ISSN: 1343-4292
Volume 140, Issue 03, June, 2022

Remediation of Eosin Stain from Wastewater using Different Techniques: An Article Review

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ABSTRACT— The production and accumulation of waste water of its various types constitutes one of the serious environmental problems, due to the fact that it contains toxic and carcinogenic substances that can lead to the deterioration of any ecosystem, and thus affect human health directly or indirectly through its transfer to it through the food chain. The components of waste water that are released to environmental systems include various types of pollutants such as heavy metals, organic materials, residues of fertilizers, pesticides, industrial detergents, petroleum and its derivatives and the associated oil residues, fats of different density, acids, alkalis, and biological pollutants such as bacteria, fungi, algae, parasites, viruses and other microorganisms, among others. The most common types of these pollutants are dyes. The current paper deals with a review of methods for treating polluted water with one of the most important and most widely used types of dyes in the field of medicine, veterinary medicine and life sciences, which is the eosin dye that is used in many industries in addition to its use in the medical and biological fields. Where there are many methods and techniques that have studied the removal of this dye from contaminated aqueous solutions with different design conditions. The purpose of this review article ends with two points. The first is to shed light on this toxic dye that is posed in not small quantities by industrial, laboratory, medical and biological activities alike. As for the second, it is the inclusion of the importance of treating the water presented to the various waterways and polluted by this character through a review of the different treatment methods for this type of pollutant, which began since the mid-nineties of the last century.

KEYWORDS: Eosin, stain, dye, acid dye, acid red 87, removal and toxicity.

1. INTRODUCTION

For decades, the environment has faced many problems that beset it and its system, which sometimes led to many changes affecting most of its components [1]. Among those problems that the environment has suffered and continue to suffer from are global warming, depletion of the ozone layer, depletion of natural resources, global warming, loss of biodiversity and natural disasters such as hurricanes, earthquakes, floods, drought and desertification [2]. At the same time, the increase in population density also contributes to increasing the burden on the capabilities of the environment, but one of the most pressing problems affecting non-living components and living organisms alike is pollution [3]. Pollution is a serious problem whose impact does not depend on a specific time and region and takes different types depending on the foreign components entering the natural environment that cause negative and harmful change [4]. It is one of the major problems facing humans and the environment, especially after the technological development accompanying contemporary life [5]. Pollution occurs in its various forms, whether it is air, water, or soil pollution as a result of the presence of some harmful organic and inorganic substances, or due to an increase or decrease in the ratios of some basic components in the environment from their natural ratios. This happens as a result of human interventions or by some natural phenomena [6]. Accordingly, environmental pollution is defined as an increase in the amount of substances in their gaseous, liquid, or solid forms, or the addition of one form of energy, such as: sound energy, thermal energy, radioactivity, and others within the environment, which makes it unable to analyze or dissipate these materials and energy, or reduced, or recycled, and are no longer able to store materials and various forms of energy in harmless forms [7], [8]. This pollution includes all substances that have a negative impact on the environment, or the living organisms that live in it [9]. The main types of pollution can be divided into: water pollution, air pollution, soil pollution, light pollution, and noise pollution [10]. One of the most severe types of pollution affecting the environment is water pollution, which is the most important main problem facing environmental workers, according to the great role of water in daily life at various levels [11]. Water is the secret of life for humans, animals and plants, as well as being an essential element in various industries and activities [12]. Therefore, pollution of water sources with any kind of pollutants poses a threat to the preservation of natural water systems. In general, almost no industry is devoid of its need for water in terms of its quantities and degree of purity, depending on the nature of the industry [13]. As a result, large quantities of polluted water with various types of organic pollutants are disposed of to water sources [14]. Dyes are organic chemicals capable of absorbing and reflecting light at selective wavelengths within the visible range of the electromagnetic spectrum. Often the dye needs a liquid medium to be able to transfer to the coloring matter, and it may need a mordant to improve the color fastness of the dyed fibers [15].

Various industries and activities such as textiles, glass, paper, paint, tanning, inks, cosmetics, plastics, photography, food, and medical specimens offer different quantities and types of dyes [16]. Through the effluent of wastewater, these dyes make their way into the aquatic systems and from there to humans, thus being an important source of pollution as many of the industrial dyes used are stable in composition, relatively stable and not easily biodegradable [17]. In addition, these dyes contribute to preventing the penetration of the sun light into the aquatic environment and prevent the occurrence of necessary light activities such as photosynthesis. Because of the complex organic composition of these pollutants, conventional treatment methods may be inefficient for removing dyes from aqueous solutions [18]. Between 10-15% of the industrial dyes used are released as liquid waste to nearby water sources [15]. Therefore, specialists in environmental treatment and pollution control affairs turned to search for ways and methods to address the problem of water contaminated with dyes [19]. There are many physical and chemical methods for removing dyes, including sintering, coagulation, adsorption, flotation, normal photolysis, catalyst, solvent extraction, electromagnetism, membrane filtration, ultrafiltration, oxidation, reverse osmosis, ion exchange, etc. [20]. Among all above technique, the adsorption is considered as the best one due to its simplicity and efficiency. It's well known that the adsorption technique is used very important media which has a large surface area, i.e. activated carbon [21], but due to the high cost of this material the researchers seeks for another cheap and active media. One of the solutions for this issue is the agricultural waste. These materials are low cost, available and cause pollution to environment when accumulated [22]. Thus, the researchers studied different types of agro-waste like rice husk [23], eggshells [24], watermelon rinds [25], lemon peel [26], orange peel [27], etc. The adsorption is proving its activity to remediate the contaminants not only from water, but also from soil [28] and crude oil [29]. Regardless of adsorption disadvantage which is the accumulation of residues which is almost be toxic, the adsorption is deemed a promising technique not only for pollution treatment but also from utilizing the adsorption waste in a benefit and economic manner, which is exactly the ZRL concept [30]. Among the various types of contaminated, dyes are toxic, carcinogenic materials or caused many factors lead to diseases [31] or deficient in multi important substance like vitamins [32], iron [33] etc. Therefore dyes must be removed or decreased its concentration to minimum. This research focuses on the biological dyes, specifically the eosin dye. So, it deals with a review of most of the methods used to treat solutions contaminated with eosin dye.

Eosin: Eosin are red crystals or reddish-brown or orange-colored powder, and the origin of the name Eosin came from the word "eos" from the Greek, meaning the dawn, and it is the name of the goddess of dawn according to the belief of the ancient Greeks. Its chemical formula is $C_{20}H_6Br_4Na_2O_5$, and its molecular weight is 691.86 g/mol, soluble in water and slightly soluble in alcohol. The concentrated solution is reddish-brown in color with a green phosphorous color. Alcoholic solutions of it show a clear phosphorous green color. It is used to make lipstick and as a guide in analytical chemistry. It is also used in the manufacture of red ink, colored paper, and others, and eosin can be used to color proteins in the cytoplasm, as well as color collagen and muscle fibers for examination under a microscope. Structures that stain easily with eosin are called eosin affinity [34]. There are many types of eosin dye, including: eosin Y: eosin yellowish or Acid Red 87, eosin S: ethyl eosin or Solvent Red 45; eosin B: eosin bluish or Acid Red 91; Eosin- phloxine and Picro-eosin. The most famous of these dyes and the most widely used is eosin yellowish, which is a form of eosin and is also called acid red 87. Chemically, it is expressed as a tetrabromo derivative of fluorescein and is called 2-(2,4,5,7-tetrabromo-6-oxido-3-oxo-3H-xanthen-9-yl)benzoate [*in its deprotonated form*], and this dye is soluble in water and alcohol. This stain has a color index of 45380 (C.I. 45380) and is widely used as an acid red dye to clarify cytoplasmic components in biological samples in histology, specifically in the H&E (Haematoxylin and Eosin) stain. Eosin yellow is one of the components of Papanicolaou stain, which is used in the Pap test. It is also used in organic manufacture as a sensitizer. Figure 1 shows the chemical structure of yellow eosin [34].

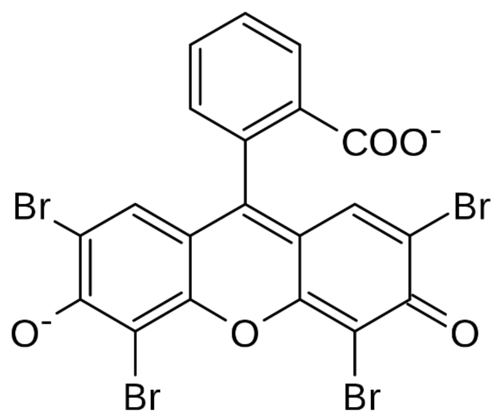

Figure 1 the chemical structure of yellow eosin [34]

Toxicity: Eosin Y is an anionic xanthan family dye in nature and highly toxic water-soluble (acid dye) and it harms water quality, it is a kind of organic dye which is difficult to degrade, dye widely used as a biological stain for human-medical cell diagnosis and serves the histological and clinical-cytological investigation of sample material of human origin. It also displays yellow-green fluorescence and with the wavelength of maximum absorbance (λmax) of 517 nm [35]. The eosin dye is harmful if swallowed, inhaled, drunk, or absorbed through the skin and causes eye and skin irritation [36], inhibits protein-protein interaction [37] and triggers genotoxicity in man [38]. Some researchers have tried to remove it by adsorption, but no one has attempted for complete degradation [39].

Removal of eosin dye: Due to the toxicity of this dye and its many uses, researchers have resorted to using multiple methods to treat water contaminated with it and get rid of its effects. In this review article, we will review the methods used to remove eosin.

[40] studied the process of separating a group of dyes, namely, brilliant green, neutral red, eriochrome black T, and eosin, from aqueous solutions by flotation method, using oleic acid as a surfactant. Eosin dye concentrations were measured using A Griffin Model 40 Colorimeter and the calibration curve was

prepared at a wavelength of 520 nm. The operating conditions used were oleic acid concentration, dye concentrations (including eosin dye), temperature and acidic function. The experimental results indicated that the flotation of eosin was the lowest among the dyes by 2%, while the increase in the concentration of oleic acid and the concentration of the dye did not affect the percentage of eosin flotation at the neutral pH. However, the efficiency of the flotation process reached 100% at the ideal conditions of pH ≤ 6, oleic acid concentration and dye concentration 10-5 and 6.63×10-3, respectively. As for the temperature, it had no effect on the efficiency of the flotation process beyond 85 °C, except that it shortened the required flotation time. None of the materials used as foreign ions by the researchers had any significant effect on the flotation process.

Photodegradation was used by [41] of the eosin using a dispersing aqueous solution of titanium dioxide (TiO_2) and at visible light. The adsorption - desorption process was carried out using a photoreactor consisting of a Pyrex glass vessel with a capacity of 70 ml. 50 ml of dye in a specified concentration with 25 mg of titanium dioxide TiO_2 was applied together and stirred overnight in a dark environment to ensure equilibrium was achieved. The change in the eosin concentration as a result of the treatment process was determined using a UV-Vis spectroscopy device (Shimadzu 1600A) at a wavelength of 514 nm. The experimental results that were carried out at a concentration of 2×10^{-5} mol/L of eosin showed that the value of the adsorbed dye was decreasing with the increase of the acidity function, as the adsorbed dye concentration became zero after it was 15 μmol/g at pH 10 and 4, respectively. While without the use of titanium dioxide, the adsorbed concentration remains constant at 1 μmol / g, despite the difference in the acidity function. The researchers calculated the initial kinetics rates and observed that they change between 1.0×10^{-7} to 2.5×10^{-7} mol/L.min. The mechanism of photolysis has also been described, which depends on plausible routes. These routes are (1) a route of singlet oxygen and (2) a route involving electron transfer from an excited state of eosin to the TiO_2 particles.

Since the beginning of the new millennium, modern and advanced nanotechnology has played a role in treating water contaminated with affinity-based ionic dyes. [42] investigated the removal of a number of dyes, including eosin blue, by using multi-walled carbon nanotubes (MWCNTs) of both types (MWCNT-P) and (C-MWCNT). These materials pick up the dyes according to the reactions of Van der Waals between the aromatic backbone of the dyes and the carbon atoms. While negative charges of large molecular weight such as humic acids and colloids of large size are excluded as a result of electrostatic repulsion and size exclusion. The conditions used were to treat 15 ml of a dye solution at a concentration of 10 μM for half an hour, and the dye concentrations were determined using a UV Spectrophotometer (Jasco UV-550). The experimental results showed that the adsorption capacity (μmol/mg)) were values of 0.73 and 0.33 for (MWCNT-P) and (C-MWCNT), respectively. While the adsorption capacity of eosin blue dye with carbon fibers (CNFs) and activated carbon (ACTC) were 0.09, 0.06, 0.25 and 0.19 for each CNF-P, C-CNF, ACTC-P and C-ACTC, respectively.

[35] indicated the possibility of using chitosan hydrobeads to remove the yellow eosin dye using the adsorption technique from aqueous solutions. Shrimps shells were used as a source for preparing chitosan hydrobeads, which were used with two different types in the experiments to treat eosin contaminated aqueous solutions. The first was ordinary chitosan hydrobeads, while the granules treated with a concentration of 0.1 M of ammonia sulfate ($(NH_4)_2SO_4$) represented the second type. The researchers studied the effect of altering the acidic function, temperature, initial concentration of dye and contact time among the operational factors and at a constant agitation speed in a batch-mode adsorption unit. The experimental results indicated that the adsorption efficiency was inversely related to the acidity function, as the value of the adsorption capacity was 77 mg/g and decreased to less than 2 mg/g at pH=4 and pH=12,

respectively, for both types of adsorbent used, but the modified material reduced pH sensitivity. As for the temperature behavior, it was similar to the previous behavior and that the highest efficiency was determined at 30°C. In addition, the adsorption was exothermic and spontaneous at all temperatures. The results of the concentration change did not differ from the temperature and the acid function, as the removal efficiency decreased from 89.76% to 55.88% by increasing the initial concentration of the dye from 50 to 200 mg/L, respectively. The isothermal results showed that the adsorption is subject to the Langmuir model with a perfect correlation coefficient. The contact time factor was not affected by the change in the initial concentration of the dye, as practical experiments proved that the equilibrium time occurs at about 600 minutes and that the adsorption matches the pseudo second order model.

A micellar enhanced ultrafiltration (MEUF) method has been used by [43] to remove the toxic eosin dye with the assist of cetyl (hexadecyl) pyridinium chloride (CPC) as the cationic surfactant. The eosin contaminated water treatment unit was a cross flow cell and organic polyamide of molecular weight cut-off (MWCO) 1000 membrane used. The studied design factors were, the dye concentration, acidity function, pressure, cross flow rate and time ranged between $4\text{-}40\times10^3$ kg/m^3, 6.8-7.05, 345 and 414 kPa and 30-75 l/h (Re= 315-790) and (1-60) min. respectively, while the CPC concentration was constant at a value of 10 kg/m^3 and all experiments were performed at 32±2 °C. The concentration of the dye was confirmed by a UV spectrophotometer (Thermo Spectronic, USA; model: GENESYS 2) at a wavelength of 517 nm. The researchers concluded that the maximum removal of eosin dye was close to 80%.

The methods of removing eosin dye were not limited to the above, but the oxidation method using the Fenton process method and the photo-Fenton process method using sunlight and an artificial light source were tried before [44]. The results showed that 94.1% of the dye could be removed in an hour and a half by the solar-Fenton process, and the removal efficiency was increased to 96% using oxalic acid. The relationship was inversely between the removal percentage for the acidity function and the initial eosin concentration. While it was positively related to the concentration of oxalic acid, the concentration of iron ions ferrous dosage, the concentration of hydrogen peroxide H_2O_2 dosage and the treatment time. Estimation of the concentration of eosin dye Y was performed using a HACHDR2000 spectrophotometer at 515 nm wavelength. The researchers were able to derive equations to describe the kinetics of removal based on the concentrations of the studied materials, where the powers of hydrogen peroxide concentration, iron ions concentration and eosin yellow dye concentration in the rate equations were 1.26, 1.14 and 0.78, respectively.

[45] used chitosan nanoparticles prepared by ionic gelation with the presence of tripolyphosphate as an adsorbent for eosin yellow dye in the adsorption batch type unit. The obtained results showed that the ideal adsorption efficiency was obtained at an acidity function less than 6, where it begins to gradually decrease until the adsorption stops at pH = 10. As for the equilibrium time, it was 120 minutes, while the relationship between efficiency and temperature was a direct relationship within the study range 20-50 ºC. The researchers studied the distribution of zeta potential and concluded that its value is 65 mV. The isothermal relationship showed that the adsorption corresponds to the Langmuir model and that the adsorption capacity was 3.333 g/g, while the thermodynamic study showed that the adsorption is physical according to the enthalpy value of 16.7 kJ/mol and that it is endothermic.

Sugarcane bagasse waste (SB) modified with Tetraethylenepentamine (TEPA) was investigated for its adsorption capacity for eosin yellow dye from aqueous solutions, at different conditions of pH, eosin initial concentration, temperature and contact time in a batch unit. The results obtained by [46] indicate that the maximum dye adsorption capacity using (TEPA) is 18 times higher compared to cane sugar residues alone

(SB). The optimum conditions for adsorption are 6, 25°C, 50 mg/l, 6 h for the pH, temperature, initial concentration and equilibrium time, respectively. On the other hand, the results of the study showed that the adsorption kinetics is identical with the pseudo-second order model, and that the Langmuir model was the closest to representing the data compared to other models.

2. Conclusions

It is noted from the reviewed references that there has been a widespread trend since the mid-nineties of the last century to treat water contaminated with eosin dye, due to its wide spread and use in many commercial, industrial and medical activities. As aforementioned the biological stains causes harmful effects on both human and environment especially when combined with other pollutants [47], [48]. On the other hand, the various methods used in trying to remove this dye give the impression that the subject of this dye is very important and requires continuous, continuous and modern studies for the purpose of getting rid of this dye in a safe, useful and environmentally friendly manner. Undoubtedly, the adsorption technology satisfies a large part of these hypotheses because it is a promising and cheap technology that does not require high operational costs. It is possible to rely on the waste because it is available and not of value and efficiency suitable for removing eosin dye from waste water and aqueous solutions.

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