

Review

# Recent Developments and Emerging Trends in Paint Industry Wastewater Treatment Methods

Nicolette Viktorová, Agneša Szarka and Svetlana Hrouzková \* 

Institute of Analytical Chemistry, Faculty of Chemical and Food Technology, Slovak University of Technology in Bratislava, Radlinského 9, 812 37 Bratislava, Slovakia

\* Correspondence: svetlana.hrouzkova@stuba.sk; Tel.: +421-915-396-928

**Featured Application:** The paper provides a review of the recently used wastewater treatment methods for paint industrial wastewater. The parameters affecting the effectiveness of the treatment process are discussed and recommendations for future improvements are given.

**Abstract:** High amounts of industrial wastewater are generated by the ever-growing demand and production of paint and coating materials. These effluents have negative effects on human health and the environment. The source of industrial effluents highly influences the properties, composition, and content of pollutants. The manufacturing of paint and coatings uses huge volumes of water and chemical reagents, consequently producing huge volumes of heavily polluted wastewater. This review is focused on summarizing various methods of industrial wastewater treatment from the paint manufacturing industry. Current trends in paint industry wastewater treatment processes have resulted in high efficiency of the reduction of chemical oxygen demand. Factors affecting the treatment processes are discussed and future trends are outlined. The effectiveness of the recently used methods is compared and the limitations of advanced treatment systems are highlighted. The review of recent developments in paint industry wastewater treatments points to the need for paying great attention to advanced analytical methods allowing the identification of individual contaminants to guarantee safe disposal limits.

**Keywords:** paint industry; purification methods; industrial wastewater; water treatment



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## 1. Introduction

Industrial wastewater is created during different types and stages of manufacture or as a by-product of instrument cleaning in factories, such as rinsing sludge, manure, or as aqueous waste from the engineering industry [1]. Industrial wastewater causes significant water pollution due to entering surface water (streams, lakes, rivers) and underground reservoirs. Chemicals, heavy metals, and microorganisms from industrial wastewater are toxic to aquatic organisms reducing organisms' life spans, abilities to reproduce and participate in the food chain. With the growing population and industrialization, global awareness has shifted towards exploring substitute methods for reducing water consumption [2]. Therefore, every effort must be made to reduce water consumption and treat wastewater to be reusable or safer for discharge into the environment.

Industrial wastewater is characterized not only by the containment of grease and oils, but also by high turbidity, high levels of chemical oxygen demand (COD) and biochemical oxygen demand (BOD), and suspended and dissolved solids. These high amounts must be reduced or eliminated before discarding the wastewater into water bodies. The main characteristics of industrial wastewater are classified into physical, chemical, and biological categories (Figure 1.). The treatment is a demanding, multi-step process with the primary purpose to reduce COD and BOD levels by removing as many contaminants as possible. Conventional industrial water treatment does not remove sufficient amounts of pollutants

entering watercourses; thus, industrial wastewater is considered potentially hazardous to freshwater ecosystems [3].

<b>Characteristics of Industrial Wastewater</b>		
<b>Physical Characteristics</b>	<b>Chemical Characteristics</b>	<b>Biological Characteristics</b>
<ul style="list-style-type: none"> <li>• <b>Color</b> – qualitative character</li> </ul>	<ul style="list-style-type: none"> <li>• <b>Inorganic chemicals</b> – nitrates, nitrites, phosphorous, heavy metals</li> </ul>	<ul style="list-style-type: none"> <li>• <b>Water borne disease for public health</b> - viruses, bacteria, protozoans, and helminths</li> </ul>
<ul style="list-style-type: none"> <li>• <b>Odor</b> - produced due to the decomposition of organic matter</li> </ul>	<ul style="list-style-type: none"> <li>• <b>Organic chemicals</b> - determines with BOD, COD and TOC</li> </ul>	<ul style="list-style-type: none"> <li>• <b>Biological processes</b> – bioaccumulation or eutrophication from nutrient additions</li> </ul>
<ul style="list-style-type: none"> <li>• <b>Turbidity</b> – by colloidal material or suspended solids</li> </ul>	<ul style="list-style-type: none"> <li>• <b>Volatile organic compounds</b> - such as benzene, xylenes, trichloroethane, dichloromethane, etc.</li> </ul>	
<ul style="list-style-type: none"> <li>• <b>Temperature</b> - higher than the normal water</li> </ul>		
<ul style="list-style-type: none"> <li>• <b>Solids</b> - soluble or insoluble organic and inorganic matter</li> </ul>		

**Figure 1.** The characteristics of industrial wastewater.

High amounts of wastewater are created by the paint-producing industries. These factories use large volumes of water, about 284–321 million liters per day, of which only about 4% is recycled [4]. The chemical content of the paint industrial wastewater varies widely and is related to the individual industrial units and the manufacturing processes [5]. The main stream of wastewater from paint industries is generated from equipment cleaning and represents around 80% of the effluents [6]. These effluents are toxic not only to the environment but can also have adverse effects on human health. The effluents contain diluted paint, which can irritate eyes, and skin or can cause headaches. The toxins present in paint industrial wastewater can also contribute to respiratory problems, muscle weakness, and liver and kidney damage. Heavy metals also pose many health risks [7]. Toxic heavy metals discharged into the environment can cause detrimental health problems in animals and ultimately humans via the food chain. They can be teratogenic and carcinogenic, and can cause oxidative stress, organ damage, nervous system impairments, and reduced growth and development [8].

The presented paper aims to review and discuss the recent trends in paint industrial wastewater treatment methods used to adequately clean wastewater. The main part of the review is classified into subchapters according to the various treatment techniques. The next part is devoted to highlighting future trends in this area.

## 2. Wastewater from the Paint Industry

### 2.1. Paint

Approximately 70% of the paint industry wastewater is discharged untreated into natural river basins [4]. All paint types contain chromophores, which are responsible for the color of the paint. However, paints differ in terms of chemical structure, process of dyeing, solubility, and the field of application. Most paints consist of similar components, a mixture of pigments, which are suspended solids, in a vehicle (liquid medium), a volatile solvent, a binder, which is a polymeric material, extenders, and suitable additives [9–11] as shown in Figure 2.

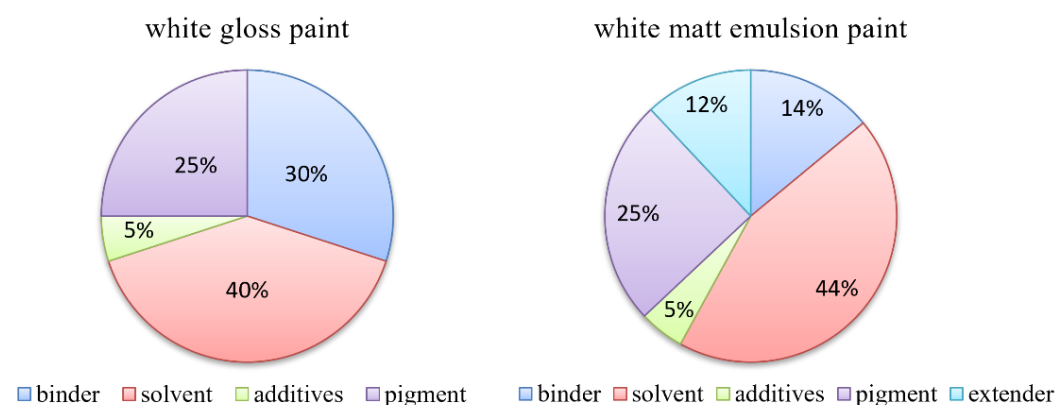


Figure 2. General paint composition [12].

The pigments are natural or synthetic compounds and can be classified by the production methods, their chemical structure, or their applications [13,14]. The pigment is a natural or artificial powdered color substance (organic or inorganic) dissolved in water, alcohol, or oils that colors the substrate. The neutral, most often white mineral pigments are made from barite, chalk, kaolin, or gypsum. Organic pigments are either natural, from plant or animal sources (e. g. saffron flowers, Brazilian purple, carmine), or produced synthetically. Synthetic organic pigments, such as dioxazine, beta naphthol, azo and phthalocyanine, are hydrocarbon compounds made from petroleum, base, and chemicals, primarily by exposure to high temperatures or pressure. Many synthetic organic pigments, especially the azo pigments, are derived from water soluble dyes. Dyes on their own bind chemically to materials in a way that prevents them from being edited after they are applied. Pigment grain size can vary from very small, up to 1  $\mu\text{m}$ , to large, with over 10  $\mu\text{m}$ . For example, zinc oxide, zinc sulfide, lithopone, and titanium dioxide are the components of basic white pigments, black pigment contains elemental carbon. The color of red pigments is made up of minerals such as iron oxide, cadmium, cuprous oxide, and various synthetic organic pigments [15,16].

The binding of the pigments is carried out by the binder or vehicle, but also oils, proteins, waxes, resins, or polymers, dissolved in a diluent that dries to a hard film are used [17]. Diluents applied in the paint industry are either organic solvents or water, depending on the consumer needs.

For saving, binder extenders are used, which are larger pigment particles. These particles improve the adhesion and strengthen the film. The physicochemical properties of the paints are adjusted by using different fillers and additives, such as chalk, gypsum, driers, stabilizers, bactericides, fungicides, etc. [12].

## 2.2. Paint Industrial Wastewater

The properties of the product are related to the type and ratio of the paint components. On the other hand, they have an impact on the characteristics of the wastewater produced by the paint industry [18]. Typical pollutants found in paint industry wastewater include those exerting high values of suspended solids (SS), COD and BOD, heavy metals, and toxic compounds [19]. Organic pollutants have recently attracted public attention, not only as they significantly degrade water quality, but also present major challenges to existing water treatment systems regarding their removal efficiency [20].

As mentioned before, around 80% of paint industry effluents come from equipment rinsing [10,21,22], so after the treatment, effluent, which is now diluted paint, may be used in manufacturing processes as a component of low-cost paints, or as a coolant or dilutant [23].

Microbial infection is typical for water-based paints because they contain inorganic and organic materials. The microbial infection occurs during the manufacturing stages of paint production and storage [24]. The changes in various kinds of physical and chemical

properties, such as high temperatures and humidity, increase the possibility of microbial contamination in paint [24]. Therefore, wastewater created during manufacturing can be highly contaminated with microorganisms. Wastewater generated during the cleaning of paint manufacturing equipment might be highly contaminated with not only organic compounds and pigments but also microorganisms [25].

The presence of any component from paint manufacturing in water is unacceptable. Untreated wastewater effluents with a significant number of dyes are considered potentially hazardous to organisms living in freshwater ecosystems. Ecological and environmental problems are caused by the discharging of these effluents directly into water sources. The contaminants included in wastewater can restrict the photosynthesis process of aquatic plants by blocking sunlight entry or poisoning the animal biota [26–28]. Therefore, sufficient treatment of paint industry effluents is important, and new ecological techniques are needed to further remove color and microorganisms present and reduce the levels of COD and BOD [29].

### 3. Paint Industry Wastewater Treatment Methods

Paint industry wastewater contains various organic solvents, pigments, surfactants, and other chemicals but also solid particles, heavy metals, oils, etc. Therefore, treatment before discharge into the environment is needed to suit the prescribed standards.

It was reported that many paint manufacturing industries discharge effluents without treatment; even though wastewater treatment plants are inefficient in bringing wastewater conditions to environmentally acceptable safe limits [30]. Industrial wastewater treatment techniques may vary depending on their composition, origin, and the specific company.

It has been shown that many paint industries, especially small-scale ones are not able to sufficiently treat the effluents before their discarding into the environment because they do not have suitable treatment systems [9]. On the other hand, the empirical data indicates that despite the installation of treatment systems in the paint industries a high level of pollution is still released into the environment. In addition to COD removal, minimizing sludge production from the coagulation process is crucial [9].

The recently used treatment techniques for the paint industry effluents can be divided according to the treatment's characteristic into physicochemical, biological, and electrochemical treatment and advanced oxidation processes (Figure 3).

The commonly used methods include coagulation, adsorption [29], flocculation, filtration, electrochemical processes [30], advanced oxidation processes, and biological processes such as composting [31–33].

Nowadays, a combination of different effective treatment methods is also used to safely dispose of or, recycle the treated wastewater [32,34].

The treatment of the paint industry effluents can be separated into four stages: preliminary, primary, secondary, and tertiary treatment (Figure 4). During the preliminary treatment neutralization and equalization take place [35]. The primary treatment includes simple and traditional techniques, such the sedimentation, chemical coagulation, flocculation, magnetic separation, and flotation. Physicochemical separation or biological oxidation is applied during the secondary treatment for the reduction of the organic compounds. Tertiary treatment is used for improving effluent treatment [36]. Some reviews of treatment methods have also shown the application of different molecular imprinted polymers for the elimination of heavy metal ions in everyday toxic paints [37].

#### 3.1. Physicochemical treatment

Physicochemical treatment methods are the most common techniques for paint industrial wastewater treatment. Table 1 contains recently used methods, such as sedimentation, filtration, adsorption, ion exchange, coagulation–flocculation, and oxidation primarily focusing on dividing the colloidal and suspended particles.

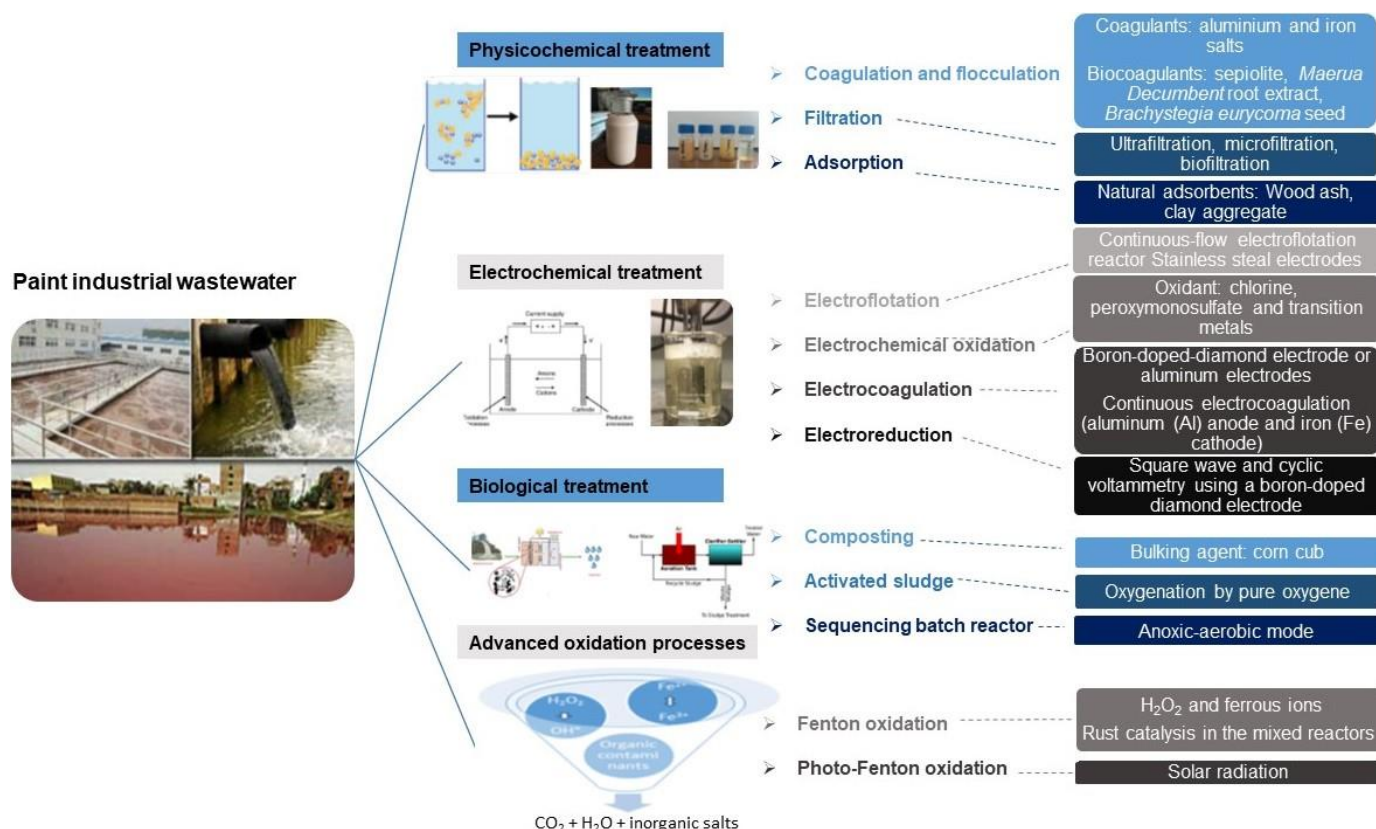


Figure 3. Distribution of industrial wastewater treatment methods according to treatment mechanism.



Figure 4. Hierarchy of stages involved in a typical industrial wastewater treatment plant.

### 3.1.1. Coagulation and Flocculation

Coagulation methods have always attracted attention for their high efficiency in removing primarily turbidity and color, but also organic matter and SS from wastewater. Coagulation–flocculation is a common treatment method for wastewaters containing dyes, paint particles, and SS [46–48]. The process of water and wastewater treatment is based on forming flocks that are settled down during sedimentation [30]. Therefore, the dye molecules and toxic aromatic compounds can be eliminated from the effluent without decomposition [49]. The amount of coagulant and adequate pH of the wastewater affects the quality of treated wastewater; therefore, these are important factors in the coagulation process (each coagulant requires a specific pH range for the coagulation process).

**Table 1.** Physicochemical treatment methods for paint industry wastewater.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Coagulation–floculation	FeCl <sub>3</sub> at pH 5.9, filtration	COD removal 40–59%	Water-based paint and allied products industry	[38]
Coagulation–floculation	FeCl <sub>3</sub> , FeSO <sub>4</sub> , and PACl pH 4.5–10	COD removal 88% SS removal: 100% Color removal 46–72%	Paint industry wastewater	[32]
Coagulation–floculation aeration process filtration	NaOH and alum as coagulant agents followed by lamella clarification process, aeration process (1–2 h) and palm hemp filtration process	Removal of COD 85% TSS 91% BOD up to 90%	Different paint factory industrial wastewater	[10]
Coagulation–floculation	2 coagulants Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> and FeCl <sub>3</sub> 2 flocculants: anionic and cationic optimal pH 5.3	BOD and COD reduction above 95%	Wastewater from the main collector of a local factory of painted steel tiles	[39]
Coagulation–floculation disinfection with sodium hypochlorite	Filtration coagulant—PAC flocculant—anionic PAM, pH = 7.3	BOD = 78.6% COD = 90.6% TSS = 99.9% TDS = 65.0% aluminum = 99.9%	Wastewater reservoir of a production plant (Latex paint company)	[40]
Coagulation–floculation	FeCl <sub>3</sub> and several natural-based materials, namely, limestone, pumice, sepiolite, bentonite, and mussel shell were used as flocculant aids	Most of the SS removed sepiolite—highest COD reduction 80%	Wastewater of paint and construction chemicals producing factory in Turkey	[41]
Coagulation–floculation	3 coagulants: FeSO <sub>4</sub> , FeCl <sub>3</sub> and Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> , optimal pH = 7.54 mixing 100 rpm 5 min, followed by slow mixing 20 rpm 30 min, settling time 30 min	The coagulant FeCl <sub>3</sub> provided the best treatment efficiency under optimized conditions removal of COD 94.1%, TSS 95.3%, color 97.1%, and turbidity 99.5%	Water-based paint wastewater from paint manufacturing plant, from washing tanks and mixing equipment	[42]
Coagulation–floculation natural coagulant Combined with electrolysis	Natural coagulant— <i>Moringa oleifera</i> fortified with Ca <sup>2+</sup> (CaCl <sub>2</sub> ) rotation 100 rpm, 1 min, slow mixing 20 min, 40 rpm 30 min sedimentation optimal pH 6.5 coagulant dosage = 80 mL/L BDD electrode	COD removal 41% turbidity removal > 99% After 1.5 h electrolysis, COD removal of wastewaters previously treated with MOAE/CaCl <sub>2</sub> and MOAE/Ca(NO <sub>3</sub> ) <sub>2</sub> was 70% and 75%, respectively	Paint manufacturing water-based paint wastewater collected directly from the plant's wastewater reservoir	[35]
Coagulation–floculation	Natural coagulant <i>Maerua decumbent</i> roots, aluminum sulphate pre-set doses of <i>Maerua decumbent</i> and alum coagulant rapid mixing (3 min at 180 rpm), slow mixing (30 min of at 20 rpm), 20–60 min of settling	Removal of turbidity 99.2%, COD 78.6%	Paint industry located in industrial area	[11]

Table 1. Cont.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Coagulation–flocculation using	<i>Brachystegia eurycoma</i> (seed) pH 8 and coagulation temperature of 35 °C	Process efficiency 96.50% at coagulant dosage of 5 g/L	Collected at batch production wash-off from a paint factory	[43]
Adsorption using	Adsorbent wood ash pH = 2, contact time 3 h, 100 g/L wood ash	Pb removal efficiency—96.1% maximum Co removal efficiency—99%	From industry equalization tank laboratory	[44]
Adsorption with	Adsorbent light expanded clay aggregate pH = 7 and exposure to 10 g/L of light expanded clay aggregate	Removal efficiency for Pb 93.75%, Cd, nearly 89.7%	Samples were taken from industry equalization tank effluent	[45]

Abbreviations: BDD electrode- boron-doped diamond electrode, BOD—biochemical oxygen demand, CFU—colony forming unit, COD—chemical oxygen demand, PAC—aluminum polychloride, PAM—polyacrylamide, TDS—total dissolved salts, TSS—total suspended solids.

Typically, chemical coagulants are widely used, especially hydrolysable metal salts based on aluminum or iron [29,42,50]. Generally, inorganic coagulants, such as aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ ), ferrous sulfate ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) [5], ferric chloride ( $\text{FeCl}_3$ ), and polyaluminum chloride (PACl) [31] have been applied during coagulation to reduce dye and SS levels [51].

Adequate pH is an important parameter for the coagulation–flocculation treatment. The COD removal was estimated in effluents from a manufacture of water-based paints using different coagulants such as  $\text{FeCl}_3 \cdot \text{H}_2\text{O}$ , sodium bentonite,  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , and  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ . It was found that the most useful coagulant used was  $\text{FeCl}_3$  at a wastewater pH of 5.9. However, this method only decreased the COD by 40–59% and was found inefficient and additional biological treatment was required [41]. On the other hand, in another study [31] applying coagulation–flocculation treatment using coagulants  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ ,  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ , PACl, and  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  at different wastewater pH levels (range of 4.5–10) showed up to 88% COD removal, 100% removal of SS, and color removal of 46–72% from paint industry wastewater with the best values at pH 8 for  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$  and  $\text{FeSO}_4$  and 8.5 for  $\text{FeCl}_3$  [29]. Hazourli et al. [38] reported sufficient COD and BOD removal using coagulants  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{FeCl}_3$  with anionic and cationic flocculants for the pre-treatment of steel painting factory effluents by the coagulation–flocculation method at the wastewater’s pH. The optimal dosage of 700 mg/L was estimated using a cationic flocculant, reducing BOD and COD values by over 95% [38].

Volume and concentration of the added coagulants are also important for the efficiency of the treatment process. In some cases, the addition lower doses of coagulants ( $\text{Al}_2(\text{SO}_4)_3$ ,  $\text{FeSO}_4$ , PACl, and  $\text{FeCl}_3$ ), under 500 mg/L, a higher removal efficiency has been achieved [31].

For the optimization of the treatment processes a jar test can be applied, which has been used for the comparison of coagulant efficiencies,  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , and  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  [49]. The best treatment efficiency was provided by the coagulant  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ; hence, optimization experiments were carried out with  $\text{FeCl}_3$ . The treatment conditions were optimized by using a central composite design. The dependent variables were COD, TSS, color, and turbidity in the optimization experiments for wastewater treatment. Treatment conditions were optimized using the design expert program. The optimum treatment parameters were reported as coagulant amount 1080.49 mg/L, pH 7.54, and mixing speed 146.16 rpm. Under optimized conditions, the treatment process was 4% more efficient than the non-optimized process. The COD, TSS, color, and turbidity removal were 94.1%, 95.3%, 97.1%, and 99.5%, respectively [49]. Other coagulant agents, NaOH and  $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ , have been studied for the coagulation–flocculation treatment method for paint industry wastewater, followed by the lamella clarification, aeration, and palm hemp

filtration processes [8]. Results indicated the removal of COD 85%, TSS 91%, and BOD up to 90% using physicochemical treatment [10].

For latex paint industry wastewater, which is characterized by the high content of SS, bacteria, and COD, effective coagulant flocculants were aluminum polychloride and polyacrylamide. The treated water fulfilled the quality standards (COD removal of around 90%, TSS removal of almost 100%) and was proven usable for the production of different acrylic latex paints after disinfection with sodium hypochlorite (NaClO), reusing about 56% of the produced wastewater [39].

Bio-coagulants such as seeds, leaves, grains, and fruits of plants can enhance the coagulation process, mainly working by adsorption mechanisms, polymer bridging, and neutralizing charge, making the smaller suspended particles settle and collectible [40]. Bio-coagulants have lots of benefits such as cost-effectiveness [52], low sludge volume, ability to keep the pH constant, creation of toxin-free treated water, and biodegradability [23]. Natural coagulants and flocculants have a typically higher ability for making the flocs, therefore, a reduced amount is needed for the treatment process, which means their use can reduce the cost of the coagulation–flocculation process. Several natural-based materials have been tested as flocculants. Limestone, pumice, sepiolite, bentonite, and mussel shell have been used in combination with  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  as flocculant aids, providing sufficient SS removal [51]. The use of the natural materials separately was reported to perform inadequately for dye removal. The combination of coagulant with sepiolite as a natural flocculant presented the highest reduction of COD (80%) in wastewater of paint and construction chemicals produced by a factory in Turkey [51].

The use of bio-coagulants, which are eco-friendly, in treatment processes is desirable because they are environmentally friendly and safe for human health. However, the optimization of the treatment process parameters is required for their efficient use in the treatment of industrial wastewater. One of the bio-coagulants effective for the coagulation–flocculation processes, *Maerua Decumbent* root extract has been used and the treatment process optimized [11]. The *Maerua Decumbent* powder with several types of heteroatomic functional groups is useful for the removal of pollutants from wastewater. It is an eco-friendly and effective alternative to conventional coagulants. In optimum conditions, *Maerua* root was reported to achieve the removal of turbidity by 99.2% and COD removal by 78.6%, which compared favorably with 98.6% turbidity and 66.2% COD removal using  $\text{Al}_2(\text{SO}_4)_3$ . Additionally, lead and chromium were removed from the wastewater by 100 and 99.97%, respectively [11].

For comparison, another natural coagulant, aqueous extract of *Moringa oleifera* seeds (MOAE), fortified with  $\text{Ca}^{2+}$  ( $\text{CaCl}_2$  and  $\text{Ca}(\text{NO}_3)_2$ ), has been used for water-based paint wastewater treatment optimization combined with electrolysis. Results after applying the coagulation–flocculation showed COD removal of 41% and turbidity removal of over 99%. The COD removal increased after 1.5 h of electrolysis for MOAE/ $\text{CaCl}_2$  to 70% and for MOAE/ $\text{Ca}(\text{NO}_3)_2$  to 75% [34], making this coagulant more efficient [11,34]. Furthermore, in the coagulation–flocculation treatment of paint wastewater, the adsorption capacity of bio-coagulant *Brachystegia eurycoma* seed was noted [53] to achieve an even higher COD treatment efficiency of 78.6%.

However, the coagulation process is not useful for the complete removal of pollutants from paint industry effluents. One of the drawbacks of this method is the dependence of the flocs dimension on temperature, small flocks are created at very low temperatures [19]. In addition, the method is not useful for the removal of microorganisms; therefore, microorganisms are still present in the effluent and can continue to grow. Using activated carbon to remove inorganic pollutants from water is a widely extended technique because of its high surface area, microporous character, and chemical nature of its surface [43]. After coagulation, the filtration of the suspended particles is needed to separate them from the liquid through a membrane or filter. Various filtration techniques (ultrafiltration, membrane filtration, microfiltration, and biofiltration) are used [10,54]. Coagulation is a simple and useful method for the elimination of the suspended particles but the chemi-



cals present in paint industry effluents can precipitate in their toxic form; therefore, high amounts of sludge can be produced which cannot be eliminated by filtration. Furthermore, the efficiency of the treatment process depends on the pH of the wastewater. It has been shown that COD removal is higher when the pH of the industrial effluent is adjusted to be alkaline. On the other hand, at the original pH of the effluent (pH 6), a higher dosage of the coagulants is needed.

### 3.1.2. Adsorption

Adsorption is also a frequently applied method for treating paint industry effluents. Heavy metals are removed from the wastewater by many different adsorbents, both chemical and natural [44,45,53,55]. In recent years, alternative adsorbents are produced, which have the ability to bind and remove heavy metals from the effluent. Low-cost materials have been tested for this purpose, such as natural zeolite, ash, rice husk, vermicompost, peat, volcanic stones, bentonite, and clinoptilolite. From these natural adsorbents, wood ash has been applied for the paint industrial treatment process. Heavy metal removal (Co and Pb) by wood ash as an adsorbent has been studied [55] and found to be easily obtainable and inexpensive. Heavy metals were reported to be effectively removed from paint industry effluents using wood ash as an adsorbent. The treatment process is influenced by the pH and the adsorbent quantity. The maximum removal efficiency was 96.1% and 99% for lead and cobalt, respectively.

Less satisfactory removal efficiency was obtained using light-expanded clay aggregate (LECA) as an adsorbent for the removal of Cd and Pb from paint industry effluents under various conditions, such as the amount of adsorbent, contact time, and pH. The removal efficiency of clay adsorbent was reported to be 93.75% for Pb and nearly 89.7% for Cd [44].

Adsorption is a proper method for the elimination of heavy metals from the wastewater effluents, meaning that for the removal of organic pollutants, dyes, and other toxic compounds the combination of other treatment processes is needed.

### 3.2. Biological Treatment

Biological treatment is another approach for the removal of paint from industrial effluents by utilizing microorganisms to treat or engineer wastewater. Pollutants produced during paint manufacture in wastewater are very difficult to eliminate biologically [56]. Usually, during the biological treatment process, an emulsion is produced which has to be separated. The concentrated paint can be reused as a thickener in the paint industry. Mostly, a single population of a microorganism dominates the effluent and this population is responsible for the rottenness of wastewater when stored for a long period of time [29]. Before discharging the effluents into biological treatment systems, the concentrated effluents have to be treated [22].

There have been some biological treatment methods (Table 2) explored for the paint industry wastewater, such as composting [32], sequencing batch reactor treatment [22], and activated sludge processes [57]. Some recent reviews also feature the biological removal of azo dyes from industrial effluents [56] and different isolation techniques by extraction for the elimination of biopolymers in wastewater [58].

Studies have shown that using composting processes for the effluents from the automotive industry using water-based paints can be used to minimize sludge in reactors by 85% using corncob as a bulking agent [32]. A pure oxygen-based activated sludge system was reported to be effective in the removal of COD and reducing other characteristic parameters in a mixture of sewage and paint wastewater; 87.8–93.6% COD and 97.7–99.2% BOD removal was obtained. The quality of the resulting wastewater was suitable according to the recommended pollution standards [56].

For the controlling of the biological treatment processes, a designed lab-scale sequencing batch reactor has been used (wastewater from a bus manufacturing factory). The study reported the removal efficiency of carbon and nitrogen to regress to levels of mainstream wastewater mixed with paint wastewater using the sequencing batch reactor, and the

inhibition of growth kinetics to 21% and hydrolysis kinetics to 33%. The effluents met the in-sewer discharge limits for COD [22].

**Table 2.** Biological treatment methods for paint industry wastewater.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Composting of paint sludge	Reactors with temperature 40 °C for a minimum of 5 days	85% of paint sludge bio-dried using corncob as bulking agent	Water-based paint sludge originating from the automotive industry	[32]
Pure oxygen-based activated sludge system	Oxygen flow rate maintained at 5.0–8.0 mL/L	87.8–93.6% COD, and 97.7–99.2% of BOD removal	Wastewater from the paint industry effluent and sewage	[57]
Sequencing batch reactor chemical pre-treatment	Volume 14 L, cycle run time 24 h followed by 2 h settling, decanting and idle period, temperature 20 ± 1 °C, pH = 6.5–7.5	Effluents met the in-sewer discharge limits for COD	Bus production factory located in Istanbul (Turkey), from coating process, treated with domestic wastewater in the existing treatment plant (after chemical pre-treatment)	[22]

Abbreviations: BOD—biochemical oxygen demand, COD—chemical oxygen demand.

### 3.3. Electrochemical Treatment

The high cost of physical and physicochemical pretreatments and the hard biodegradability of paint industry pollutants led to the development of electrochemical treatment processes, from which electrochemical oxidation is becoming a widely used alternative to these techniques. During the electrochemical treatment process, an electric current is applied and coagulation of the particles takes place by the transfer of electrons. Different electrochemical processes have been studied for paint industry wastewater purification, including electroflotation, electrochemical oxidation, electro-reduction, and electrocoagulation (Table 3). Electrochemical treatment methods have high electricity consumption because of the formation of an oxide film which makes them unsustainable [56,59].

#### 3.3.1. Electroflotation

The first use of electroflotation was in mineral processing, but the advantages of the process showed its effectiveness in the field of wastewater treatment. During the process, thanks to electrolysis, bubbles are formed and effectively used for the removal of suspended particles by flotation [60]. The main advantages of this method over flotation are in the size of the produced bubbles (smaller), the higher surface area, and the suitability for the removal of finer particles. Electroflotation has been used as a method for the removal of synthetically prepared paint from automotive paint systems [60], with NaOH addition for adjusting the pH of the water. TSS removal of 90.39 to 97.43% was achieved [60].

The continuous flow electroflotation method has also been developed [61] for the design and application of apparatus with stainless-steel electrodes for the treatment of paint wastewater from the car coating industry regarding the key variables, e.g., initial solids concentration, applied current density, and retention time. The system efficiency is reduced with higher concentrations of the suspended solid particles. On the other hand, there is a direct relationship with the current used for the treatment and the time of the whole process. The efficiency of TSS removal increased to 95 ± 7% when applying a higher current density and longer hydraulic time [61].

It should be concluded that the control and operation of the electroflotation systems are simple, and the construction is flexible because the electrodes can be designed based on the size and dimensions of the reaction tank. In electroflotation, suspended particles collide with gas bubbles, attachment occurs, and they slowly rise to the surface to be skimmed off. Each phase of this process is time dependent. Therefore, adequate retention time is needed for the collision and attachment of bubble–particle in the solid–water separation

process [61]. Furthermore, the efficiency of electroflotation treatment is also dependent on the pH of the effluent, such as in the case of coagulation.

**Table 3.** Electrochemical treatment methods for paint industry wastewater.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Electroflotation	NaOH added to mixture to adjust pH total testing time was 40 min	TSS removal from 90.39 to 97.43%	Automotive paint solvent-based auto paint (clear coat and primer) synthetically prepared in the laboratory	[60]
Continuous flow electroflotation	Stainless-steel electrodes	TSS removal rate $95 \pm 7\%$ under initial TS 500 mg/L, current density 100 A/m <sup>2</sup> , retention time 8 min	Automotive paint wastewater was synthetically prepared in the laboratory	[61]
Electrochemical oxidation	Batch-wise in the presence of NaCl electrolyte with carbon electrodes	Removal of COD—65.68%, color—98.74%, and turbidity—96.56%	Synthetic water-based paint wastewater	[62]
Indirect electrochemical oxidation	Reaction time = 1 h pH = 3–12 graphite electrodes NaCl = 1–3 g/L	COD removal 55%	Mixed wastewater from textile and chemical industry	[63]
Electrooxidation	Peroxydisulfate and transition metals (Fe <sup>2+</sup> , Cu <sup>2+</sup> , Zn <sup>2+</sup> ) graphite cathode different anode materials (Ti/IrO <sub>2</sub> , Ti/RuO <sub>2</sub> , Ti/SnO <sub>2</sub> )	COD and true color removal efficiencies as a result of validation studies 74.28%, 99.03%, respectively	Paint manufacturing industry wastewater	[22]
Electrooxidation	pH = 4, temperature 39.99 °C, NaCl concentration 100 mM, feed rate of 40 mL/min current 5.21 A	COD removal 80.95%. color removal 79.12% under optimum conditions	Paint manufacturing plant wastewater	[64]
Electrochemical treatment in tubular reactor	Batch-wise in the presence of NaCl electrolyte with carbon electrodes	COD removal 44.3% color removal 86.2% turbidity 87.1%	Synthetic water-based paint wastewater (acrylic copolymer water-based white primer)	[18]
Electrocoagulation	BDD electrode current dosage $i = 10 \text{ mA/cm}^2$ (90 min) coagulant: aluminum sulfate dosage 12 mL/L	12 mL/L of Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> TS decreased from 10.7 kg/m <sup>3</sup> to 0.37 kg/m <sup>3</sup> (97% removal TS) COD removal 85%	Wall paint manufacturing plant wastewater reservoir	[28]
Electrocoagulation with advanced electricity and flow control	Current density for batch process $i = 14.2 \text{ mA/cm}^2$ and for continuous process $i = 14.4 \text{ mA/cm}^2$	COD removal 69.7% in continuous flow reactor and 68.0% in batch process aluminum removal: 62.1% continuous process 79.8% batch process	Paint industry wash water from storage tank	[65]
Electrocoagulation	Reaction time = 1 h applied voltage = 1–2 V pH = 3–10 aluminum electrodes	COD removal 43–55%	Mixed wastewater from textile and chemical industry	[63]

Abbreviations: BDD electrode—boron-doped diamond electrode, BOD—biochemical oxygen demand, COD—chemical oxygen demand, kGy—kiloGray (1 kGy = 1000 J/kg), TS—total solids, TSS—total suspended solids.

### 3.3.2. Electrochemical Oxidation

The paint industrial wastewater is mostly treated by indirect electrochemical oxidation. During this process, a strong oxidizing agent, mostly chlorine, which is produced by the oxidation of chlorides, is electro-generated at an anode surface. After that, it is applied to destroy pollutants in the bulk solution [66,67]. The main advantage of indirect electrolysis is the prevention of electrode fouling, avoiding direct electron transfer between organics and the anode surface. It was reported by a batch study of electrochemical oxidation of synthetic water-based paint effluents that the technique could be used as an alternative to traditional physicochemical processes in treating water-based paint wastewater by using NaCl electrolytes with carbon electrodes. The COD removal of almost 66% and significant removal in color and turbidity (over 96%) were obtained. On the other hand, the process is less economic and requires supervision [62]. Similar results were obtained by a tubular reactor batch-wise study, using NaCl as an electrolyte [18]. Results showed a COD removal of 44.3%, color removal of 86.2%, and turbidity removal of 87.1% [18]. The addition of external chloride into the system improved the removal efficiency by up to 55% for COD and almost completely removed color within 1 hr. The addition of sodium chloride significantly increased the efficiency of both COD and paint particle elimination [63]. It has been shown that pH is also important during electrochemical oxidation. The COD and color removal of 80.95% and 79.12%, respectively, were obtained at pH 4 with NaCl as an electrolyte [64].

Peroxymonosulfate and transition metals have been tested for the electrochemical oxidation process of paint manufacturing industry wastewater [68]. The best results were reported to using  $\text{Fe}^{2+}$  as a catalyst. In validation studies this catalyst provided higher removal efficiency and lower costs. COD and true color removal efficiencies of 74.28% and 99.03%, respectively, were experimentally obtained [68].

It can be concluded that the addition of external chloride into the system can improve the efficiency of the electrochemical oxidation treatment process. The method is also a pH dependent process; in comparison with the above-mentioned techniques the acid pH adjustment is preferred.

### 3.3.3. Electrocoagulation

Electrocoagulation uses an electric current for the removal of SS from paint industry effluents. The aim of electrocoagulation is the removal of particles from wastewater by destabilizing/neutralizing the repulsive forces that keep the particles suspended in water. When the repulsive forces are neutralized, the suspended particles will form larger particles that can settle down for easier separation from water [69]. The current creates an extreme temperature, concentrated in the liquid, coagulating its contents. Different electrode materials have been studied for this purpose, such as boron-doped diamond electrodes (BDDE) [28] or aluminum electrodes [63]. Better results, according to the COD removal, were obtained for BDDE using  $\text{Al}_2(\text{SO}_4)_3$  as a coagulant to treat water-based acrylic wastewater from the paint industry [28]. COD removal by using the BDDE has been studied in the pH range between 4 and 7 using  $\text{Al}_2(\text{SO}_4)_3$  as a coagulant agent. The COD removal was 85% with a smaller dose of coagulant (97% removal TS) and treated water was appropriate for disposal. In the case of aluminum electrodes, COD was reduced in total by 55% and the color of the wastewater by 56% after 60 min of electrolysis [63]. On the other hand, the application of BDDE increased turbidity with a higher coagulant dose required due to saturation of the solution [28,70].

A comparison of electrocoagulation with monopolar and bipolar wiring performance (aluminum electrodes) showed that monopolar connection is more efficient at reducing the COD value by 55%, (bipolar connection only 43%) with an applied voltage of 1.5 V during 1 h of electrolysis [63].

A novel continuous electrocoagulation system has been developed and studied for the treatment of paint industry effluents. The system uses advanced electricity and flow control [65]. For both batch ( $i = 14.2 \text{ mA/cm}^2$ ) and continuous ( $i = 14.4 \text{ mA/cm}^2$ ) processes

a higher current density than in a previous study [28] was used causing a slight increase in wastewater pH levels, the pH of the original wastewater was reported at 6.6 and it did not significantly change during the electrocoagulation process (7.15 after the batch process and 7.0 after the continuous process). Comparison of the two parallel analyses of both processes, similar results were obtained. The average efficiency of chemical oxygen demand removal was 68% in batch and 69.7% in the continuous system [65].

The use of chemical coagulants/flocculants such as metal salts or polyelectrolytes is the main advantage of electrocoagulation over chemical coagulation/flocculation. The coagulants in electrocoagulation are generated in situ by the electrolytic oxidation of an appropriate anode material which results in much less sludge generation.

### 3.4. Treatment by Advanced Oxidation Processes

Advanced oxidation processes (AOPs) can efficiently decompose degradation-resistant organic waste and remove it from wastewater [71]. Some of these processes are, for example, Fenton's oxidation, photo-Fenton, ozonation, or photo-catalytic degradation [72]. These techniques have been found to be very effective tools for the treatment of toxic organic pollutants in dye- and heavy metal-containing effluents. Of the mentioned AOPs techniques, Fenton's oxidation and photo-Fenton processes have been studied for the treatment of paint industry effluents (Table 4) [73].

**Table 4.** AOPs treatment methods for paint industry wastewater.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Fenton's oxidation process using rust particles	10 g rust particles/L within 70 h	COD removal 80% rust materials practically not an efficient source of Fe ions	Grab from painting unit of Profilo Telra plant, manufacturing televisions	[74]
Fenton and photo-Fenton processes	All processes were carried out in pH 3.0 $15.15 \times 10^{-3}$ mol/L FeSO <sub>4</sub> 0.30 mol/L H <sub>2</sub> O <sub>2</sub> during 6 h	Photo-Fenton process assisted with solar radiation, reductions of 99.5 and 99.1% of COD and TOC levels photo-Fenton's is very efficient in treating alkyd resin using solar light compared to artificial source	Wastewaters generated during alkydic resins manufacture	[75]
Fenton's oxidation process	Sample volume 250 mL, sample temperature 24 °C, pH = 3 stirring for 5 min at 200 rpm and subsequently for 45 min at 20 rpm, supernatant was withdrawn, pH adjusted to 8.5–9 by Ca(OH) <sub>2</sub>	81% of COD removal sludge generated in the Fenton's process was not as much as in coagulation, hazardous chemical sludge clearance necessary	Treatment and recycling facilities of highly polluted water-based paint wastewater from electronics industry	[76]
Photocatalysis	Ternary photocatalyst containing iron nitride, carbon nitride, and hematite (Fe <sub>3</sub> N/Fe <sub>2</sub> O <sub>3</sub> /C <sub>3</sub> N <sub>4</sub> ) 0.04 g catalyst dosage, 5 ppm rhodamine B concentration, pH 3.5	Removal of rhodamine B solutions and CO <sub>2</sub> photoreduction 98% of the contaminant was removed over the photocatalyst in the aqueous phase	Rhodamine B solutions	[77]
Photocatalysis	Ag/AgCl@T-C <sub>3</sub> N <sub>4</sub> photocatalysts	Removal efficiency of 98% after repeated use 5x	Organic dye compound AB92	[78]

Abbreviations: BOD—biochemical oxygen demand, COD—chemical oxygen demand.

During the Fenton's oxidation process, a solution of H<sub>2</sub>O<sub>2</sub> and ferrous ions is applied for the oxidization of the contaminants in the wastewater. The process can be affected by the dose of H<sub>2</sub>O<sub>2</sub>, the pH, and the catalyst concentration. The treatment of water-based

paint wastewater using Fenton's oxidation process has been tested using different Fenton's catalysts and photo-Fenton's oxidation [74–76].

Different catalysts for Fenton's oxidation process were studied for enhancing the COD removal efficiency by estimating the effects of column dimension, size of packing material, the dosage of reactive, pH, and reaction time based on packed columns and mixed reactors [74]. Oxidized iron rust particles were used as the catalyst for the Fenton's process, enhancing the removal efficiency of COD to around 80%. Despite enhanced COD removal efficiency, using oxidized iron materials as catalysts is an impractical and inefficient source of Fe ions because low pH can cause the generation of dissolved metal ions, and the small particles of Fe can harm the rate of filtration [74].

The correlation has been studied between the effectiveness of Fenton's and photo-Fenton's oxidation and the application of sunlight and artificial radiation for the removal of COD, total organic carbon, and phenolic compounds from alkyd resin manufacturing wastewater. The best results were obtained with photo-Fenton's process using solar radiation, with 99.5% COD removal efficiency and 99.1% total organic carbon removal, whereas using artificial irradiation presented a less effective but still notable removal of 60–80% for both parameters. Application of photo-Fenton's process with artificial irradiation resulted in 95% removal of total phenols [75].

On the other hand, Fenton's oxidation has been used for coagulation and flocculation (using  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{FeSO}_4$ ) with membrane processes to treat and reuse extremely contaminated water-based paint effluents from the electronics industry [76]. The studied wastewater had very high levels of COD and SS, alkaline nature, and was black. The molar ratio of  $[\text{H}_2\text{O}_2]/[\text{Fe}^{+2}] = 10:1$  showed COD removal up to 81% and a lower sludge production was attained; however, hazardous chemical sludge purification was required. Membrane filtration enabled the reuse of filtrate from sludge created during coagulation [76].

A new ternary  $\text{Fe}_3\text{N}/\text{Fe}_2\text{O}_3/\text{C}_3\text{N}_4$  photocatalyst, prepared by thermal pyrolysis, was able to completely remove 5 ppm of dye rhodamine B solution under acidic conditions in less than 30 min [77]. Photocatalytic capacity for the removal of rhodamine B solutions and  $\text{CO}_2$  photoreduction was studied. More than 98% of the contaminants were removed with photocatalyst in the aqueous phase in under 30 min under optimized conditions (catalyst dosage 0.04 g, RhB concentration 5 ppm, pH 3.5) [77].

Photocatalysis has also been studied using  $\text{Ag}/\text{AgCl}@T\text{-C}_3\text{N}_4$  photocatalysts for dye removal [78]. The removal efficiency of an organic dye compound AB92 was 98% after repeated photocatalysis ( $5\times$ ) [78]. Both photocatalysts could be promising candidates for practical application, achieving the same removal rate.

Despite the advantages of AOPs, there are some disadvantages such as the stability of oxidants that leads to limitation in their storage and transport, in addition to the strict pH requirements of the system (2–4). Therefore, new strategies resolving the current issue have to be developed.

### 3.5. Combination of Treatment Processes

For higher COD removal from paint industry wastewater, a combination of the various treatment methods has proven to be successful [6,30,32,34] (Table 5). Regarding electrocoagulation improvement and obtaining an effluent of suitable quality for agricultural purposes, a combined method of radiation, coagulation, and adsorption with granular activated carbon has been used to treat wastewater from the paint industry [29].

For the decomposition of the pollutants short-lived reactive species, which are produced during the radiolysis of water should be used. Ionizing radiation is an effective tool for the degradation of numerous compounds and the inactivation of microorganisms. The effectivity of the process depends on the type of energy, dose rate, and absorbed dose [79–81]. El-Sawy et al. [29] studied the efficiency of the combined process of radiation and coagulation with  $\text{Al}_2(\text{SO}_4)_3$  and adsorption using granular activated carbon for paint industry effluents and its reuse for different purposes. Radiation at 2 kilograys (kGy)

proved to be sufficient for the complete elimination of Gram-positive bacteria. However, it was not enough to reduce COD and BOD values. Therefore, a higher radiation dose (10 kGy) was tested and a 60% COD and 80% BOD removal was obtained. However, the obtained values for COD and BOD were insufficient to meet the guideline for effluent discharge. For this reason, coagulation with  $Al_2(SO_4)_3$  was applied to improve the treated water quality. The combination improved COD removal by up to 92% and BOD removal by up to 98.5% [29]. Aggregates were removed by sedimentation and filtration, and analysis of sulfate ions from the coagulant were conducted by standard methods for water and wastewater treatment [29].

The coagulation–flocculation process in combination with a biological process, using an aerobic feed batch operation, has also been shown to be highly effective in the treatment of paint manufacturing effluents [27]. COD and BOD removal was 96% and 92.5%, respectively. In comparison with the method developed by El-Shazly et al. [10] (85% for COD and 90% for BOD), using a chemical/physical treatment followed by an aeration and filtration process through a palm hemp agro fiber filter media, the aerobic feed batch operation produced better results.

A multilevel contact oxidation system was designed [82] for automobile painting wastewater treatment. By producing activated sludge and optimizing the method conditions, the system could effectively decrease the COD of the paint industry effluents. Under steady process conditions, the final removal of COD was 84% and SS 82.5% [82].

Hybrid electro-thermochemical wastewater technology was studied for the treatment of paint industry wastewater [6]. Treatment of industrial wastewater was carried out with modifications of additives (organic and inorganic) during the treatment procedure. The effect of both additives on the pollutant removal efficiency was studied in two different potential settings resulting in a variety of treated water samples (Figure 5). Treatment efficiency was monitored using the six most representative analytes as markers. It was found that the removal of the selected organic pollutants, in some cases, was higher than 90%, indicating options for further technology upgrades [6].



Figure 5. The hybrid electro-thermochemical wastewater treatment technology [6].

Table 5. Combination treatment methods for paint industry wastewater.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Radiation in combination with coagulation and adsorption	Coagulant $Al_2(SO_4)_3$ 10 g/L, pH 8 adsorbent granular activated carbon absorbed radiation dose range 0.5–10 kGy	COD removal 92% BOD removal 98.5% efficient radiation dose 2 kGy to remove nearly 100% of bacteria 10 kGy for reduction of COD, BOD and TSS	Discharge of a paint factory	[29]

Table 5. Cont.

Treatment Method	Treatment Parameters	Results	Source of Wastewater	References
Combination of chemical coagulation-flocculation with aerobic biological process	FeCl <sub>3</sub> and coagulant aid centrifuged at 6000 rpm 25 min	96% of COD removal, 97% of color removal and 92.5% of BOD removal	Collected from equalization tank of a paint factory	[19]
Multilevel contact oxidation system	25 L active sludge, 30 L painting wastewater and 25 L clean water evenly mixed and then fed to biological tank with special biological stuffing total process time 30 days	Removal of COD 84% SS 82.5% with hydraulic retention time of 8 h	Painting wastewater pretreated by physicochemical process	[82]
Hybrid electro-thermochemical wastewater treatment technology	Additives—two types: organic, inorganic—added during the treatment procedure both additives were used for 2 different potential settings resulting in 4 treated water samples	Removal efficiency factor (REF) 90%	Water discharge of paint factory	[6]

Abbreviations: BOD—biochemical oxygen demand, COD—chemical oxygen demand, kGy—kiloGray (1 kGy = 1000 J/kg), REF—removal efficiency factor, TSS—total suspended solids.

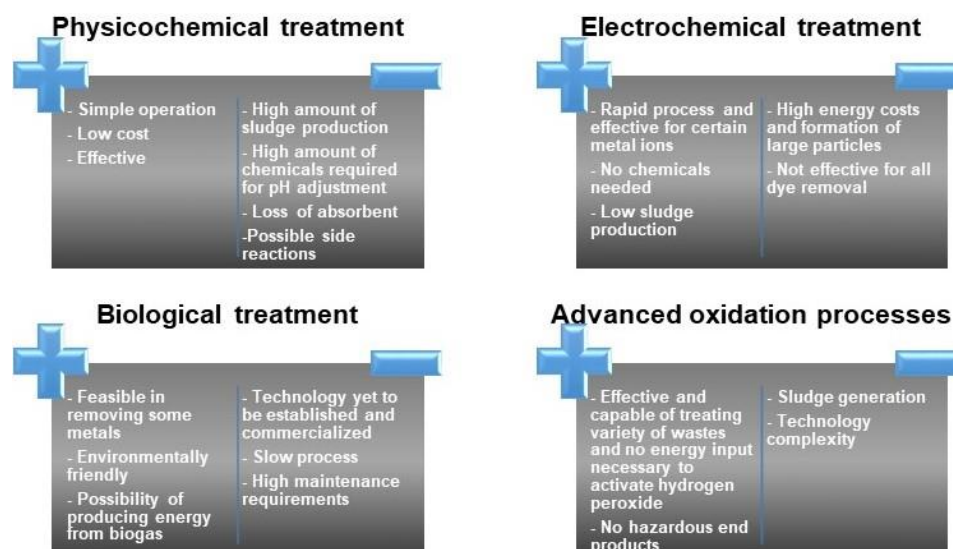
#### 4. Comparison of Techniques and Future Trends

In recent years, there has been an increasing demand for high-quality wastewater treatment; this coincides with stricter regulations regarding wastewater reuse. The main characteristics of the paint industrial wastewater are color, pH, high chemical oxygen demand, and low biodegradability. Suspended solids are the dominant contaminants that could be decreased by the impressive implementation of conventional treatment techniques in facilities. Traces amounts of heavy metals in pigments occur in industrial wastewater. Oil and grease are the other major contaminant indicators in the paint industry [83]. Different treatment techniques have been used for the treatment of paint industry wastewater and the main advantages and disadvantages of these techniques are summarized in Figure 6.

In the last decade intense efforts have been dedicated to developing additive chemistries that are more environmentally friendly. Therefore, the principles of green chemistry should be accepted also in paint industry effluent treatments. It should be concluded that mostly physicochemical methods are applied. In comparison with other techniques, these methods are more simple and cheaper. However, the effect of the amount and concentration of coagulant and the pH on the quality of wastewater treated is an important factor and must be optimized [30]. To eliminate the shortcomings of chemical coagulants, which are related to a high carbon footprint associated with their production, natural plant-based coagulants are beginning to be used, which represents a renewable, non-hazardous, degradable, potentially carbon-neutral option, to replace conventional coagulants [40].

The biological degradation of the organic pollutants included in paint industrial wastewater is very difficult. Therefore, biological methods are rarely used. However, biofiltration has been successfully applied for the treatment of different types of wastewaters and should be a promising biological alternative for paint industrial effluent treatment thanks to the advantages of a smaller footprint, shorter retention times, lower sludge production, and effective removal performances [84].





**Figure 6.** The main advantages and disadvantages of paint industrial wastewater treatment methods.

Under the current circumstances, advanced water treatment technologies are urgently needed to guarantee high-quality water, reduce chemical and biological contaminants, and strengthen industrial production processes. Modern wastewater treatment is based on the combination of simple physicochemical processes with electrochemical oxidation, ozonation, or advanced techniques. Nevertheless, future research should focus on optimizing the process and reducing costs. At the same time, an important issue that will concern final users is the disposal of the produced sludge, especially in light of the circular economy and minimizing the total cost. However, in processes such as electrochemical oxidation, there are various future research directions, such as the development of new materials with increased activity or the combination with other processes, where the existence of synergy can significantly increase efficiency and, therefore, the viability of the combined or hybrid processes [85].

It was previously mentioned that the paint industry effluents are mainly formed during washing equipment, so effluents are de facto diluted forms of paint. Therefore, separation of the components of the wastewater should be considered and the recovery of the substances, such as pigments, solvents, and heavy metals, upstream of the conventional wastewater treatments would reduce emissions into the environment.

To guarantee that the treated water meets the safe disposal limits, besides the traditional parameters, such as toxicity, TOC, COD, BOD, turbidity, etc., the individual identification of selected water contaminants in raw and treated wastewater should be realized through advanced analytical methods allowing the identification of individual contaminants, particularly by separation and spectrometric detection.

## 5. Conclusions

Wastewater from the paint industry is highly polluted and requires additional attention in terms of cleaning methodologies and techniques. Industrial companies produce large volumes of wastewater during the production of dyes, resulting in new cleaning methods, including physicochemical and biological methods, which have constantly been developed in recent decades. Nevertheless, these processes are insufficient for the effective treatment of paint industry wastewater due to its different composition and toxicity. This paper gives an overview of the development of techniques for the treatment of industrial wastewater from the production of paints. As can be seen from this review, the focus of most research is to develop physicochemical and electrochemical processes for water treatment, to optimize operating conditions of existing technologies, to investigate the potential use of natural adsorbents, and to develop combined technologies for water recycling and reuse. Therefore, the combination of physicochemical and electrochemical processes with greater COD and

SS reduction could potentially be effective treatment methods to address the problems caused by large volumes of wastewater that are typically disposed of directly into surface water sources.

However, it is still challenging to increase the efficiency, sustainability, and cost-effectiveness of wastewater treatment processes in the paint industry; therefore, research should focus on making them more attainable and environmentally friendly. It is essential that the treatment of wastewater from the paint industry that is discharged into large water bodies is treated to the required disposal quality to ensure environmental protection.

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