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Advanced oxidation processes based on zero-valent aluminium for treating textile wastewater

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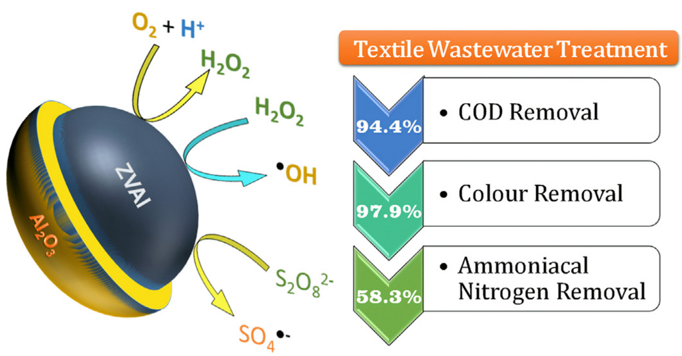
HIGHLIGHTS

- Textile wastewater treatment by ZVAl based AOPs.
- Enhancement in process efficiency of ZVAl/O_2 process with Fe^{3+}, H_2O_2 and persulfate addition.
- 97.9% colour, 94.4% COD and 58.3% ammoniacal nitrogen removal.

GRAPHICAL ABSTRACT



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ABSTRACT

Treatment of textile wastewater by advanced oxidation processes (AOPs) based on zero-valent aluminium (ZVAl) were carried out in the present study. COD removal efficiencies of ZVAl/O_2, ZVAl/Fe^{3+}/O_2, ZVAl/Fe^{3+}/O_2/H_2O_2 and ZVAl/Fe^{3+}/O_2/persulfate processes were monitored and optimized. Colour and ammoniacal nitrogen removal efficiencies of each process at the optimal operating conditions were compared. Pollutant removal efficiencies of AOPs were followed the order as: ZVAl/Fe^{3+}/O_2/H_2O_2 > ZVAl/Fe^{3+}/O_2/persulfate > ZVAl/Fe^{3+}/O_2 > ZVAl/O_2. Maximum COD, colour and ammoniacal nitrogen removal efficiencies of ZVAl-based AOPs were found as 97.9%, 94.4% and 58.3%, respectively at 1 g/L ZVAl, 0.5 g/L Fe^{3+}, 6.7 g/L H_2O_2 and after 3 h of contact time. External addition of tert-butyl alcohol to the processes revealed that in-situ hydroxyl and sulfate radicals are the main oxidants responsible for the oxidation of pollutants. Overall, ZVAl-based AOPs are efficient for treating effluents generating from textile industry.

1. Introduction

Environmental pollution increases day by day due to industrial activities which is a worldwide problem. Industries are polluting the environment directly or indirectly [1]. Air, water, land, living organisms, plants, animals etc. get affected due to pollution generated through industries. When industrial wastewater is discharged in water stream, it is dangerous to aquatic life and human beings. Textile industries are one of the major sources of water pollution. The textile dye industry effluents contains organic and inorganic salts, dyes, and heavy metals [2,3]. Moreover, 10–15% of the dyes used in textile industry enter into the environment as wastes [4,5]. As textile wastewater is toxic, mutagenic and carcinogenic due to presence of dyes [6,7], it is undesirable for any use. The physicochemical methods like adsorption [8–10],

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coagulation [11] and membrane processes [12], electrochemical separation methods [13–15] and biological treatment methods [16,17] are found effective for the removal of dyes from water medium, while these methods are in-effective for the mineralization of dyes and other recalcitrant compounds present in textile wastewater [18].

Advanced oxidation processes (AOPs) are the effective and efficient method of treating water and wastewater. AOP is used for removing toxic and non-biodegradable matters present in wastewater [19]. As compared to conventional treatment techniques AOPs are more efficient and capable of degrading recalcitrant organic pollutants. In AOPs organic matters are removed by oxidation through generated hydroxyl or sulfate radicals [20,21]. AOPs such as ozonation [22,23], photocatalysis [24–26], Fenton [27,28], electro-Fenton [29–31], anodic oxidation [32,33], photo-Fenton [34,35], peroxicoagulation [36,37], persulfate oxidation [28,38], sonochemical methods [39,40], hydrodynamic cavitation [41–43] etc. were found very effective for the complete mineralization of dyes from water medium and treatment of real textile wastewater. Performances of these AOPs for dye removal from aqueous medium are well reviewed and documented [15,44–49].

Zero-valent metal based AOPs are emerging field in water and wastewater treatment. Different kind of zero-valent metals like iron, nickel, copper, aluminium, magnesium, etc. are available for the treatment of water and wastewater [50]. Similar to other heterogeneous AOPs, zero-valent metal based AOPs have several advantages over homogeneous AOPs such as reusability and effective operation in wide pH conditions [45]. From last decade, zero-valent aluminium (ZVAl) got attention for treating contaminated water [51]. Bokare and Choi [52] reported in first time the ZVAl-based AOP for water and wastewater treatment. It has been reported the ZVAl-based AOPs are effective for the removal of various water and wastewater pollutants like 4-chlorophenol [52], triton X-45 [53], phenol [52], iopamidol [54], dichloroacetate [52], nitrobenzene [52], etc.

Textile wastewater treatment capacity of various ZVAl-based AOPs was checked in the present study. ZVAl/O_2, ZVAl/Fe^{3+}/O_2, ZVAl/Fe^{3+}/O_2/H_2O_2 and ZVAl/Fe^{3+}/O_2/persulfate processes were considered. The performance of ZVAl was checked on the basis of COD, colour and ammoniacal nitrogen removal.

2. Materials and methods

2.1. Materials

Aluminium fine powder (98% extra pure) from LobaChemie, ferric chloride, potassium persulfate, sulfuric acid and hydrogen peroxide from Merck were used for the experiments. All the chemicals were used without further purification. Deionized water was used for preparing reagents.

2.2. Textile wastewater

Raw textile wastewater was collected in a 20 L capacity plastic container from a textile industry situated in Ahmadabad, Gujarat, India. The textile wastewater was characterised as per standard methods [55]. Colour of wastewater was monitored at 500 nm using Microprocessor UV–visible spectrophotometer (LT – 290, Labstronics, India).

2.3. Experimental procedure

Initially, pH of textile wastewater was maintained to the required level using 1 N sulfuric acid. 500 mL of the solution was taken in a 1000 mL glass beaker. Required amount of ZVAl was added and aeration provided using an aquarium pump. ZVAl/O_2 process experiments were carried out by varying ZVAl dosages as 1 g/L, 4 g/L and 6 g/L. In order to find the best combination and efficiency, both ZVAl dosage and ferric ion concentration were varied for ZVAl/Fe^{3+}/O_2 process. ZVAl dosages of 1 g/L and 2 g/L were considered, while ferric ion concentrations were changed from 0.5 g/L to 2 g/L. ZVAl/Fe^{3+}/O_2/H_2O_2 and ZVAl/Fe^{3+}/O_2/persulfate processes were carried out by changing the concentrations of hydrogen peroxide (1.3 g/L, 4 g/L, 5.3 g/L and 6.7 g/L) and persulfate (0.18 g/L, 0.36 g/L, 0.75 g/L and 1 g/L), respectively for the optimal operating condition of ZVAl/Fe^{3+}/O_2 process. Aeration was continued till the end of the experiments. Samples were collected at regular interval and the removal efficiency of process was monitored by checking residual COD of treated wastewater. All the experiments were conducted at room temperature. COD removal efficiency, colour reduction and ammoniacal nitrogen removal efficiency were determined using following formula:

$$\text{COD removal efficiency}(\%) = \frac{\textit{Initial COD}-\textit{COD at time t}}{\textit{Initial COD}} \times 100 \tag{1}$$

$$\text{Colour reduction} = \frac{\textit{Absorbance of sample at time t}}{\textit{Absorbance ofwastewater}} \tag{2}$$

$$\text{Ammonical nitrogen removal efficiency}(\%) = \frac{\textit{Initial concentration}-\textit{Final concentration}}{\textit{Initial concentration}} \times 100 \tag{3}$$

3. Result and discussions

3.1. Characteristics of textile wastewater

The characteristics of textile wastewater are: pH 7.10–7.80, COD 2155 mg/L, BOD 500 mg/L, ammoniacal nitrogen 699 mg/L, chloride 1463 mg/L, total dissolved solids 5190 mg/L and total suspended solids 513 mg/L. BOD/COD ratio of raw wastewater was found as 0.23. Thus, biological wastewater treatment methods are not a feasible solution. AOPs are the best option to treat such types of wastewater as mentioned earlier.

3.2. ZVAl/O_2 process

ZVAl can generate hydroxyl radicals as in the Eqs. (4) and (5). In acidic medium and in the presence of dissolved oxygen, ZVAl generates hydrogen peroxide in the water medium and further ZVAl reacts with in-situ generated hydrogen peroxide to produce hydroxyl radicals. These reactions are reported in first time by Bokare and Choi [52]. Lin et al. [56] reported that pH 2 is the ideal conditions for generating hydrogen peroxide by aerating ZVAl and subsequent production of hydroxyl radicals. Similarly, Zhang et al. [57], Liu et al. [58] and Fan et al. [59] observed higher pollutant removal efficiency for ZVAl/O_2 process at pH near to 2. Similar to the previously published articles, COD removal from textile wastewater by ZVAl/O_2 process was examined at pH 2 and the observed results are shown in Fig. 1. Initially the experiments were carried out at ZVAl dosage of 1 g/L and 33.9% of COD removal was observed at the end of 3 h reaction time. Experiments were also carried out to test the COD removal by aeration and ZVAl without aeration. Aeration is efficient to remove 25% of COD from textile wastewater. COD removal efficiency of ZVAl (in the absence of air) was observed as 21.3%, which is significantly lesser than aerated process. The reason behind lower efficiency of ZVAl without aeration is mainly attributed to lesser amount of dissolve oxygen in the water medium and which leads to the lower hydrogen peroxide and hydroxyl radical productions. These results indicate the importance of oxygen for the generation of hydroxyl radicals, as explained by Bokare and Choi [52]. The authors observed insignificant removal of 4-chlorophenol in the absence of oxygen and the removal was enhanced significantly with the aeration.

$$2Al^0 + 3O_2 + 6H^+ \rightarrow 2Al^{3+} + 3H_2O_2 \tag{4}$$

$$Al^0 + 3H_2O_2 \rightarrow Al^{3+} + 3OH^- + 3HO^{\cdot} \tag{5}$$