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Investigating the use of ozonation process with calcium peroxide for the removal of reactive blue 19 dye from textile wastewater

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ABSTRACT

Colors have a complex molecular structure, often toxic, mutagenic, nondegradable biologically, and stable, which, when they enter the environment, cause environmental problems. Advanced oxidation processes are the most efficient and effective methods for decomposing dangerous organic contaminants which are resistant and nonbiodegradable in aquatic environments. The study aimed to determine the efficiency of the ozonation process with calcium peroxide for the removal of reactive blue 19 dye from textile wastewater. Synthetic samples were prepared with blue dye concentrations of 10–300 mg/L. The removal efficiency of reactive blue 19 dye and chemical oxygen demand (COD) by ozonation with calcium peroxide in different conditions of pH, calcium peroxide concentrations, contact time, concentration of the reactive blue 19 dye, and 1 g of ozone/min was examined. The optimal conditions for synthetic samples were obtained. The maximum removal of blue dye and COD with a concentration of 5 mg/L reactive blue 19 dye, pH = 3, 0.025 mg/L of calcium peroxide, contact time of 40 min, were 100% and 85.6%, respectively, in synthetic samples and 90% and 76.5%, respectively, in real samples.

Keywords: Reactive dye; Calcium peroxide; Advanced oxidation; Aquatic solution

1. Introduction

Many industries, such as the cosmetics, food, tanning, pharmaceutics, dye, paper and paperboard industries, plastics, and textiles industries, produce colored wastewater [1]. Annually, more than 7 × 104 tons of paint are produced worldwide, and some of it is discharged through industrial wastewater [2,3]. The textile industry is one of the biggest water consumers, and it produces colored wastewater with dye concentrations of 10–200 mg/L [4]. Almost 50% of reactive colors, 8%–20% of dispersed paints, and 1% of pigments are discharged into the environment through wastewater [5]. Colors have a complex molecular structure, mostly toxic, carcinogenic (producing amine groups in anaerobic decomposition), mutagenic, nonbiodegradable, and persistent, that cause environmental problems when they enter the environment [6,7]. Colors are categorized into different types based on their chemical compositions and applications [8]. On the basis of chemical structure, colors are divided into 20–30 groups, the most important of which are azo, anthraquinone, and phthalocyanine [9]. Azo colors comprise the biggest and most important group of colors. They are characterized by the presence of one or more azo groups (–N=N–) that make a relationship between their annular structures. The colors used in textiles are divided into the three main classes of anionic (direct, reactive, and acidic), cationic (alkaline colors), and nonionic disperse colors [10]. The discharge of colored sewage into the environment and into aquatic ecosystems damages the natural aspect, prevents the penetration of light into the depths of the water,

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disrupts photosynthesis, destroys aquatic plants, increases the growth of some types of algae, decreases the amount of dissolved oxygen in water, and jeopardizes fish life [11]. Most colors are resistant to light and heat. Conversely, the technology that causes these colors to resist bleaching, sunlight, and oxidation causes them to be unremovable with conventional wastewater treatment systems [12]. Different methods have been considered for the treatment of colored sewage and natural organic material, including electrocoagulation, coagulation and flocculation, chemical oxidation, electrochemical treatment, ion exchange, advanced oxidation processes (AOPs), enzymatic decomposition, adsorption, photocatalysts, and nanotechnology [13–15]. The management of wastewater that has reactive colors is very important. Reactive dyes are sulfonated and very soluble in water. Their absorption onto biological masses is very weak, and they do not decompose under aerobic conditions [16,17].

Reactive blue 19 dye is a component of the anthraquinone colors, and it is very resistant to the chemical process that is currently used in the textile industry. This color has a low stabilization efficiency (80%–75%) due to the competition between the reactive state (vinyl sulfone) and hydrolysis reactions. Chemical oxidation, such as chlorine, hydrogen peroxide, and ozone, can overcome the disadvantages of other methods [18]. Among all the oxidation methods, it seems that ozonation is an efficient method for the treatment of colored sewage. In most applications such as removing dyes, disinfecting, and removing odors, flavors and organic compounds, ozone has strong oxidizing properties. At a standard temperature and pressure, it is unstable and has low solubility in water [19]. Simplicity, low cost, and high efficiency make the AOP one of the most common technologies used for the removal of pollutants, especially decomposition, and the removal of dangerous organic contaminants which are resistant and nonbiodegradable in aquatic environments [20]. This process can convert organic compounds into water and carbon dioxide [21]. To date, different types of AOPs for the treatment of water and wastewater have been expanded and examined, including the original use of ozone as an oxidant [22]. Calcium peroxide is an effective source for the production of hydrogen peroxide. The unique features of calcium peroxide include slow dissolution in water and the release of oxygen molecules [23]. In addition, calcium peroxide decomposes in water and is converted to hydrogen peroxide and calcium oxide. The advantages of calcium peroxide include its environmental compatibility, ease of handling, low cost compared with other substances, long-term, and high impact [24].

In 2016 in Iran, Jamshidi et al. [25] used reed powder for the removal of reactive blue dye. In 2016 in Iran, Malakootian et al. [26] used calcium peroxide for the removal of reactive red 198 dye. In 2014 in Iran, Ghaneian et al. [27] used jujube stem powder for the removal of reactive dye. Wijannatrong et al. [28] used the ozonation process for the removal of reactive dyes from textile wastewater in Thailand in 2013. Sancar et al. [29] used O_3 and O_3/ultrasound for the decolorization of dye in Turkey in 2013.

The study aimed to determine the efficiency of the ozonation process with calcium peroxide for the removal of reactive blue 19 dye from textile wastewater.

2. Materials and methods

This empirical research was accomplished in the first half of 2017 at the Research Center of Environmental Health Engineering at the Kerman University of Medical Science. Stock solutions with a reactive blue 19 dye concentration of 1,000 ppm were prepared on a daily basis. Thereafter, spectrophotometry was employed to scan the blue 19 dye. Maximum absorption was obtained at a wavelength of 518 nm. The amount of ozone of 1 mg/min was determined. The gas output from the ozone generator was passed for 10 min from two containers containing a 2% solution of potassium iodide. Both containers contained 250 mL of potassium iodide solution. After 10 min of ozonation, 200 mL of the potassium iodide solution was removed. Thereafter, 10 mL of 2 N sulfuric acid was added. Subsequently, the solution was titrated using 0.005 N sodium thiosulfate until the yellow color of iodine disappeared. Thereafter, one to two drops of starch were added, and titration was continued until the blue color disappeared. Ultimately, the volume of consumption of sodium thiosulfate was recorded. The ozone produced was determined with Eq. (1). The calibration curves for pH values of 3, 5, 7, 9, and 11 were drawn. Concentrations of 10, 50, 100, 150, 200, 250, and 300 mg/L of stock solution were taken. Thereafter, they were mixed with concentrations of 0, 0.025, 0.05, 0.075, and 0.1 mg of calcium peroxide and stirred by a shaker. Next, the solution was ozonized at times of 5–50 min. The solution was then centrifuged. In the next step, the amount of the sample absorbed was measured by spectrophotometry. The schematic figure for the experimental setup is shown in Fig. 1. The real sample was prepared from wastewater from the Yazdbaf textile company (Yazd, Iran). All tests were performed on the real sample. All experiments were carried out in laboratory temperatures (22°C ± 1°C). Experiments were repeated three times, and the results were reported as means. Data analysis was carried out using descriptive statistics. Finally, the reactive blue 19 dye removal efficiency was calculated with Eq. (2) [30]:

$$\text{Ozone concentration (mg/L)} = ((A + B) \times N \times 24)/(T\ (\text{min})) \quad (1)$$

where A: consumption of sodium thiosulfate for the first container (mL); B: consumption of sodium thiosulfate for the second container (mL); T: ozonation time (min); and N: normality of sodium thiosulfate.

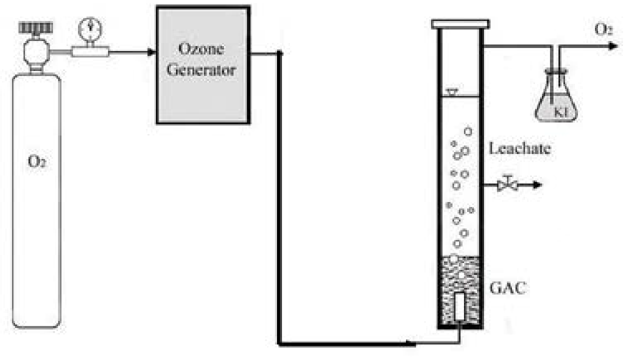

Fig. 1. Schematic figure for experimental setup.

Removal efficiency (%) = $(C_i - C_f)/C_i \times 100$ (2)

where C_i: the initial concentration of reactive blue dye (mg/L) and C_f: the residue concentration of reactive blue dye (mg/L)

Reactive blue 19 dye was purchased from Alvan Sabet Hamedan Company (Hamedan, Iran). Other substances, such as calcium peroxide with a purity of 98%, sulfuric acid, and sodium hydroxide, were purchased from Merck (Germany). In this study, the ozone generator devices ARDA (model (MOG-5 G/H), air compressor (model FL25), pH meter (Model EDT-R357), flow meter, centrifuge (model-150), and spectrophotometer (model Schimadzu/UV-visible 1800) were used.

3. Results and discussion

Changes in the UV-visible light spectrum and the determination of reactive blue 19 dye, which is the highest point in the reactive blue 19 dye prototype, was observed at a wavelength of 518 nm.

3.1. Effect of pH

The effects of pH on the removal of reactive blue 19 dye is shown in Fig. 2.

Fifty minutes after the ozonation process with calcium peroxide at pH = 3, the maximum removal efficiency was 100% for synthetic samples. With increases in pH, the efficiency of the process was reduced. At pH 11 and 50 min after the process, the removal efficiency in the synthetic sample was 62.3%. Calcium peroxide at low pH has high stability and higher solubility [31]. The hydrogen peroxide obtained from the ozonation process with calcium peroxide played an important role in the oxidation of organic compounds [32]. When pH was decreased, removal efficiency was increased. Moreover, with the release of calcium carbonate, pH was increased, and it created the appropriate conditions for discharging wastewater into the environment [32]. The calcium peroxide reaction in an acid solution was carried out according to Eqs. (3)–(5):

$$CaO_2 + 2H^+ \rightarrow Ca_2^+ (aq) + 2H_2O_2 \quad (3)$$

$$Ca(OH)_2 + 2H^+ \rightarrow Ca_2^+ (aq) + 2H_2O \quad (4)$$

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 (s) + H_2O \quad (5)$$

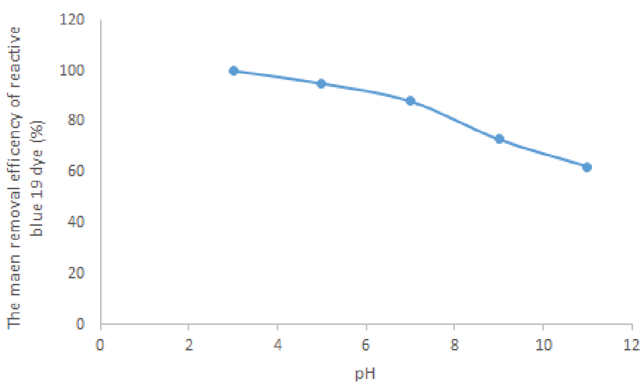

Fig. 2. Effects of pH on the efficiency of the ozonation process with calcium peroxide on the removal of reactive blue 19 dye.

These results are in agreement with those of Malakootian et al. [26], who studied the use of calcium peroxide for the removal of reactive red 198 dye in 2016 in Iran, Qian et al. [33], who used calcium peroxide for the removal of toluene in 2013 in China, Northup et al. [24], who used calcium peroxide in Reform Fenton in 2008 in the United States, and Rezaee et al. [34], who used UV/H_2O_2 for the removal of reactive blue dye in 2008 in Iran. In acidic pH, the production efficiency of hydrogen peroxide and dissolution rate of calcium peroxide increase [31].

3.2. Effect of calcium peroxide

The effects of calcium peroxide concentration on the removal of reactive blue 19 dye are shown in Fig. 3.

A concentration of 0.025 mg/L of calcium peroxide removed 100% of the reactive blue 19 dye in the synthetic sample. With a reduction in the calcium peroxide concentration, the removal efficiency was reduced. When the amount of calcium peroxide was 0.1 mg/L, a reactive blue 19 dye removal efficiency of at least 59% was obtained in the synthetic sample. With an increase in calcium peroxide, the reactive blue 19 dye removal efficiency was reduced. This result corresponds with the study results of Malakootian et al. [35], who used the ozonation process with calcium peroxide for the removal of metronidazole in 2017 in Iran, and Rahmani et al. [32], who used the ozonation process with calcium peroxide for the removal of ciprofloxacin in 2015 in Iran. The presence of radicals is limited in solution; they act as radicals to capture and cause the oxidization of radicals to intermediate materials [36]. When ozone gas is transferred into water, the dissolved ozone reacts with the organic and inorganic compounds. In the initial reaction of ozone with the chromophores group of dyes in wastewater, ozone can react with organic compounds at the double bonds of carbon, nitrogen (C=C, N=N), and aromatic rings. The cause is a product of the destruction of the bonds. Ozone can decompose to oxygen by splitting into radicals such as hydroxyl radicals (OH^{o}), OH_3, OH_4, and super oxide (O_2^-) [28]. The radical that occurs is a very strong oxidant that reacts with various substances. The mechanism of ozone decomposition with hydrogen peroxide is done according to Eq. (3), and

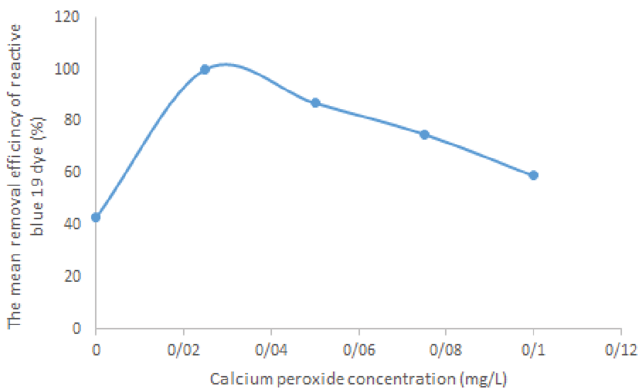

Fig. 3. Effects of calcium peroxide on efficiency of ozonation process with calcium peroxide on the removal of reactive blue 19 dye.

causes the production of hydroxyl radicals. Simultaneously, the following Eqs. (6)–(8) are also involved in the aquatic solution that causes reductions in the efficiency of the process when the presence of hydroxyl radicals in the solution is increased [32]:

$$OH^{o} + H_2O_2 \rightarrow H_2O + HO_2^{o} \quad (6)$$

$$OH^{o} + HO_2 \rightarrow H_2O + O_2 \quad (7)$$

$$HO_2^{o} + HO_2 \rightarrow H_2O_2 + O_2 \quad (8)$$

3.3. Effect of reactive blue 19 dye concentration

The effects of the initial concentration of reactive blue 19 dye are shown in Fig. 4.

The removal efficiency decreased with increases in the concentration of reactive blue 19 dye. Hence, at a concentration of 10 mg/L, the maximum removal efficiency of reactive blue 19 dye maximum from the synthetic sample was 100%, and at a concentration of 300 mg/L in the synthetic sample, the removal efficiency was 68.9%. Increasing the reactive blue 19 dye concentration caused removal efficiency to be reduced. These results are in agreement with the study results of Ghaneian et al. [27], who used jujube stem powder for the removal of reactive dye in 2014 in Iran and Olyaie et al. [36], who used calcium peroxide for the removal of arsenic from aquatic solutions in 2012 in Iran. Increasing the concentration of pollutant increased the consumption of oxidizing substances such as ozone molecules and hydroxyl radicals. Whenever the concentration was lower, the contaminant removal efficiency was higher, and decomposition of the pollutant and production of the intermediate was reduced [32].

3.4. Effect of contact time

The effects of contact time on the removal of reactive blue 19 dye are shown in Fig. 5.

By increasing contact time, removal efficiency was also increased. Hence, at the time of 5 min, the removal efficiency in the synthetic sample was 50.9%. At a contact time of 50 min, the maximum removal efficiency in the synthetic sample was 100%. By increasing contact time, the removal efficiency was also increased. By increasing contact time, the removal efficiency was increased, because more hydroxyl radicals were produced than ozone molecules. Contact time, that is, the time required to achieve the desired targets in a refining process, is an important variable in designing and leading the process of oxidation. These results are in agreement with those of Malakootian et al. [26], who used calcium peroxide for the removal of reactive red dye in 2016 in Iran, and Jamshidi et al. [25], who used reed powder for the removal of reactive blue dye in 2015 in Iran.

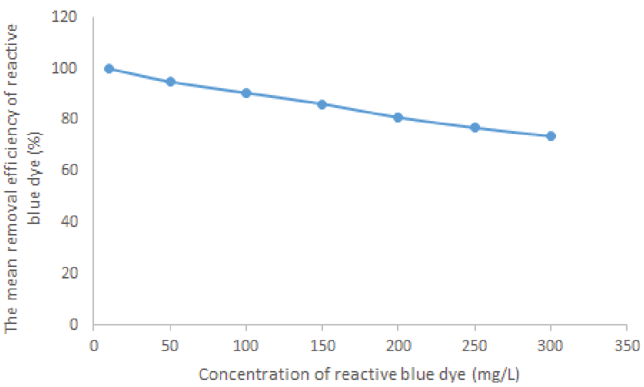

Fig. 4. Effects of initial concentration of the reactive blue 19 dye on the efficiency of ozonation process with calcium peroxide.

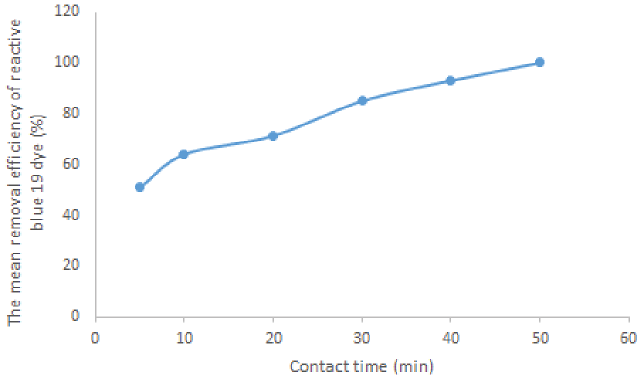

Fig. 5. Effects of contact time on the efficiency of ozonation process with calcium peroxide for the removal of reactive blue 19 dye.

3.5. Effects of calcium peroxide, ozonation, and calcium peroxide with ozonation

The effects of calcium peroxide, ozonation, and calcium peroxide with ozonation on the removal of reactive blue 19 dye are shown in Fig. 6.

The maximum removal rates by calcium peroxide, ozonation, and ozonation with calcium peroxide were 43.3%, 68.4%, and 100%, respectively. The removal efficiency of ozonation with calcium peroxide was greater than the other two methods. According to Eq. (3) and Eqs. (6)–(8), calcium peroxide produced hydrogen peroxide and reacted with ozone to produce radical OH^{o}, and radical OH^{o} has a great effect on dye removal [28,32].

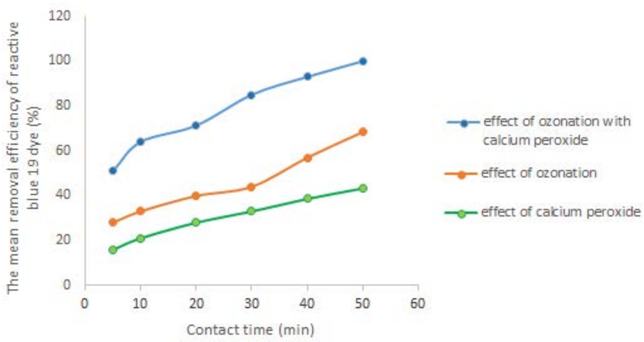

Fig. 6. Effects of calcium peroxide, ozonation, and calcium peroxide with ozonation on the removal of reactive blue 19 dye.

Table 1
Quality of turbidity, COD, TSS, EC, pH, carbonate, and chloride of Yazdbaf textile wastewater

Parameter	Amount
COD (mg/L)	1,400
TSS (mg/L)	1,800
pH	11
EC (µs/m)	4,340
Turbidity (NTU)	520
Carbonate (mg/L)	7.3
Chloride (mg/L)	531

The results of determining the quality of the Yazdbaf textile wastewater are shown in Table 1.

Under optimal conditions (pH = 3, calcium peroxide concentration = 0.025 mg/L, reactive blue 19 dye concentration = 5 mg/L, and contact time = 50 min), the removal efficiency in the real sample was 85.6%. In addition, the amounts of chemical oxygen demand (COD) removed from the synthetic and real samples were 85.6% and 76.5%, respectively.

Under optimal conditions (pH = 3, calcium peroxide concentration = 0.025 mg/L, reactive blue 19 dye concentration = 5 mg/L, and contact time = 50 min), the removal efficiency of reactive blue 19 dye and COD in the synthetic sample were 100% and 85.6%, respectively. In the real sample, these values were 90% and 76.5%, respectively. The removal efficiency of reactive blue 19 dye and COD in the real sample was less than that of the synthetic samples. The reason for this reduced efficiency is the existence of organic substances such as turbidity, TSS, carbonate, and chloride in textile wastewater that caused the consumption of oxidizing substances such as ozone and hydroxyl radical molecules before other substances such as colors and organic materials; therefore, the removal efficiency of reactive blue 19 dye and COD was less in the real sample than in the synthetic sample.

4. Conclusion

The relatively high efficiency of the ozonation method with calcium peroxide for the removal of reactive blue 19 dye in addition to its advantages such as strong oxidization, decomposition of refractory organic compounds, indivisibility, and high efficiency make this method advisable for use in the removal of reactive blue 19 dye from textile industry wastewater.

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