

## Scientific article

# The behaviour and removal of synthetic printing dyes from wastewaters

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**Abstract:** *One of the problems in the graphic industry is the discharge of untreated colored effluents into recipients. Printing dyes are classified as poorly biodegradable with a tendency to exhibit negative effects for the environment. Not enough is known about the interactions of these dyes with other pollutants, as well as about the effects of interactions and their impact on wastewater treatments. The applied wastewater conventional treatments are accompanied by shortcomings (the incomplete dye degradation, large amount of coagulants used, the regeneration of materials, the high cost of waste disposal, etc.) which causes a reduced efficiency of applied processes. In the last few years, studies are based on the application of improved oxidation processes, as a technique that is a prominent among treatments for reducing organic pollutants concentration. The advantages of these processes are the generation of hydroxyl radicals, powerful oxidizing species, with a tendency to degrade poorly biodegradable compounds.*

**Keywords:** *printing dyes; wastewaters; treatments; advances oxidation processes*

## 1. Introduction

Along with the textile industry, the graphic industry is one of the biggest consumers of synthetic dyes. One of the problems is the loss of about 15% of synthetic dyes during the printing process, that end in water recipients and present an environmental issue [1-3]. Also, the wastewater generated after the printing process is enriched, in addition to dyes and numerous solvents, with dioxins, dibenzofurans, pesticides, polychlorinated biphenyls, acids, bases, heavy metals, surfactants, additives, highly hazardous and toxic substances [3, 4]. They are characterized by high pH value and temperature, high conductivity, high content of suspended matter and total organic carbon (TOC), elevated values of chemical oxygen demand (COD), low values of biological oxygen demand (BOD) and low the BOD/COD ratio, which indicates a high content of non-biodegradable organic matter in wastewater [4 – 7].

The maximum allowed concentrations of certain pollutants in the wastewater of the graphic industry in the Republic of Serbia are defined by the "Regulation on limit values of emission of polluting substances into water and deadlines for their achievement" [8]. However, this

Regulation exempts flexo printing. In addition, dyes as a group of organic pollutants are not included in the systematization of "Emission limit values at the point of discharge into surface waters".

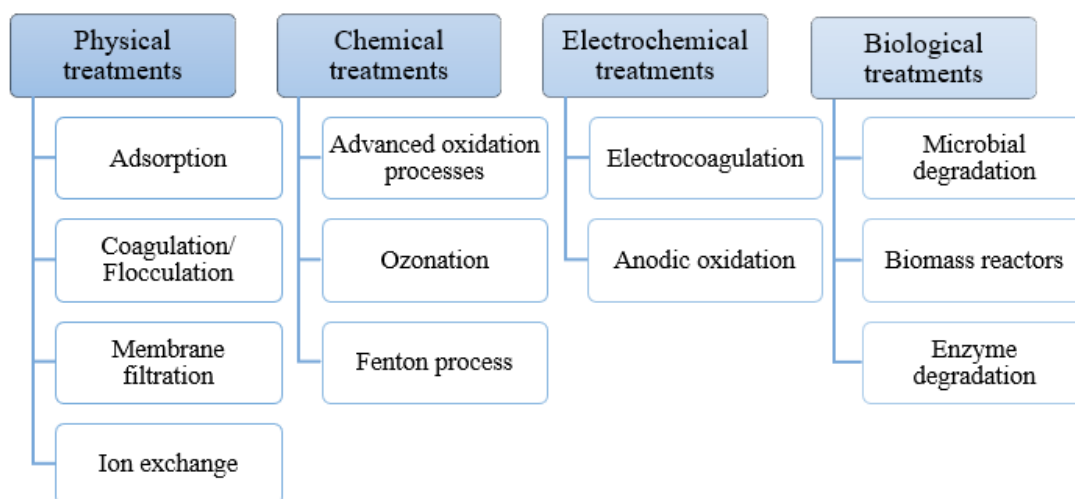
If we compare solvent dyes and water-based dyes, it is certain that water-based dyes have a greater ecological potential, since their introduction reduces the emission of volatile organic compounds into the environment. However, their use and removal from wastewater is a particular challenge. These dyes remain dissolved in water in the form of colloidal particles, which in their structure contain hydrophilic residues less than 5  $\mu\text{m}$  stabilized by electrosteric mechanisms via an ionized binder [9, 10].

Colored wastewaters are rich in high-molecular aromatic compounds, which in their structure mainly contain azo groups in the form of chromophores. Because of that, it is difficult to apply conventional biological treatments [11-13]. Even if they are present in low concentrations ( $< 1$  ppm), dyes affect not only aesthetic properties and transparency, but also the absorption and solubility of atmospheric gases in water [4, 14]. Dyes can limit the oxygenation of water surfaces by hindering the growth of aquatic species, preventing photosynthesis and reducing the biological activity of aquatic organisms [1, 12]. This means that all the available oxygen is spent on the degradation of the present organic substances, so this kind of environment very quickly becomes anaerobic [14].

Physical, chemical and biological characteristics define the type and composition of wastewater, which is the most important parameter in determining the treatment that will be applied to purify the water. This paper will describe the treatments used to remove printing dyes in wastewater treatment.

## 2. Discussion

Numerous studies have been carried out in the field of wastewater treatments with the aim of removing dye molecules, and the ideal method should ensure the efficient removal of a large amount of dyes in a short period of time without creating secondary pollution, whereby the treated wastewater could be reused [15-20]. The terms decolorization of treated wastewater and degradation of dye molecules should be distinguished. Decolorization implies the loss of dye in the form of a reduction of its initial concentration, whereby water treated in this way can still be rich in organic matter. If the dye removal process itself is accompanied by a decrease in COD and TOC, then it is a phenomenon of dye degradation [13, 20, 21]. Conducted studies point to the fact that there is no single method that applies to all types of colored effluents, and the type of industry that generates wastewater, as well as the pronounced variability of the nature of the organic dye, must be taken into account. Figure 1 shows selected dye removal methods [13, 20, 22, 23].



**Figure 1** Selected methods of dye removal from printing wastewater [13]

Biological treatments include aerobic and anaerobic microbial decomposition, a combination of these two processes, and the use of pure enzymes [2, 20, 23, 24 – 27]. The high efficiency of biological treatments in reducing dye concentration up to 90% and COD up to 80% is accompanied by advantages such as reduced energy consumption and low costs of performing the treatment [6]. Authors Qiu et al. [28] have effectively treated wastewater originating from the graphic and textile industry using aerobic activated sludge, but with the disadvantages of the process such as the retention of a certain amount of sludge and the formation of foam. At the same time, anaerobic processes become attractive for overcoming the key disadvantages of aerobic treatments, which are characterized by improved system stability, biogas production, limited energy consumption and increased efficiency of the process itself [29]. The application of biocatalysts in the form of enzymes as cheap, efficient and selective biological substances in liquid form has gained the greatest importance in the last few years [23]. The advantage of their application is reflected in their availability, ecological and non-toxic nature, pronounced functionality in a wide range of pH values and at low temperatures, as well as in the possibility of reuse [23, 30, 31]. The application of enzymes has only one drawback - the unreliable amount of material during the synthesis, which is certainly negligible compared to the disadvantages of other conventional techniques, and is most easily overcome by the appropriate selection of raw materials for efficient enzyme extraction [2, 23].

The disadvantages of biological methods are the long duration of the process and the resistance of certain dyes to aerobic treatment. It was also determined that certain carcinogenic compounds such as aromatic amines, aniline, phenol and aliphatic compounds that tend to bioaccumulate, recalcitrate and inhibit microbial activity remain in the treated effluent after the applied anaerobic treatment [4, 6, 20].

Conventional physical methods, such as adsorption [2, 23, 32-34], coagulation/flocculation [35], ion exchange [36], membrane filtration, nanofiltration, ultrafiltration [37-39] and reverse osmosis [40] are the most commonly used methods in the treatment of colored wastewaters [41].

The highest percentage of efficiency was established using the adsorption method, which proved to be extremely effective in the treatment of cationic, acidic, direct and reactive dyes, using different adsorbents, such as activated carbon, clay materials, resins, biopolymers and agricultural waste [2, 23, 32, 33]. Similar to the method of enzymatic degradation of dyes, adsorption as a

highly efficient method can be applied several times, until the adsorbent is saturated or there is no need for its regeneration or final disposal as waste. Since enzymatic degradation and adsorption stood out the most within biological and physical treatments, it would be desirable to consider the combination of the mentioned methods in the future, into a unique treatment of colored wastewater [2].

The adsorbent used significantly affects the efficiency of the adsorption mechanism. The performance of most adsorbents is controlled by various factors, the main ones being initial dye concentration, pH value, temperature, contact time and the amount of adsorbent. Various adsorbents are used to remove dyes from wastewater, such as activated carbon, clay materials, resins, biopolymers and agricultural waste [2, 23, 32, 33].

Coagulation/flocculation is most often based on the application of aluminum and iron salts (sulfates and chlorides), whereby colloidal particles are first destabilized by neutralizing their charge, while during flocculation, smaller particles are grouped together, which enables their further separation during the final phase, sedimentation [7, 42-46]. Numerous factors affect the efficiency of the coagulation/flocculation process, such as the type and dosage of the coagulant/flocculant, pH of the medium, stirring speed, reaction time and temperature, but a well-optimized coagulation/flocculation process can greatly reduce the turbidity and color of the wastewater, as well as the content of organic matter. Khannous et al. [47] pointed out the importance of the combination of the coagulation process and biosorption using fungal cultures in the treatment of wastewater from the graphic industry, where the efficiency of the process was recorded at 80%. Excessive application of coagulants negatively affects the treatment processes of dye removal from wastewater, which is determined by measuring the organic load in the form of COD, particle size distribution, pH value and suspended solids potential. In addition to removing COD and other organic loads, reducing the amount of sludge produced during the coagulation process is also essential. The efficiency of the coagulation process varies depending on the dose and experimental conditions. High removal efficiency (>95%) was achieved by introducing smaller doses of coagulants (<500 mg/l) under suitable experimental conditions [7]. Zampeta et al. [46] applied a coagulation/flocculation process to remove black dye used for packaging printing. They proved that 99% dye removal and 96% COD removal in wastewater treatment was achieved using different coagulants (aluminium-sulfate-Alum, poly-aluminium chloride - PAC and ferric chloride - FeCl<sub>3</sub>) in different concentrations, where the best results were obtained with PAC concentration of 4500 mg/L.

One of the methods for dye removal is membrane filtration, a process characterized by the passage of wastewater through a porous membrane, where dissolved substances larger than the pore size of the membrane remain trapped. Wastewater containing dyes, salts, monovalent and divalent cations and anions and other dissolved substances can be removed by passing through a porous membrane. The dye molecules will be separated from the water due to a difference in size or charge relative to the membrane. In general, larger dye molecules and those with a similar charge to the membrane will easily be rejected. The high aromaticity and reduced reactivity of dyes make their biological and chemical degradation processes unfavorable, which gives membrane technology an efficient approach to separation. On the other hand, membrane separation has disadvantages, such as high operating costs and easy contamination of the membrane, which leads to loss of performance. Furthermore, in addition to conventional membrane filtration techniques consisting of membrane materials, such as polysulfone, polydopamine, and polypyrrole, advanced integrated membrane filtration technologies have become popular in the treatment of colored wastewater. Integrated membrane systems show improvement in membrane properties resulting from additional functionalities such as stability, porosity and increased surface area. Therefore,

membrane filtration technologies for dye removal based on conventional membrane filtration (microfiltration, ultrafiltration and nanofiltration), advanced integrated membrane techniques (nanocomposite membranes and membrane distillation) and reverse osmosis are distinguished [23].

Ultrafiltration membranes with pore sizes between 0.1 to 0.001 microns are more economical and efficient because they require lower pressure than nanofiltration and reverse osmosis. However, the low rejection is caused by the large pore size, so ultrafiltration is more suitable for use as a pretreatment. A porous polyester sulfone membrane is used to remove dyes from wastewater [20].

Nanofiltration is a new advanced membrane technology applicable to a variety of wastewater treatment and decontamination applications. Nanofiltration membranes effectively remove dyes because they have a charge and a pore size between 0.5 and 2.0 nm in diameter. In continuous mode, nanofiltration eliminates almost 100% of dye, reduces up to 94.7% of COD and 76.3% of salt. In addition, nanofiltration has advantages that include less tendency of polluting and lower energy consumption, lower osmotic pressure difference, higher permeate flux, higher retention of multivalent salts, relatively low investment and low operating costs [20].

Reverse osmosis is defined as the process that uses semi-permeable spiral membranes to separate and remove dissolved solids, organic matter, submicron colloidal matter, pyrogens, nitrates, bacteria and dye from wastewater. This membrane filtration process has significant industrial application in the treatment of wastewater containing dyes and thus ensures high quality of purified water, yielding almost deionized water. The advantages of this technology include the possibility of separation without a change of state and without the use of heat energy or chemicals [20].

The latest scientific achievements have been achieved by the application of nanoparticles in the form of cellulose fibers, whose main advantages are reflected in their renewable nature, biodegradability, non-abrasive properties, easy manipulation, but also the availability of different sources of cellulose, whereby the basic cellulose fibers as the primary raw material are treated with various mechanical, chemical or enzymatic treatments [10, 20, 48].

However, certain shortcomings have also been established within the physical methods: incomplete degradation of the dye, with its translation from one form to another (from liquid form to sludge), where secondary pollution is formed, which requires the application of additional techniques for the treatment of newly formed sludge [4, 41, 20].

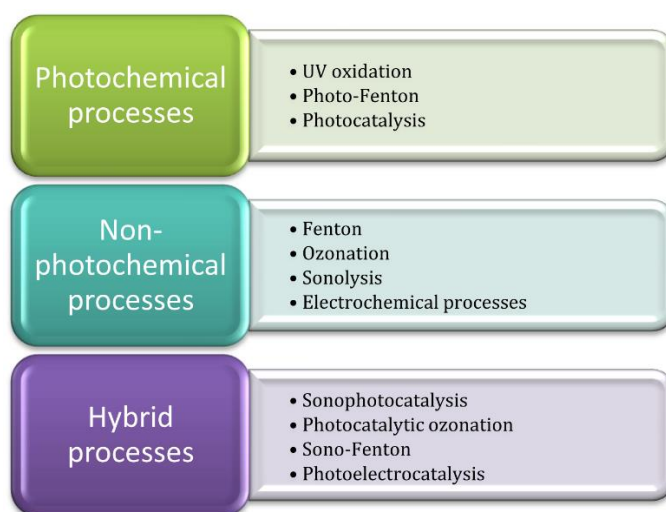
High efficiency in the field of colored effluent treatment was observed using conventional and advanced chemical processes, including advanced oxidation processes (AOPs). The diversity of AOPs in terms of different ways of producing hydroxyl radicals ( $\text{HO}\cdot$ ) it allows researchers to select the appropriate process that fits the specific needs of the experiment [20, 49, 50].

The integration between chemical oxidation processes and conventional or innovative physical and biological treatments contributes to minimizing the disadvantages of individual methods and increasing the efficiency of the applied treatment [13]. Rajabi et al. [51] have established that photocatalytic oxidation and adsorbents based on magnetic nano-particles achieve high efficiency in the removal of dyes from aqueous effluents. Ma and Xia [9] applied the Fenton process in combination with coagulation in order to remove water-based dyes from effluents originating from the graphic industry, while Tung et al. [5] proposed titanium dioxide as the optimal material for performing the electrooxidation process. Electrochemical processes, which enable high efficiency without the large use of oxidizing agents, can be a valid alternative, even if they are

still in the study phase, whereby in recent years some researchers have emphasized the application of combined bioelectrochemical treatments [52-54].

AOPs were first introduced in the 80s by Glaze and Kang [55, 56] within the treatment of drinking water, while their study was extended to the field of wastewater treatment [1, 57]. Among the water treatments that have been used so far, AOPs have a pronounced potential for treating a wide range of contaminants in industrial wastewater, which are characterized by high chemical stability and low biodegradability, such as dyes, solvents, pesticides, antibiotics, herbicides, insecticides and endocrine disruptors [58-60].

AOPs that are most often used in wastewater treatment are ozonation, photocatalysis, electrochemical oxidation, Fenton and Fenton-like processes. In general, AOPs can be classified into: photochemical processes, non-photochemical processes and hybrid processes, shown in Figure 2 [50, 61].



**Figure 2** Types of AOPs treatments [61]

AOPs are based on the in-situ production of highly reactive oxidants, which have the ability to degrade difficult-to-biodegradable compounds and convert organic contaminants into less toxic species, and under ideal conditions ensure complete mineralization and degradation of organic components to  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and inorganic salts. However, achieving such a reaction outcome is a challenge for researchers since organic compounds are characterized by high molecular mass and structural complexity [1, 62]. Among numerous oxidizing agents, the hydroxyl radical, as the strongest oxidizing agent right after fluorine, ranks among the most frequently applied oxidants within the AOPs process. It is characterized by high reactivity, non-selectivity, easy generation, pronounced control of the chemical reaction speed, electrophilic behavior and short life, which enables rapid and complete degradation of polluting substances in water.

Reactions of hydroxyl radicals with organic pollutants take place in three ways [63]:

1. electron transfer – redox reaction (equation 1)
2. by abstraction of a hydrogen atom from a C-H, N-H or OH group - dehydrogenation (equation 2)
3. by electrophilic addition - hydroxylation of organic compounds that contain an aromatic ring or multiple carbon bonds in their structure (equation 3)



where RX and PhX are aliphatic and aromatic halides, respectively.

In the mentioned reactions, organic radicals are generated and the development of radical chain reactions begins in interaction with oxygen (followed by the formation of peroxy radicals), whereby the formed reaction intermediates undergo further oxidation reactions with the generated oxidation radicals ( $HO^{\bullet}$ ,  $HO_2^{\bullet}$ ,  $H_2O_2$ ) until complete mineralization of organic pollutants [63].

Due to the partial decomposition of non-biodegradable organic substances during the AOPs process, the formation of numerous biodegradable intermediates can occur. For this reason, the application of AOPs in the form of pretreatment followed by some physical or biological treatment represents an economically profitable treatment that will result in high degradation efficiency of the organic component [64].

### 3. Conclusion

There is no single method that can be applied to all types of colored effluents. Conventional methods for the treatment of graphic industry wastewaters (biological, physical, chemical, electrochemical) are accompanied by shortcomings (incomplete degradation of dyes, use of a large amount of coagulants, regeneration of applied materials, high cost of waste disposal, etc.) which comprehensively causes a reduced efficiency of the applied processes. In the last few years, studies are based on the application of improved oxidation processes. The advantages of the mentioned processes are the generation of hydroxyl radicals, powerful oxidizing species, with a pronounced tendency to degrade difficult biodegradable compounds, such as synthetic dyes. Research should be focused on the development and application of affordable and effective methods for removing dyes from effluents, both in terms of decolorization of wastewater and their degradation, as well as minimizing or removing the toxicity of potentially formed byproducts of the reaction.

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