In many cases, the sub-products are more toxic than the molecule to be degraded -This treatment can be expensive because it uses reactive in huge quantities, and they will have a high concentration.
Electrochemical	-The use of electric power can break down the dyes. -This method can treat vast quantities of effluents with dyes.	After applying this treatment, the formation of a massive amount of sludge that has the sub-products of the degradation of the dye
Biological	-Use more than one kind of bacteria. -The system does not need the addition of more reactive because the dye can be used as food for the bacteria.	-This methodology can apply only to a few quantities of dyes. -The biodegradation process is slow.
UV oxidation	-Using UV-C light to break the links of dye molecules. -The treatment can be applied for a short time.	-The high energy of the light can produce a health problem for the people around it. -High consumption of electricity.
Photocatalysis	-Require only a semiconductor and light. -This methodology can eliminate a considerable volume with a low concentration. -The system can be activated using solar radiation. -Generally, the system does not generate more toxic compounds. -The method can be achieved in many cases via the mineralization process of the dye.	-Some semiconductors must be modified to achieve a high percentage of dye degradation. -In some cases, the separation process of the semiconductor from the water.

One initial method for treating azo dyes was adsorption onto a substrate [19–23]. This approach was also necessitated by early water treatment stages involving sand, gravel, or clay filters that partially adsorbed these dyes [24,25]. However, this initial option was specialized, prompting the search for more efficient treatment alternatives since these filters needed frequent replacement, incurring higher costs (Table 2). Thus, pre-treatment stages were developed where adsorbents with high adsorption capacities, such as activated carbon, were utilized [26]. Activated carbon cannot only be used as an adsorbent in the removal of dyes [8,27,28]; it has other applications such as air and gas purification, gold recovery, chemical catalyst support, solvent recovery, gas masks and protective gear, cosmetics and beauty products, and other [29–32]. However, it is important to mention that the adsorption method often involves transferring dyes from one medium to another, i.e., from water to a solid substrate (Table 2). Hence, the appropriate treatment method must be sought to prevent future contamination [33]. For example, chemical oxidation treatments aim to oxidize dyes using reactive agents, but in some cases, these dyes contain halogens that produce even more toxic byproducts, exacerbating environmental issues (Table 2) [34].

Other methods for the degradation of azo dyes have been explored via electrochemistry, which can effectively eliminate dyes but often generate sludge requiring further treatment [35]. The biological treatment has gained traction as bacteria have evolved to tolerate more toxic environments and break down complex dye molecules as a food source, facilitating partial or complete azo dye degradation [36,37]. However, this approach has not been effective for all dyes. Another option involves using UV-C light for dye oxidation; its high energy can break chemical bonds, resulting in partial dye degradation [38]. However, a high consumption of electricity is necessary. Combining multiple degradation techniques has been essential in many instances to break down the dyes fully.

Recent research has delved into a novel treatment method using a semiconductor, a light source, and a dye, where these components interact to achieve complete dye mineralization; this process is called photocatalysis [18]. The concept of using semiconductor materials such as TiO_2 in photocatalytic treatments emerged in the 1990s. Researchers discovered that titanium dioxide could generate electrons (e^-) and holes (h^+) when exposed to UV light. These two photo-species enter the surrounding medium, where the electrons can interact with dissolved oxygen in water, forming superoxide ($\cdot\text{O}_2$). Simultaneously, the photogenerated holes (h^+) interact with water molecules, producing hydroxyl radicals ($\cdot\text{OH}$) capable of oxidizing organic matter [39].

Over the years, this application has evolved by using solar radiation as an environmentally friendly light source. Additionally, semiconductor materials have improved via enhanced synthesis methods, modifications, or doping [40–43]. In some cases, entirely new semiconductors have been developed, expanding the possibilities for treating dye-contaminated effluents.

1.3. Fundamentals of Metal–Organic Frameworks (MOFs) as Semiconductors

Metal–organic frameworks (MOFs) are three-dimensional crystalline materials characterized by metallic clusters connected by organic ligands [44]. Beyond separation and gas storage applications, MOFs have found use in various fields, including catalysis (e.g., photo- CO_2 reduction, and photocatalytic dye degradation), drug delivery and transport (enabled by mesopores for substance storage), optical sensors due to their semiconductor properties or as photocatalyst, and more. Currently, MOFs are of great interest because some have photocatalytic properties such as NH_2 -MIL-125 (Ti) [45–49], UIO-66 (Zr) [50], NH_2 -UIO-66 (Zr) [51–53], MIL-53 (Al) [54,55], NH_2 -MIL-101 (Fe) [47,56], and new MOFs with a complex ligand or mixed ligand [57–59], among others. Figure 3 shows how research into MOFs as photocatalysts has been growing in recent years, as well as their use as photocatalysts for the degradation of dyes.

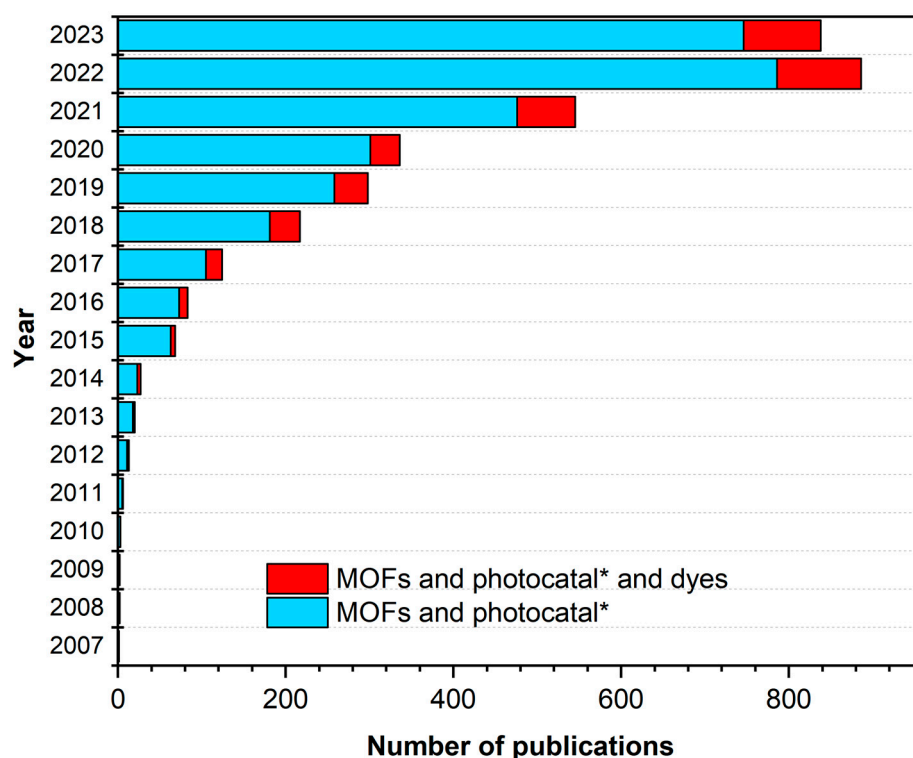


Figure 3. Historic publications of the MOFs as semiconductors. (Source: Scopus; keywords: MOFs and photocatal * and dyes).

When a beam of light interacts with the MOF, it allows the formation of charge carriers (holes (h^+) and electrons (e^-)) that are produced by the absorption of energy equal to or greater than the bandgap (E_g) [44]. However, these materials, due to their particular structure and organic nature, must take into account some additional and complementary considerations to common metallic semiconductors, such as (a) light absorption and photoexcitation: MOFs with photosensitive properties can play an active role since organic ligands and metal centers can absorb photons and convert a photogenerated excited state and transfer electrons from the ligand to the metal center [60–62]. This is possible due to the multiple possibilities that MOFs must be modified by replacing their organic linkers or metal sites, introducing molecules and nanoparticles within their structure, creating defects, or forming composites with other materials. In MOFs, charge transfer can occur via various mechanisms, leading to distinct charge transfer complexes (CTCs) with unique characteristics. Table 3 shows some common types of charge transfer in MOFs and their characteristics.

Table 3. Some types of charge transfer in MOFs and their characteristics.

Charge Transfer	Characteristics	Applications	Reference
Metal-to-Ligand Charge Transfer (MLCT)	In MLCT, electrons transfer from the metal center to the ligand. This typically involves a transition metal cation donating electrons to ligand molecules.	MLCT can enhance the photocatalytic activity of MOFs, making them useful for light-driven reactions.	[63–65]
Ligand-to-Metal Charge Transfer (LMCT) or Ligand-to-Cluster Charge Transfer (LCCT)	In LMCT, electrons transfer from the ligand to the metal center. This occurs when ligands act as donors to the metal ions.	LMCT can affect MOFs' electronic and optical properties, making them suitable for sensors and photoluminescent materials.	[66–69]

Table 3. Cont.

Charge Transfer	Characteristics	Applications	Reference
Ligand-to-Ligand Charge Transfer (LLCT)	LLCT involves electron transfer between two or more ligands within the MOF structure. This can lead to changes in the color and electronic properties of the MOF.	LLCT is often associated with color changes and can be utilized in chemical sensing and colorimetric indicators.	[70]
Guest-to-Host Charge Transfer (GHCT)	GHCT occurs when guest molecules or ions transfer electrons to or from the MOF framework. This type of charge transfer is essential in gas adsorption and separation.	GHCT is essential in gas storage and sensing applications, where guest molecules interact with the MOF structure.	[71]
Linker-Based Transition (LBT)	linker orbitals form the VB and CB.	This is valuable in applications like water and pollutant degradation	[72,73]
Metal-Based Transition (MBT)	In MBT, electrons transfer between metal centers within the MOF structure.	This is valuable in applications like pollutant degradation	[74]
Host-to-Guest Charge Transfer (HGCT)	HGCT occurs when the MOF's host structure transfers electrons to guest molecules or particles.	HGTC is significant in applications like gas sensing and photocatalytic degradation of dyes.	[73,75]

The characteristics and applications of these charge transfer types can vary depending on the specific MOF structure, the metal ions, the ligands involved, and the environmental conditions. The ability of MOFs to facilitate these types of charge transfer processes upon exposure to light makes them promising photocatalysts for a wide range of applications, including solar energy conversion, environmental remediation, and sustainable chemistry, and b) about the efficiency of these processes: these depend mainly on the type of material, its crystallinity and particle size. For example, high charge mobility is commonly associated with highly crystalline semiconductor materials in which oriented charge mobility is observed. Semiconducting nanocrystals with small particle sizes generally possess satisfactory charge transfer performance; However, if the particles are too small, recombination may occur due to enhanced surface defects [76]. Charge carrier recombination is a process in which photogenerated electrons and holes will recombine and, as a result, release energy obtained during excitation in the form of fluorescence or heat. In general, this process can be avoided mainly by decreasing the particle size of the photocatalyst.

In addition, this charge carrier recombination process can also reduce doping/impregnation/support in MOFs of metals (Au, Ag, Pt, Pd, etc.) [47,50,77], oxides, or mixtures of metal oxides. (TiO₂, CoFe₂O₄, ZnFe₂O₄, Ag₃VO₄, etc. [45,47,55], graphene oxide [78,79], quantum dots [80,81], and g-C₃N₄ [82,83], among others. The synthesis of these composites makes it possible to reduce the recombination of charge carriers by separating the photogenerated electrons and sending them to a metal, metal oxide, graphene oxide, or another MOF with a reduction potential less than the activated one. Figure 4 provides a scheme of the charge transfer pathway in the photocatalytic degradation of dyes employing a MOF and a semiconductor; a MOF (support), a semiconductor and a metal; and a MOF and a semiconductor with Z-scheme.

Figure 4a shows charge transfer mechanism proposed when a MOF and a semiconductor (SC) interact, where the MOF is activated. After, the photogenerated electrons migrate to the conduction band of the semiconductor, generating reactive species that oxidize of dye molecules and the holes in the valence band, also they generate ($\cdot$ OH) hydroxyl radicals that oxidize the dye molecules. While Figure 4b exemplifies a composite based on a MOF, a semiconductor, and a metal. In this case, the semiconductor is activated, generating the charge carriers, where the electrons migrate to the metal, and in this, the active species responsible for the oxidation of the dye are formed. In addition, water oxidation reactions take place in the valence band, creating hydroxyl radicals responsible for the

oxidation/degradation of the dye molecule. Finally, Figure 4c shows a composite with a Z-scheme, where the MOF is activated, generating charge carriers, the electrons from the conduction band migrate to the valence band of the semiconductor, these electrons migrate again to the conduction band of the semiconductor, generating in this site the reactive species responsible for the oxidation of the dye.

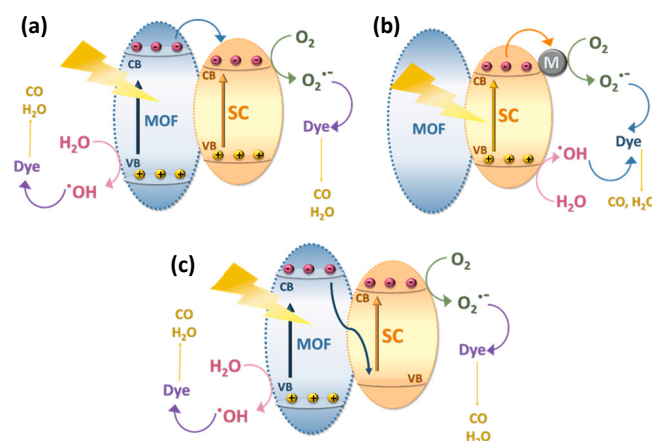


Figure 4. Scheme of charge transfer pathway in composite based MOFs; (a) MOF and semiconductor; (b) MOF, semiconductor and metal; and (c) MOF and semiconductor with Z-scheme.

1.4. Paragraph in the Degradation of Dyes Using MOFs

In recent years, there has been a growing interest in improving MOFs for use as semiconductors, particularly in photocatalytic applications [84–87]. These improvement strategies can be categorized into two fundamental axes, as depicted in Figure 5: (i) the synthesis of new MOFs with simple or complex ligands or mixtures of complex ligands and (ii) the synthesis of composite materials incorporating metals, metal oxides, carbon materials, or other MOFs, often combined with quantum dots (QDs) of carbon and metal oxides. Almost in all cases, this last axis allows us to improve the physicochemical, structural, optical, and textural properties of conventional MOFs, their thermal stability, and in aqueous environments, a key challenge when water is involved in MOF applications.

In the case of thermal stability, Healy et al. [88] identified several factors that affect the thermal stability of MOFs, such as the nature and position of functional groups, metal hardness, and the presence of coordinated solvent molecules. Also, they mentioned that thermal stability was found to have a relatively modest impact on the synthesis of MOFs by isorecticular expansion and interpenetration, where mixed ligands are generally present. They also proposed a possible thermal decomposition mechanism of MOFs, which they divided into two categories depending on whether the decomposition is ligand-centered, inorganic node-centered, or SBUs. However, ligand-centered decomposition is the dominant factor in determining the decomposition temperature of MOFs.

This section will review recent advancements along these two axes, analyzing strategies to enhance semiconductor efficiency in the photocatalytic degradation of dyes found in wastewater via heterogeneous photocatalysis.

1.5. Possible Degradation Mechanism of Dyes

Several studies have shown that the main reactive species in the photocatalytic degradation of dyes can be superoxide radicals ($\cdot\text{O}_2^-$), hydroxyl radicals ($\cdot\text{OH}$), and holes (h^+) [89,90]. These species are responsible for oxidizing the dye molecule until their mineralization ($\text{CO}_2 + \text{H}_2\text{O}$) or degradation of these into molecules that are sometimes more toxic than the dye itself. To achieve the above, the dye molecule, in interaction with the reactive species form reaction intermediates, which are finally mineralized into inorganic products. For example, Shi et al. [89] showed a possible reaction mechanism for the degradation of methyl blue (MB) dye (Figure 6). They mention that during the degradation, both S and N

in the molecular structure of MB underwent redox reactions to form amines and sulfonic acid groups as intermediates, which are finally further mineralized into inorganic products.

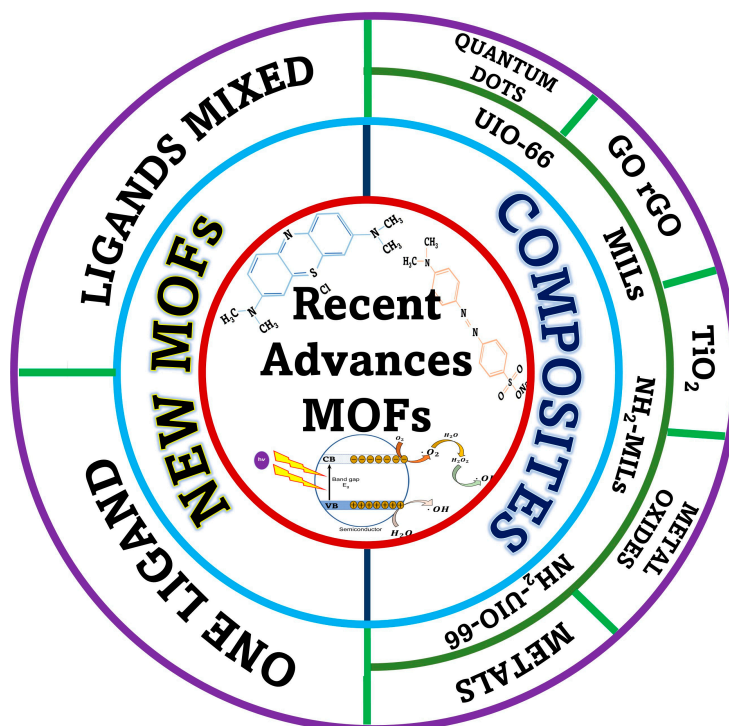


Figure 5. Strategies to improve the degradation of dyes using MOFs as semiconductors.

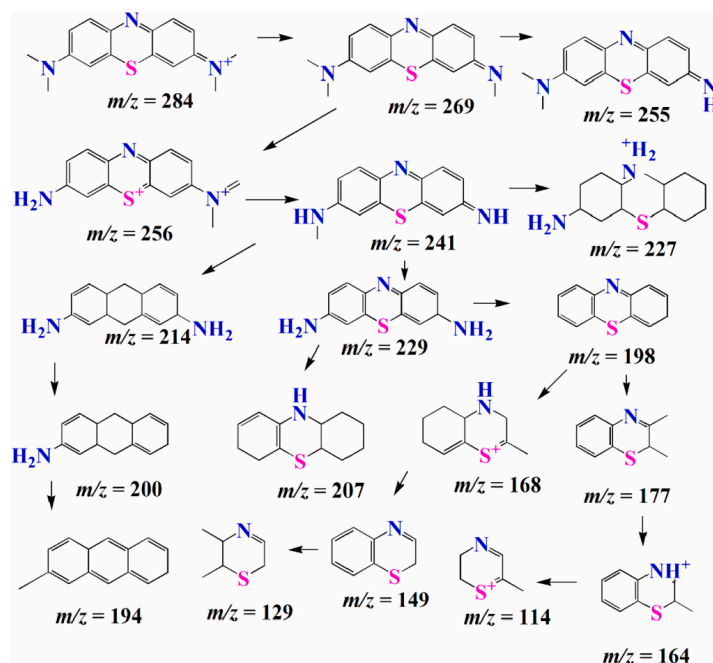


Figure 6. Step diagram of gradual degradation of MB dye. Taken with permission of Ref. [89].

Other authors also proposed a possible degradation mechanism of Rhodamine B (RhB) [51,90], methyl violet (MV) [91], and azure I [92].

Li et al. [90] determined using liquid chromatography–mass spectrometry (LC-MS) the possible intermediates of the reaction and therefore a possible degradation mechanism of Rhodamine B (RhB) (Figure 7). As shown in Figure 7, the proposed reaction mechanism for the degradation of Rhodamine B could begin with the formation of B1 ($m/z = 443.19$),

which is characteristic of the protonated form ($C_{28}H_{31}N_2O^{+3}$) of RhB (mechanism 1), or by the methyl oxidation of the latter to B2 ($m/z = 458.69$) (mechanism 2). Via mechanism 1, the protonated form of RhB could undergo a series of N-de-ethylation reactions to form macromolecular intermediates B3–9 ($m/z = 415.16, 387.15, 362.30, 330.90$), which subsequently, via de-amination, de-alkylation, de-carboxylation, and cleavage of the chromophore reactions allow the formation of smaller molecular intermediate B10–22 ($m/z = 318.27, 310.07, 279.11, 274.27, 258.19, 254.17, 244.03, 230.24, 213.07, 212.15, 167.03, \text{ and } 122.00$), which are finally further mineralized into inorganic products. Meanwhile, via mechanism 2, the direct formation of macromolecular intermediates B3–9 is possible, which also forms small molecular intermediates B10–22 via de-amination, de-alkylation, de-carboxylation, and cleavage of the chromophore reactions, where finally the latter are mineralized to inorganic products.

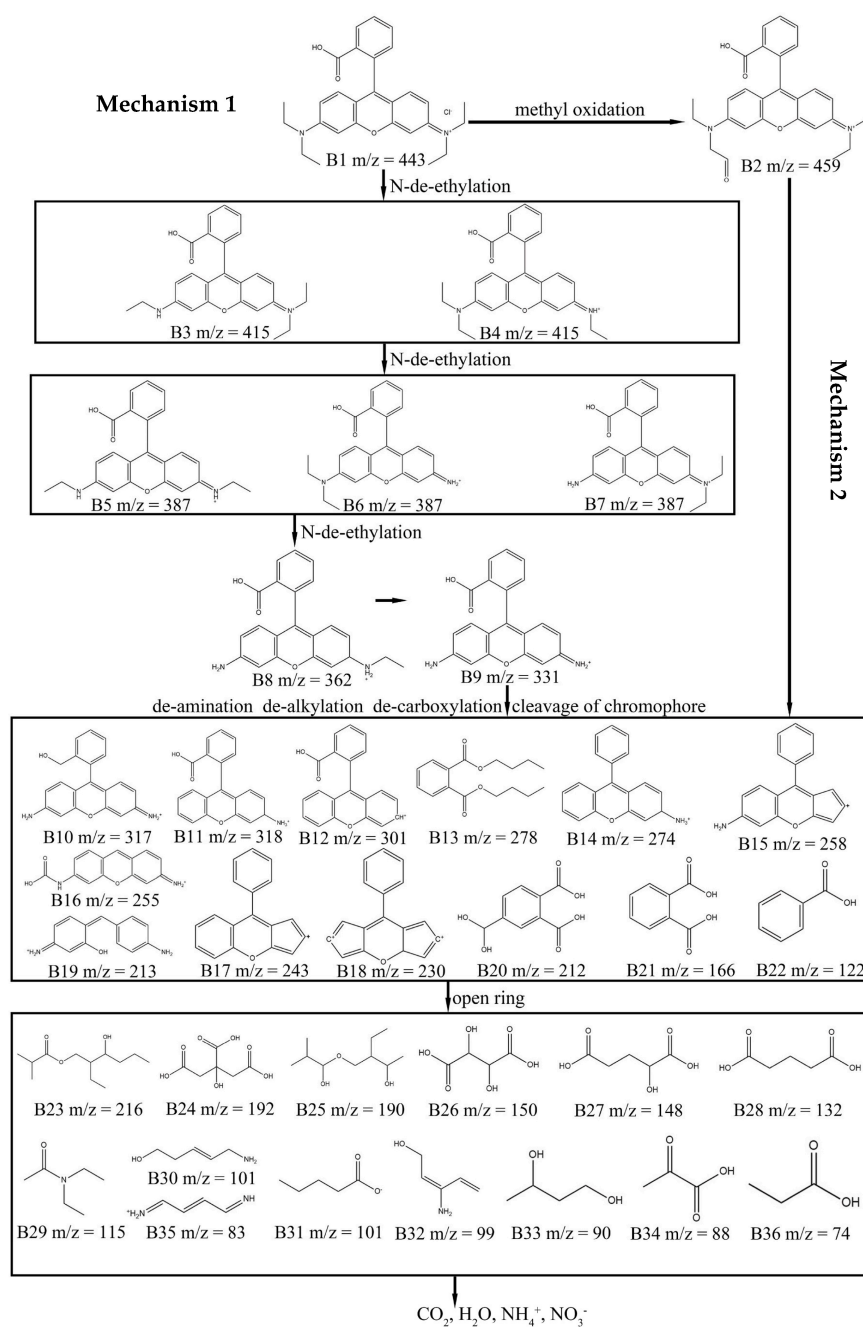


Figure 7. A possible degradation pathway of RhB. Taken with permission of Ref. [90].

However, despite the efforts made in the search for a reaction mechanism for the photocatalytic degradation of dyes, it is still necessary to use more sophisticated techniques, such as in situ and even theoretical studies, which allow for a more accurate determine the possible reaction intermediates, and thus be able to establish a more realistic reaction mechanism.

2. Recent Advances in the Photocatalytic Degradation of Dyes According to Classification

2.1. Acid Dyes

Table 4 presents recent advances in using MOFs as photocatalysts for degrading various dyes, including acid, reactive, direct, and mordant. Methyl Orange (MO) (I.C. Acid Orange 52), an azo-type dye (Figure 8), has been a critical acid dye used to evaluate the performance of MOFs in photocatalysis. Dyes are commonly employed in characterizing semiconductors due to their convenient monitoring via spectrophotometry. The degradation process is also visually observable, as the reduction in coloration can be detected with the naked eye. Recent research on MOFs in photocatalysis for dye degradation has explored several avenues, including synthesizing new MOFs, using multiple ligands, and doping MOFs with other semiconductors.

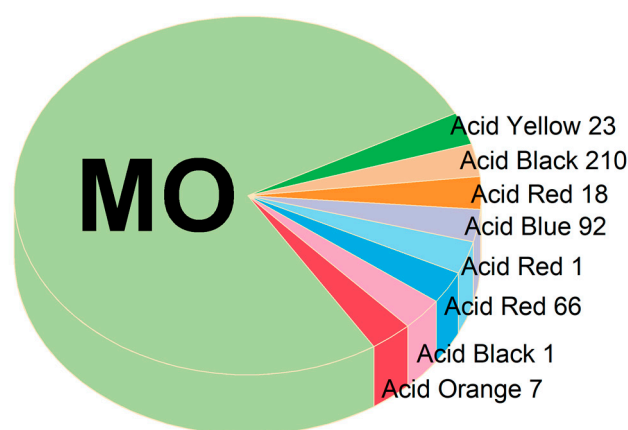


Figure 8. Acid dyes are most used in the characterization of MOFs in this review.

In recent years, the utilization of pristine MOFs has declined due to their inherent limitations, primarily related to their stability both thermally and in aqueous media. These MOFs rapidly decompose upon contact with water molecules. In the context of acid dyes, novel MOFs have been introduced, such as $[\text{Cu}_3(\text{L})_2(4'\text{-bipy})]$ [89], designed to enhance stability in aqueous media.

Another noteworthy example is the MOF La-PTC, composed of perylene-3,4,9,10-tetracarboxylate ligand and lanthanum metal cluster. This MOF achieved a remarkable 99.6% degradation of MO at 240 min, with the addition of hydrogen peroxide facilitating the formation of hydroxyl radicals, leading to the complete degradation of the dye. MOF La-PTC was activated using visible light [93], wherein the dye facilitated light absorption; this is activated, generating electrons that transitioned to the valence band of the MOF, ultimately yielding superoxide radicals (Figure 9).

The combination of ligands in the synthesis of MOFs has been applied in the last four years, providing better characteristics to the MOF and increasing its photocatalytic activity. For instance, MIL-88A catalysts with mixed $\text{C}_4\text{H}_6\text{N}_2$ -fumaric acid ligands were introduced, featuring small, highly crystalline particles. This MOF efficiently degraded to MO at a concentration of 100 ppm for 45 min [40]. In another case, a new 8-fold interpenetrated MOF was synthesized via a mixed-ligand system of benzene-*p*-dicarboxylic acid and tris(4-(4H-1,2,4-triazol-4-yl) phenyl)amine under hydrothermal conditions, demonstrating excellent water stability. Figure 10 illustrates the structure of the MOF with two ligands, highlighting the band positions. This MOF eliminated nearly 91% of MO due to the mixed

ligands, which reduced the band gap of the MOF (Figure 10d). Remarkably, it could be reused five times under the same conditions, achieving over 90% elimination of MO [94].

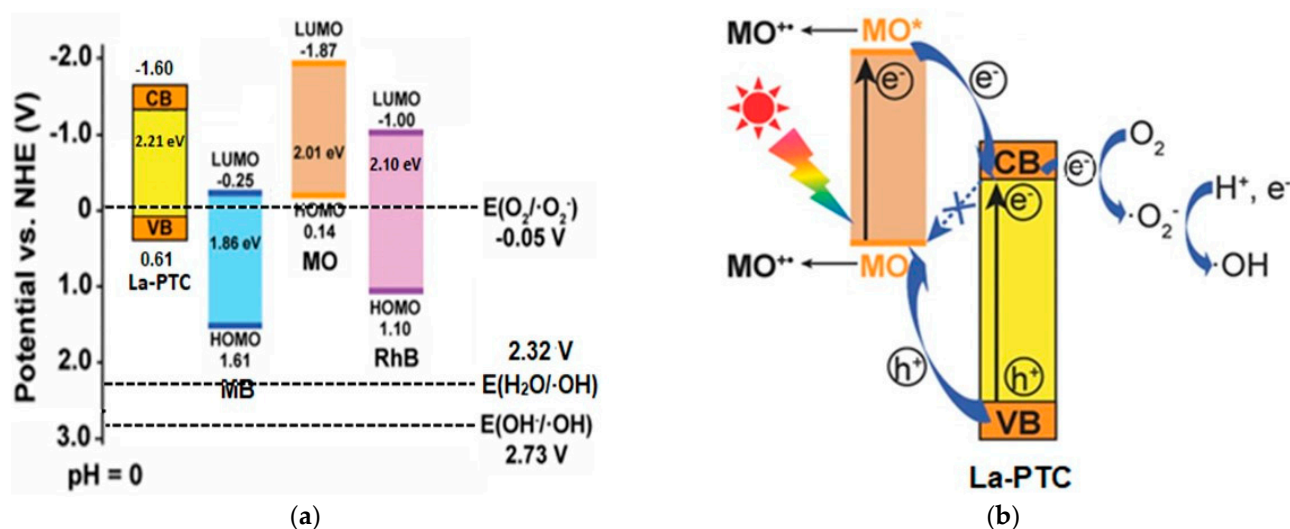


Figure 9. (a) positions of valence and conduction band of La-PTC photocatalyst and dyes, and (b) charge transfer mechanism for MO degradation over La-PTC photocatalyst under visible-light irradiation. Taken with permission of Ref. [93].

The mixed ligands also contributed to more excellent thermal stability, allowing these modified D-Fe-MOFs to eliminate 97% of MO in 48 min [95]. In a related study, identical ligands were applied to obtain Fe-MOFs, which exhibited a larger specific surface area, smaller particle size, a band gap of 2.55 eV, and a degradation rate of 97% within 48 min for MO (20 mg/L) under visible light [96].

The third configuration involves modifying MOFs by doping them with another semiconductor, forming a heterojunction between the two semiconductors. This configuration enhances the separation of generated photo species, increases the surface area, and improves light absorption in the visible region. Various semiconductors, such as MoS_2 [49], GO [41], and TiO_2 [42,43,97], have been employed, consistently achieving excellent results with over 90% degradation of MO. Moreover, all doped MOFs were activated using visible light. For instance, when MOFs were modified or doped with Ag_2CrO_4 [98,99], AO7 dye was effectively degraded in both cases, surpassing 90% degradation.

2.2. Basic-Dyes

There are several types of basic dyes, but in the literature, basic organic dyes like RhB, MV, and MB have been extensively investigated due to their common occurrence in wastewater, as evident in Table 4. Moreover, these dyes serve as target molecules in photocatalytic degradation reactions to assess the photocatalytic properties of semiconductors, including TiO_2 , ZnO, g- C_3N_4 , Fe_2O_3 , composite materials, and more recently, metal–organic frameworks (MOFs) and their composites. Simultaneously, another objective has been focused on the search for highly active photocatalysts for complete dye mineralization.

Several semiconductors have been studied in the photocatalytic degradation of dyes, such as ZnO, g- C_3N_4 , CeO_2 , TiO_2 , etc. However, TiO_2 is one of the most used semiconductors in the photocatalytic process. Nevertheless, it presents several disadvantages, such as a wide gap band, high recombination of the photogenerated charge carriers (electron-hole), low mechanical stability, etc. Hence, exploring alternative semiconductors for dye degradation reactions, like MOFs, is imperative. MOFs find extensive use as semiconductors in the photocatalytic degradation of dyes due to their exceptional properties, including a high specific surface area, large pore volumes, and exposed metal centers that facilitate catalytic

and photocatalytic reactions. Additionally, MOFs can adsorb substantial quantities of dyes due to their abundant active sites [20].

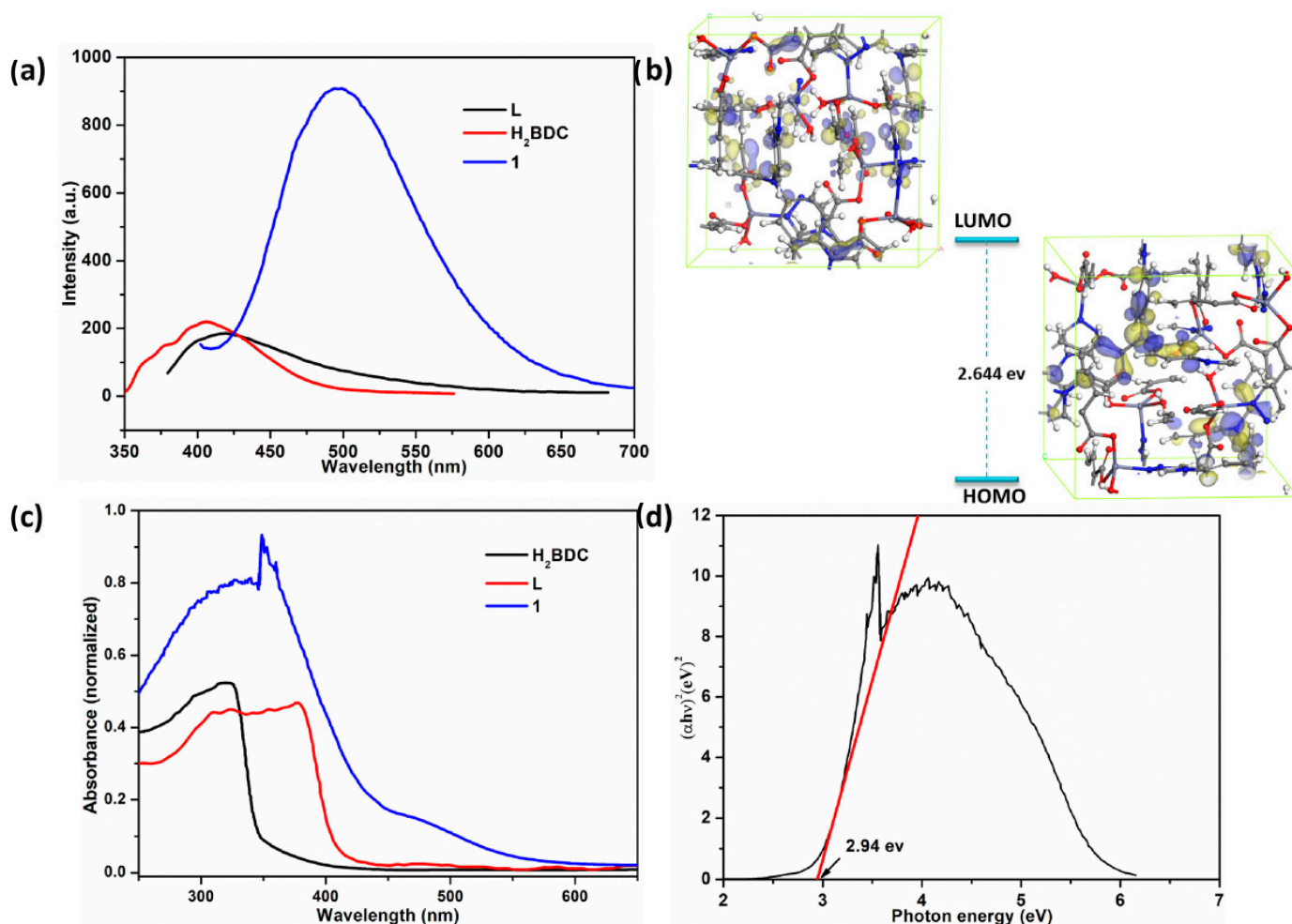


Figure 10. (a) The solid-state luminescent emission spectra of MOF and the ligands. (b) The calculated HOMO and LUMO for the periodic network of MOF. (c) UV-vis diffuse reflectance spectra for MOF and ligands. (d) Diffuse reflectance spectra of Kubelka–Munk function versus energy of MOF. Taken with permission of Ref. [94].

In the degradation of basic dyes, it is noteworthy that the most researched MOFs include UiO-66, NH₂-UiO-66, MIL-53 (Al), MIL-53 (Fe), NH₂-MIL-53 (Fe), NH₂-MIL-125 (Ti), MIL-88 (Fe), NH₂-MIL-101 (Fe), MIL-100 (Fe), and Cu-BDC. These MOFs have gained attention due to their excellent properties, such as high specific surface area, large pore volumes, and good stability in aqueous environments. Various research groups have concentrated on enhancing pristine MOFs by doping them with metals, metal oxides, mixed oxides, and even quantum dots. These materials modify pristine MOFs' physical, chemical, optical, and textural properties. The primary focus has been changing their optical properties to reduce the bandgap and minimize the recombination of charge carriers, thereby enhancing their photocatalytic activity.

Recent advancements in the photocatalytic degradation of Rhodamine B (also known as “Basic Violet 10” according to color index) have been centered on identifying solid materials for its degradation and mineralization (Table 4). For example, Gayathri et al. investigated the band structure of Zr-based metal–organic frameworks (Zr-MOFs) with different ligands, such as terephthalic acid (UiO-66), 2-amino terephthalic acid (NH₂-UiO-66), and 2-methylimidazole (UiO-66-2M), synthesized via the hydrothermal method. They applied them in the photocatalytic degradation of Rhodamine B [53] (Figure 11). They

demonstrated that the optical properties of MOFs are influenced by the type of ligand, which can act as an antenna to capture photons and direct them to metal sites for oxidation and reduction reactions. $\text{NH}_2\text{-UiO-66}$ exhibited the highest efficiency, achieving 95% degradation at 120 min (under reaction conditions: pH = 7, 10 ppm RhB, and catalyst concentration = 0.4 mg/L). The presence of amino groups in the organic ligand (2-amino terephthalic) used during MOF synthesis enables photon capture electron generation to fill the LUMO energy level and, subsequently, reduction in the bandgap, thereby improving photocatalytic activity in Rhodamine B degradation.

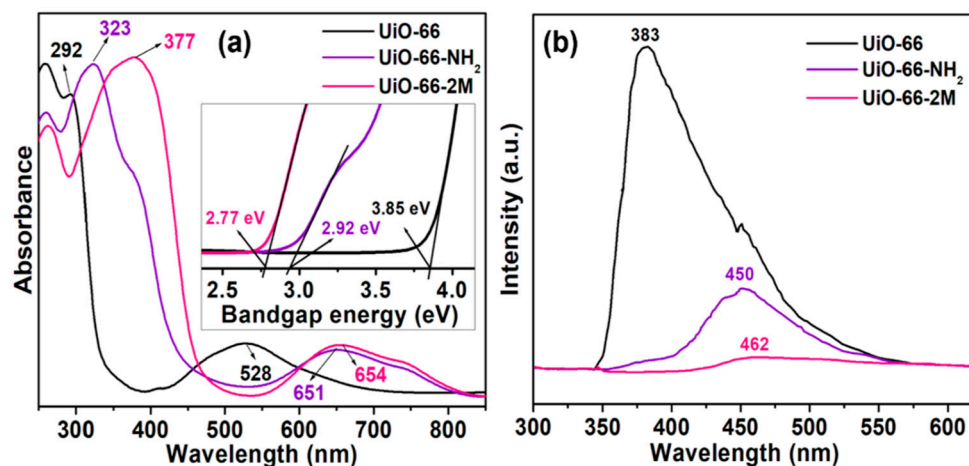


Figure 11. (a) UV-visible absorption spectra (insert-Tauc's plots for band gap estimation) and (b) photoluminescence spectra of the synthesized different liganded-UiO-66 MOFs. Taken with permission of Ref. [53].

The photocatalytic degradation of Methyl Violet (also known as “Basic Violet 3” according to the Color Index) has primarily focused on synthesizing new MOFs using novel or mixed ligands. The development of MOF-composite materials has been a primary strategy to enhance the optical properties of pristine MOFs and their photocatalytic performance. Sofi et al. [50] demonstrated that incorporating Ag_2WO_4 and silver nanoparticles into UiO-66 improves the efficiency of Rhodamine B degradation (Figure 12). They observed that this enhancement was attributed to the synergistic effect between Ag_2WO_4 and UiO-66, along with the surface plasmonic impact of silver nanoparticles. Furthermore, this composite material exhibits excellent stability even after five cycles.

Another interesting study was conducted by Liu et al. [54] via a hydrothermal method and employed them in the solar light-driven photocatalytic degradation of Rhodamine B. They degraded 30 mg/L of RhB within 10 min using a catalyst of 0.3 g/L under solar light. While using sacrificial agents in contaminant degradation is not typically recommended, their presence enhances photocatalytic activity, as demonstrated in a study by Mahmoodi et al. [47]. They showed that adding H_2O_2 (20 mL) as a sacrificial agent considerably enhances the photocatalytic activity of $\text{Ag}/\text{CoFe}_2\text{O}_4/\text{NH}_2\text{-MIL-125 (Ti)}$ composite materials. Their results revealed that nearly 98.8% of 20 mg/L RhB could be removed within 10 min under visible light irradiation. Additionally, they established the stability of this composite material.

Figure 13 shows the semiconductor's activation mechanism without and with a sacrificial agent such as hydrogen peroxide. Figure 13a illustrates the formation of the active species when they do not have a sacrificial agent. Moreover, Figure 13b shows the interaction of a sacrificial agent (H_2O_2) with electrons photogenerated in the conduction band of the semiconductor, allowing the formation of hydroxyl radicals, which are generally the active species that promote the degradation process of dyes [100,101]. In both mechanisms, the formation of the free radical is possible; however, when the sacrificial agent is present, the formation is direct.

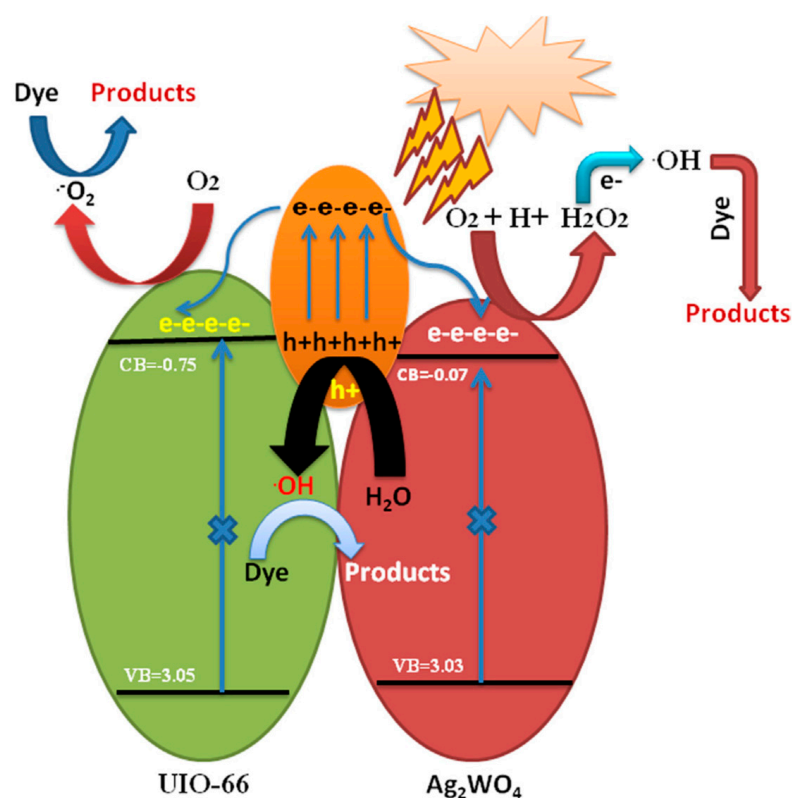


Figure 12. Schematic diagram showing the band structure and Z-scheme separation of photoinduced electron-hole pairs at interface of Ag/Ag₂WO₄/UIO-66 catalyst under visible light irradiation. Taken with permission of Ref. [50].

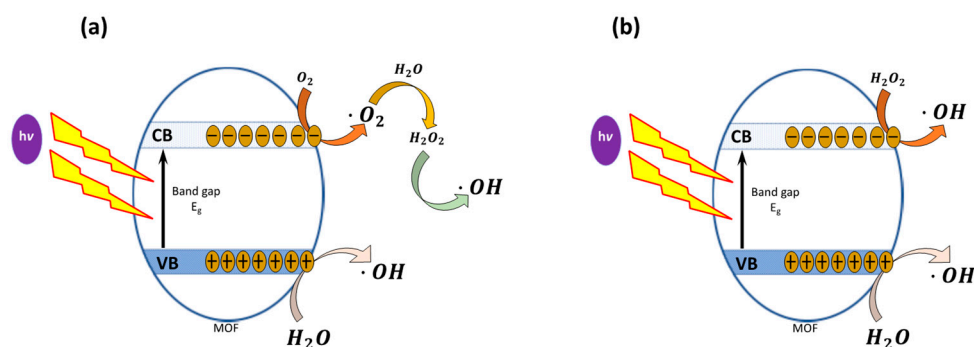


Figure 13. Scheme of the generation of active species for the degradation of dyes, (a) without sacrificial agents (hydrogen peroxide) and (b) with sacrificial agents (hydrogen peroxide).

In recent years, the search for new MOF-based semiconductors for the photocatalytic degradation of dyes has focused on using complex ligands [94,102,103]. For instance, the synthesis of 3D supramolecular structures via L (H₄L = 1, 1':4', 1'':4'', 10''-quaterphenyl-2, 5, 20'', 50''-tetracarboxylic acid) ligand linkage has been explored. Wu et al. [17] synthesized two isomorphic MOFs of manganese and cadmium with H₄L ligand. These materials were used in the photocatalytic reaction of Rhodamine B under UV irradiation (Table 2). However, MOF-based manganese shows a higher catalytic performance compared with MOF-based cadmium. Also, the authors proposed a possible reaction mechanism for the photocatalytic degradation of dye using LC-MS. They found that the chains/rings in the main structure of RhB happened redox reactions to generate amine groups, which are oxidized by strong oxidizing groups, mainly OH radicals.

Other authors have studied MOFs using a mixed-ligand system [89,94,104,105], where these materials have shown good photocatalytic activity in the degradation of basic dyes

such as MB, RhB, and MV. Chen et al. [94] show that MOF synthesis based on zinc clusters, benzene-*p*-dicarboxylic acid (H_2BDC), and tris(4-(4H-1,2,4-triazol-4-yl) phenyl)amine (L) as organic ligands (Figure 14) offers excellent water stability. They also found that both ligands enable inter-ligand-based charge transfer and reduce the energy gap compared to MOF with the BDC ligand. This new material is promising in the degradation of basic dyes. Also, this can be activated with visible irradiation (Table 4).

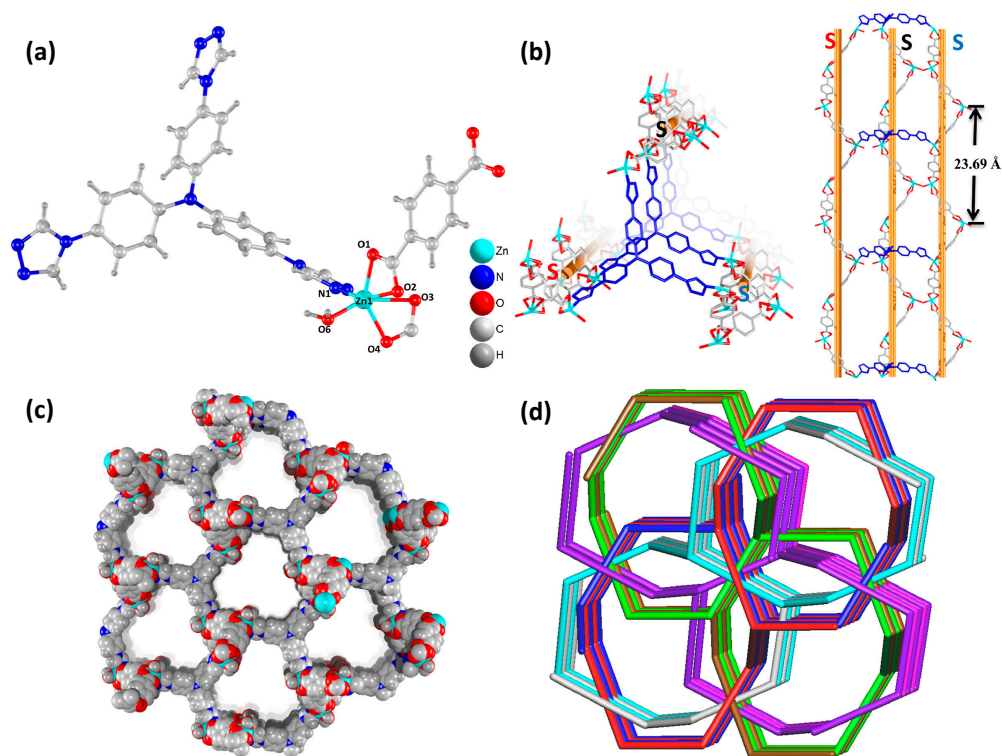


Figure 14. (a) View for the six coordinated Zn (II) ions of MOF; (b) the left-handed helical chains of MOF; (c) the 3D porous framework ligands and helical chains; (d) the 8-fold interpenetrated *srs* net of MOF. Taken with permission of Ref. [94].

For the photocatalytic degradation of cation dye azure I, Ren et al. [92] synthesized Mg-MOF using $MgCl_2$, H_4tptc , and CH_3CN via solvothermal method at 150 °C. The degradation rate of Az I reached 100% within 50 min. After five cycles, the catalyst showed that its original structural framework remained intact during the photocatalytic degradation.

2.3. Reactive Dyes

In the photocatalytic degradation of the synthetic textile dye Reactive Yellow 145 in an aqueous solution, different MOF photocatalysts were used under UV and visible irradiation. Metallic (Co-BTC/CN and Fe-BTC/CN) and bimetallic (Co-Fe-BTC/CN) nanocomposites were examined in the degradation of Reactive Yellow 145 by Nguyen et al. [106]. They introduced nanocomposites based on metallic and bimetallic MOF and CN sheet using the microwave-assisted hydrothermal method and investigated the effect of Co:Fe molar ratio on the decomposition of Reactive Yellow 145 in water. The Co-Fe-BTC/CN sample exhibited a higher efficiency than CN, Fe-BTC, and Fe-BTC/CN, with almost 99.38% removal of RY-145 after 40 min. The stability of the Co_2-Fe_8-BTC/CN catalyst was determined, and no change in RY-145 dye removal efficiency was observed after six reaction cycles, indicating that the catalyst is stable and reusable. On the other hand, $\alpha-Fe_2O_3@C@SiO_2/TiO_2$ magnetic nanocomposites were successfully synthesized from MOFs and then coated with silica and titanium particles via sol-gel and solvothermal methods by Mousavi et al. [107]. The removal efficiency of Reactive Yellow 145 was about 97.5% at 150 mgL^{-1} of dye concentration, pH = 2, 15 $mg/100 mL^{-1}$ of photocatalyst, 25 °C reaction

temperature, and 90 min under UV irradiation. The particles exhibited excellent stability and reusability because they can maintain a relatively high removal efficiency of about 97% of their initial performance after five reuse cycles. It can be concluded that TiO_2 coating on magnetic particles increases their photocatalytic degradation efficiency and reusability.

For Reactive Red 198 degradation, AC, MIL-88B (Fe), and AC/MIL-88B (Fe) photocatalysts were synthesized by Mahmoodi et al. [108]. This study used a newly prepared activated carbon from Kiwi peel to synthesize AC/MOF nanocomposite to improve the MOF catalytic activity. The photocatalytic degradation of Reactive Red 198 (RR198) by the AC/MIL-88B (Fe) composite achieved 99% removal at 0.08 g/L of catalyst dosage, pH of 3, and a dye concentration of 50 mgL^{-1} .

2.4. Other Dyes

To study the degradation of Direct Red 28 under sunlight at room temperature, Z. Saedi et al. [109] used TMU-16. The experimental results showed a dye degradation of 96% after 1 h with a 20 mgL^{-1} dye concentration. After three cycles, the degradation of the dye remained at about 95%. When TMU-16 is exposed to light photons, the $\text{Zn}(\text{O})_4$ metal–oxygen clusters act as semiconductors, forming electron–hole pairs. So, in this work, the ability of TMU-16 for significant photocatalytic degradation of Congo red (CR). Furthermore, researchers [110–112] reported strong adsorption of CR, which influences the degradation process and results in a negligible or prolonged degradation rate. The amino groups in CR achieved the adsorption of CR onto the catalyst as both donors and acceptors of electrons, enabling H-bonds to form between the amino organic dye and the uncoordinated O atoms of carboxyl groups in MOFs [112]. In the same research, Z. Saedi et al. [109] used TMU-16 to study the degradation of Eriochrome Black T under sunlight at room temperature. Eriochrome Black T showed 96% degradation after 180 min. Alternatively, a new metal–organic framework $\{[\text{Co}_3(\text{BTC})_2(\text{Bimb})_{2.5}] \cdot 2\text{H}_2\text{O}\}_n$ (1) (Bimb = 1,4-bis[(1H-imidazol-1-yl)methyl]benzene and H_3BTC = 1,3,5-Benzenetricarboxylic acid) was synthesized by Somnath et al. [113]. The mixed ligand 3D Cobalt was examined in the degradation of Eriochrome Black T in an aqueous solution with almost 77.5% removal of EBT after 135 min under UV light. The complex displayed high reusability, efficiency, and low cost of adsorbency with great potential in handling dye-contaminated wastewater.

Table 4. Review the most recent studies using MOFs for the photocatalytic degradation of dye.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
Acid Red 1	Cl-M88A (MIL-88A (Fe)) (1) NIT-M88A (MIL-88A (Fe)) (2)	2.17 2.19	120 90	100 100	20 mg/L dye; pH = 7; conc. catalyst = 1 g/L; 0.03 mL of H ₂ O ₂ ; Xe lamp	stable	[40]
PBS/Acid Red 66 RhB/Basic Violet 10	Ag/Ag ₂ WO ₄ /UiO-66	2.06	25 (PBS) 60 (RhB)	95 (PBS) 90 (RhB)	60 mg/L dye, pH = 7; conc. catalyst = 0.2 g/L; Halogen Lamp	stable	[50]
Acid Black 1	MIL-53 (Cr)/polymer (1) HKUST-1 (Cu) (2)	2.1–2.4	30	96 90	UV lamp	stable	[114]
MB/Basic Blue 9 MV/Basic Violet 3 RhB/Basic Violet 10	[Co ₄ (bbibp) ₅ (HCOO) ₈ (H ₂ O) ₂] _n (1) [Co(bimmb)(NO ₃) ₂] _n (2)	2.50 3.07	75	(1): 100 (MB); 50.8 (MV); 61.0 (RhB) (2) 94 (MB); 68.7 (MV); 64.4 (RhB)	6 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; H ₂ O ₂ = 0.5 mL; UV high pressure lamp	-	[57]
RhB/Basic Violet 10 CB/Crome blue MB/Basic Blue 9	[Mn ₄ (NDC) ₂ (bb) ₄]	2.53	100	94.33 (RhB) 34.91 (CB) 29.35 (MB)	10 mg/L dye; pH = 7; conc. catalyst = 0.8 g/L; Hg Lamp	stable	[102]
AO7/AO7 MB/Basic Blue 9	CF/MoS ₂ /NH ₂ -MIL-125(Ti)	2.5	120	67.9 (AO7) 94.3 (MB)	10 mg/L dye; pH = 7; conc. catalyst = 1.5 g/L; visible light	stable	[49]
MB/Basic Blue 9	SrAl ₂ O ₄ :Eu ²⁺ ,Dy ³⁺ /g-C ₃ N ₄ @NH ₂ -UiO-66	2.77	30	95	20 mg/L dye; pH = 7; conc. catalyst = 0.4 g/L; Xe lamp	stable	[51]
MV/Basic Violet 3	[Co ₂ (μ ₃ -OH) (L)(H ₂ O) ₃ ·2·75H ₂ O] _n (1) [Co ₂ (μ ₃ -OH)(L)(bib)(H ₂ O) ₂] _n (2)	3.02 1.91	40	55.3 (1) 61.3 (2)	10 mg/L dye; pH = 7; conc. catalyst = 0.1 g/L; Hg lamp	stable	[115]
MV/Basic Violet 3	[Zn ₂ (1,4- BDC) ₂ (bmp)•DMF] (1) [Cd ₃ (H ₂ O) ₂ (bmp) ₂ (DMF) (BTC) ₂ •3•5H ₂ O•1•5DMF](2)	- -	40	70.1 (1) 37.83 (2)	-	stable	[100]
MV/Methyl Violet CV/Basic Red 5 NR/Basic Red 5 CR/Direct Red 28	{[Co ₃ (BTC) ₂ (Bimb) _{2.5}]·2H ₂ O} _n	3.94	135	68.7 (MV) 77.5 (CV) 19.5 (NR) 15.8 (CR)	10 mg/L dye; pH = 7; 5 mL of methanolic solution (0.01 mmol powder in 50 mL methanol); UV lamp	-	[113]
MB/Basic Blue 9 MV/Basic Violet 3	[CoL (bimb) _{0.5}] _n (1) {[CoL (bimmb)] DMF} _n (2) [CoL (bimmb) _{0.5}] _n (3) [CoL (bbibp)] _n (4)	- -	120	MV: 85.5 (1); 78.4 (2); 84.2 (3); 96.2 (4) MB: 45.8 (1); 65.0 (2); 82.9 (3); 84.0 (4)	6 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; Hg lamp	stable (4)	[58]

Table 4. Cont.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
RhB/Basic Violet 10	$\{[\text{Cu}_8\text{Cl}_5(\text{CPT})_8(\text{H}_2\text{O})_4](\text{HSiW}_{12}(\text{H}_2\text{O})_{20}(\text{CH}_3\text{CN})_4)_n\}$	2.69	70	97	10 mg/L dye; pH = 7; conc. catalyst = 0.375 g/L; H_2O_2 = 1 mL; visible light	stable	[94]
CV/Basic Violet 3 AzI/52010 NR/Basic Red 5	Mgtptc based on MgO_6 octahedral (Mgtptc)	2.84	55 50 55	90.94 (CV) 100 (AzI) 94.39 (NR)	-	stable	[92]
MB/Basic Blue 9 MV/Basic Violet 3 MG/Basic Green 4	$\{[\text{Mn}_2(\text{L})(\text{tib})(\text{H}_2\text{O})] 3\text{DMA } 4\text{H}_2\text{O}\}_n$ (1) $\{[\text{Co}_2(\text{L})(\text{bipd})_2] 3\text{H}_2\text{O}\}_n$ (2) $\{[\text{Co}(\text{L})_{0.5}(\text{bimb})] 3\text{H}_2\text{O } 0.5\text{O}_2\}_n$ (3)	3.35 3.37 3.22	120	(1): 59.6 (MB); 63.6 (MV); 55.2 (MG); (2) 85.8 (MB); 88.8 (MV); 82.3 (MG) (3) 91.9 (MB); 93.8 (MV); 92.8 (MG)	6 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; UV light	stable (compound 2,3)	[116]
MV/Basic Violet 3 MB/Basic Blue 9	$\text{Co}(\text{L})(\text{tib})(-\text{H}_2\text{O})_2)_n$ (1) $[\text{Co}(\text{L})(\text{bip})_{0.5}]_n$ (2)	2.63 2.80	120	(1): 92.1 (MV); 75.2 (MB) (2): 89.7 (MV); 57.7 (MB)	10 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; Hg lamp	stable	[59]
MB/Basic Blue 9 MV/Basic Violet 3 RhB/Basic Violet 10	Cu-tipe/GO composite	1.35	30 45 60	90.4 (MB) 92.2 (MV) 91.3 (RhB)	10 mg/L dye; pH = 7; conc. catalyst = 0.4 g/L; H_2O_2 = 0.5 mL; visible light	-	[78]
MB/Basic Blue 9	MIL-53(Al)@ TiO_2	3.05	240	95	20 mg/L dye; pH = 7; conc. catalyst = 0.1 g/L; visible light	-	[55]
MB/Basic Blue 9	Cu(tpa)-1@GO	-	30	93.3	visible light	-	[79]
RhB/Basic Violet 10	$[\text{EuCd}(\text{pbbp})_2(\text{SO}_4)_2\text{Cl}_2(\text{H}_2\text{O})_3]$	-	120	92	1×10^{-5} M dye; pH = 7; conc. catalyst = 1 g/L; H_2O_2 = 1 mL; Xe lamp	stable	[117]
CR/Direct Red 28 MB/Basic Blue 9 RhB/Basic Violet 10	Co-doped MIL-53(Al)	2.63	16 (CR) 12 (MB) 16 (RhB)	99 (CR) 99 (MB) 99 (RhB)	30 mg/L dye; pH = 7; conc. catalyst = 0.3 g/L; Sunlight	stable	[54]
RhB/Basic Violet 10 MB/Basic Blue 9 MO/Acid Orange 52	$\{[\text{Zn}_3(\text{L})(\text{BDC})_3(\text{H}_2\text{O})_3] 3.5\text{H}_2\text{O}\}_n$	2.94	70	90 (RhB) 90 (MB) 90 (MO)	15 mg/L dye; pH = 7; conc. catalyst = 0.33 g/L; UV light	-	[94]

Table 4. Cont.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
MB/Basic Blue 9	[Cd(4-Hptz) ₂ (H ₂ O) ₂ Cl ₂] (1) [Cu(btx) ₂ (ClO ₄) ₂] _n (2) [Cu(btx)(ClO ₄)] _n (3)	4.12 3.58 2.59	100	93.43 (1) 47.32 (2) 14.60 (3)	10 mg/L dye; pH = 7; conc. catalyst = 0.175 g/L; Hg lamp	stable	[118]
MB/Basic Blue 9 MO/Acid Orange 52 RhB/Basic Violet 10	[Cu ₃ (L) ₂ (4,4'-bipy)]	2.82	100	23.19 (RhB) 9.26 (MO) 81.17 (MB)	10 mg/L dye; pH = 7; catalyst: 40 mg	stable	[89]
RhB/Basic Violet 10	bimetallic MOF anchored corncob calcined derived activated carbon (CCAC)	2.55	75	100	20 mg/L dye; pH = 7; conc. Catalyst = 0.05 g/L; Xe lamp	stable	[119]
RhB/Basic Violet 10	Ni-MOF (1) NH ₂ -MIL-101(Fe) (2) Ti-MOF (3)	2.77 1.63 2.55	90	9 (1) 90 (2) 50 (3)	20 mg/L dye; pH = 7; conc. Catalyst = 1 g/L; Solar light	stable	[56]
RhB/Basic Violet 10	Zr ₆ O ₄ (OH) ₄ (ABDC) ₆ /natural sepiolite (Sp) clay	2.85	120	64	20 mg/L dye; pH = 5.9; conc. catalyst = 0.5 g/L visible light	-	[120]
NR/Basic Red 5	{[Co(bcpt)(bib) _{3/2} (H ₂ O)] H ₂ O} _n (1) {[Ni(bcpt)(bib) _{3/2} (H ₂ O)] H ₂ O} _n (2)	3.6 2.9	100		See information supplementary		[121]
MV/Basic Violet 3	[Co ₄ (μ ₃ -OH) ₂ (H ₂ O) ₂ (L) ₂ (bib) _{1.5} (CH ₃ CN) 3H ₂ O CH ₃ CN] (1) [Co ₈ (μ ₃ -OH) ₄ (L) ₄ (bib) ₄ 8CH ₃ CN] (2)	3.64 2.88	100	67.99 78.22	UV light	-	[91]
MV/Basic Violet 3	[NH ₂ (CH ₃) ₂][Zn ₂ O(bmp)(BTC)] (1) [NH ₂ (CH ₃) ₂][Zn ₂ O(bmp)(BTC)]·1.5DMF (2)	3.63 3.96	40	38.25 55.95	10 mg/L dye; pH = 7; conc. catalyst = 0.8 g/L; UV lamp	Stable	[122]
MB/Basic Blue 9	CdS/Cd-MOF	2.36	100		20 mg/L dye; pH = 7; conc. catalyst = 0.1 g/L; Xe lamp	Stable	[123]
MB/Basic Blue 9 CR/Direct Red 28 MO/Acid Orange 52 RhB/Basic Violet 10	CuCd-BMOF/GO-x	2.7	40	90.1 (MB) 85.4 (CR) 79.6 (MO) 87.6 (RhB)	20 mg/L dye; pH = 7; conc. catalyst = 0.6 g/L; Xe lamp	Stable	[41]
MV/Basic Violet 3 MG/ RhB/Basic Violet 10	NH ₂ -UiO-66/FeOOH quantum dots	2.53	180	82.2 (MV) 94.3 (MG) 81.5 (RhB)	20 mg/L dye; pH = 7; conc. catalyst = 0.6 g/L; 30 mM H ₂ O ₂ ; LED lamp	stable	[80]

Table 4. Cont.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
MB/Basic Blue 9	NH ₂ -MIL-125(Ti)/polysulfone membrane	-	300	88	10 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; visible light	-	[48]
RhB/Basic Violet 10	Bi-TATB	3.21	180	99.1	3×10^{-5} M dye; pH = 7; conc. catalyst = 0.1 g/L; LED light	stable	[124]
MB/Basic Blue 9	Ce-MOF/g-C ₃ N ₄ -TS	2.45	120	100	10 mg/L dye; pH = 7; conc. catalyst = 0.25 g/L; UVlight	stable	[125]
MB/Basic Blue 9 MV/Basic Violet 3 RhB/Basic Violet 10	{[Ni(L)(bpt)(H ₂ O)] 2H ₂ O} _n	-	90	89.5 (MB) 84.7 (MV) 76.4 (RhB)	6 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; UV light	-	[104]
MO/Acid Orange 52	STA-12Fe	2.6	40	12	10 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; Sun light	stable (Fe)	[126]
MB/Basic Blue 9 CR/Direct Red 28 MO/Acid Orange 52 RhB/Basic Violet 10	CuCd-BMOF/GO-x	2.7	40	92 (Mb) 90.1 (CR) 85.4 (MO) 79.6 (RhB)	10 mg/L dye; pH = 7; conc. catalyst = 0.6 g/L; Xe lamp	stable	[41]
MB/Basic Blue 9 MV/Basic Violet 3 RhB/Basic Violet 10	{[Cd(HL)(tib)(H ₂ O)] 1.5H ₂ O} _n (1) {[Co ₃ (L) ₂ (tib) ₂] 4H ₂ O} _n (2) [Cu(L)(3,5-bibp)] _n (3)	3.13 3.32 3.37	90	(1): 87.4 (MB); 67.3 (MV); 66.5 (RhB) (2): 83.7 (MB); 48.4 (MV); 33.5 (RhB) (3) 86.9 (MB); 55 (MV); 53.4 (RhB)	6 mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; UV light	stable	[112]
RhB/Basic Violet 10	[Mn(L) _{0.5} (H ₂ O)] (1) [Cd(L) _{0.5} (H ₂ O)] (2)	2.84 3.05	150	90.5 (1) 70.5 (2)	10 mg/L dye; pH = 7; conc. catalyst = 1 g/L; UV light	stable	[105]
RhB/Basic Violet 10	Ag/AgCl@CoFe ₂ O ₄ /NH ₂ -MIL-125(Ti)	2.73	10	97	20 mg/L dye; pH = 9; conc. catalyst = 0.8 g/L; H ₂ O ₂ = 20 µL; Visible light	stable	[47]
MB/Basic Blue 9 RhB/Basic Violet 10	MoS ₂ @MIL-88(Fe)	2.75	60	98.5 (MB) 97.4 (RhB)	50 mg/L RhB; pH = 7; conc. catalyst = 1 g/L; Xe lamp	stable	[127]
MB/Basic Blue 9	Ag ₃ PO ₄ /UiO-66(Zr)/g-C ₃ N ₄	2.9	240	61.7	10 mg/L dye; pH = 7; conc. catalyst = 0.7 g/L; Visible light		[128]

Table 4. Cont.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
MB/Basic Blue 9 MV/Basic Violet 3	[Cd(bimd)(Hbipa)(H ₂ O)] _n (1) [[Zn(bipd)(Hbipa)] H ₂ O] _n (2) [Zn(bimb)(Hbipa)] _n (3)	2.24 2.55 2.36	120	(1): 90.3 (MB); 85.7 (MV) (2): 89.8 (MB); 89 (MV) (3): 92.9 (MB); 95.3 (MV)	6mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; UV light	-	[111]
AO7/Acid Orange 7 MB/Basic Blue 9 CR/Direct Red 28	Ag ₂ CrO ₄ /MIL-53(Fe)	2.22	140	95.5 (AO7) ~60 (MB) ~80 (CR)	20mg/L dye; pH = 6; conc. catalyst = 1 g/L; Halogen lamp	stable	[98]
MB/Basic Blue 9 MV/Basic Violet 3	NH ₂ -MIL-101(Fe)@CuCoNi	2.56	120	99.8 (MB) 93.8 (CV)	25mg/L dye; pH = 6; conc. catalyst = 1 g/L; 20 µL H ₂ O ₂ ; Visible light	stable	[129]
MG/Basic Green 4 MO/Acid Orange 52	[[Zn ₂ (DIPA) ₂ (bimp) ₅]-DMF 2H ₂ O] _n (1) [[Zn ₂ (HBPPA) ₂ (bip) ₂]-2H ₂ O] _n (2)	2.08 2.26	120	(1): 96.6 (MB); 78.6 (MV) (2): 98.3 (MB); 96.9 (MV)	6mg/L dye; pH = 7; conc. catalyst = 0.2 g/L; Hg light	stable	[130]
RhB/Basic Violet 10	g-C ₃ N ₄ /ZnFe ₂ O ₄ /UiO-66	2.8	60	98	visible light	stable	[83]
RhB/Basic Violet 10	NH ₂ -MIL-101(Fe)/copper oxide		50	100	50 mg/L dye; pH = 6.5; conc. catalyst = 0.5 g/L; LED	-	[131]
MB/Basic Blue 9 MO/Acid Orange 52	Al-PcCl@NH ₂ -MIL-125 (1) CoPc@NH ₂ -MIL-125 (2) CoPcPA@NH ₂ -MIL-125 (3)	2.47 2.42 2.54	120	(1): 85(MB); 19 (MO) (2): 79 (MB); 25 (MO) (3): 99 (MB); 18 (MO)	30 mg/L dye; pH = 7; conc. catalyst = 0.5 g/L; Visible light	Stable	[46]
RhB/Basic Violet 10	NH ₂ -UiO-66/BiOBr/Bi ₂ S ₃		60	92	20 mg/L dye; pH = 7; conc. catalyst = 0.5 g/L	stable	[52]
RhB/Basic Violet 10 CR/Direct Red 28	UiO-66 (1) UiO-66-NH ₂ (2) UiO-66-2M (3)	3.85 2.92 2.77	120	(1): 48 (RhB); 52 (CR) (2): 95 (RhB); 64 (CR) (3): 69 (RhB); 48 (CR)	10 mg/L dye; pH = 7; conc. catalyst = 0.4 g/L	stable (3)	[53]
BG/Green Basic 4	Cu ₄ (O) (OH) ₃ (bb) ₄ (NO ₃) _{30.3} H ₂ O 3CH ₃ CN] _n	2.32	120	96.8	40 mg/L dye; pH = 5; mass catalyst = 30 mg	-	[132]
MB/Basic Blue 9 RhB/Basic Violet 10	Ag ₃ VO ₄ @MIL-125-NH ₂ (1) Ag ₂ WO ₄ /MIL-125-NH ₂ (2)	2.27 2.57	60	(1): 75 (MB); 75.3 (RhB) (2): 65 (MB); 50.3 (RhB)	5 mg/L dye; pH = 7; conc. catalyst = 1 g/L; visible light	stable	[45]

Table 4. Cont.

Dye Common Name/Color Index	Sample	Band Gap (eV)	Time (min)	Degradation Efficiency (%)	Reaction Conditions	Stability	Ref.
RhB/Basic Violet 10	FCAU-17 (flakes) (Bi) (1) CAU-17 (rods) (Bi) (2)	3.75 3.83	80	78.6 (1) 96.1 (2)	5 mg/L dye; pH = 7; conc. catalyst = 0.1 g/L		[133]
MB/Basic Blue 9	amino acid-based carbon quantum dot/MIL-53 (Fe) binary composite	2.63	120	98	35 mg/L dye; pH = 9; conc. catalyst = 0.1 g/L; Xe lamp	stable	[81]
RhB/Basic Violet 10	NH ₂ -MIL-125 (Ti)/Ag/NiFe layered double hydroxide (Ti-MOF/Ag/NiFeLDH)		50	95	15 mg/L dye; pH = 7; conc. catalyst = 0.5 g/L; Xe lamp	stable	[134]
RhB/Basic Violet 10	MIL-125/g-C ₃ N ₄ /sodium alginate	-	120	99.41	310 mg/L dye; pH = 5; Xe lamp	stable	[82]
MB/Basic Blue 9	Ag@[Zn ₂ (bta)(bpy)(H ₂ O) ₂] 2H ₂ O] _n	2.86	160	86	10 mg/L dye; pH = 6; conc. catalyst = 1 g/L; Xe lamp	-	[77]
MB/Basic Blue 9	[Zn ₄ (tmlb) ₄ (H ₂ O)] _n (1) [Ni ₄ (tmlb) ₄ (H ₂ O)] _n (2) [Cu(tmlb)] _n (3) [Co(tmlb)] _n (4) [Pb(tmlb)(H ₂ O)] _n (5)	3.92 3.89 3.9 3.75 3.8	125	87.1 (1) 69.6 (2) 94.4 (3) 75.4 (4) 74.2 (5)	6 mg/L dye; pH = 6; conc. catalyst = 0.2 g/L; Hg lamp	stable	[110]
MV/Basic Violet 3	[[Co ₂ (bdc) ₂ (bmp) _{1.5}]-DMF·1.5H ₂ O] _n		40	68	10 mg/L dye; pH = 6; conc. catalyst = 0.8 g/L; Hg lamp	stable	[101]
MV/Basic Violet 3	[Co ₂ (μ ₂ -H ₂ O)(L)(2,2'-bipy) ₂] _n (1) [Co ₂ (μ ₂ -H ₂ O)(L)(phen) ₂] _n (2)	- -	40	69 (1) 61 (2)	10 mg/L dye; pH = 6; conc. catalyst = 0.8 g/L; UV lamp	-	[135]
AO7/Acid Orange 7 MB/Basic Blue 9 RhB/Basic Violet 10	Ag ₂ CrO ₄ /Cu (BDC)	2.92	100	98 (AO7) 68 (MB) 46 (RhB)	20 mg/L RhB; pH = 6; mass catalyst = 15 mg; Halogen lamp	stable	[99]
MB/Basic Blue 9	AmCoPc/UiO-66-NH ₂ amino cobalt phthalocyanine (AmCoPc)	2.21	180	80.17	10 mg/L RhB; pH = 6; conc. catalyst = 1 g/L; Xe lamp	stable	[136]
MB/Basic Blue 9	Ce/Co bimetallic MOFs with the mixed ligands	3.2	120	82.3	10 mg/L RhB; pH = 6; conc. catalyst = 1 g/L; Xe lamp	stable	[137]

Ponceau BS (PBS); acid orange 7 (AO7); Azure 1 (AzI); malachite green (MG).

3. Challenges, Perspectives, and Conclusions

Photocatalysis as a treatment method for the degradation/elimination of recalcitrant compounds of wastewater provides advantages compared to other methods, such as being a non-selective method, not generating sludges, and sometimes achieving complete dye mineralization. It is relatively inexpensive and has a simple configuration. Heterogeneous photocatalysis is understood to be the process that involves the joint use of light (light being understood as the entire electromagnetic spectrum) and a solid inorganic material called a semiconductor. Thus, employing MOFs (new MOFs, combination of ligands, and modification with other semiconductors) for the degradation of dyes offers advantages such as faster degradation, separation of the photo-generated species (electrons and holes) to make the degradation process more efficient, and providing stability to the MOF, thus increasing its useful life in the photocatalytic process. The dyes are relatively straightforward to characterize in the MOF reactions, as they can break the chromophore group in the dyes. The primary degradation mechanism for these dyes involves the generation of hydroxyl radicals. Those modifications to the MOFs have been explored and have shown promising results in treating recalcitrant compounds, some of which are persistent and considered carcinogenic in some instances.

In this study, we observed that the research on dye degradation using MOFs focuses on two key aspects. Firstly, the quest for new MOFs that allow working in the spectrum's visible region and, perhaps most importantly, their stability in aqueous environments. Despite the hundreds or even thousands of studies conducted in recent years, many MOFs are unstable in aqueous media, as exemplified by HKUST (CuBTC) or MOF-5 (Zn). This limitation hinders their use in the photocatalytic degradation of dyes in wastewater. Hence, recent advancements are concentrated on synthesizing MOFs with favorable physicochemical and optical properties and good stability. As shown in Table 4, recent research primarily focuses on two axes: the synthesis of new MOFs with complex ligands and the synthesis of composite materials. Both approaches aim to enhance MOF stability and improve its optical properties, such as the band gap and the recombination of charge carriers. While the research results over the past five years have been significant, we believe that using complex ligands for MOF synthesis may significantly limit their practical application as photocatalysts for dye removal. This limitation arises from the challenges in obtaining these ligands due to specific synthesis conditions or their high acquisition costs. Therefore, we suggest that research should be directed toward developing new MOFs composed of simpler ligands, such as terephthalic or trimesic acid.

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