

Acid Red 18 Dye Removal from Aqueous Solution Using Combined Process of Coagulation and Nanoparticle Zero Valent Iron (nZVI)

Babak Goudarzi¹, Hasan Pasalari², Kavoos Dindarloo¹, Hamid Reza Ghaffari¹,
Vali Alipour*¹, Abdolhamid Tajvar³, Amin Ghanbarnejad⁴

¹Department of Environmental Health Engineering, Faculty of Health, Hormozgan University of Medical Sciences, Bandar Abbas, Iran.

²Department of Environmental Health Engineering, School of public Health, Iran University of Medical Sciences, Tehran, Iran.

³Department of Occupational Health Engineering, Faculty of Health, Hormozgan University of Medical Sciences, Bandar Abbas, Iran

⁴Department of Public Health, Faculty of Health, Hormozgan University of Medical Sciences, Bandar Abbas, Iran.

*Corresponding author: E-mail: v.alipour53@gmail.com,

ABSTRACT

The color decrease aesthetical quality of drinking water and consumer's tendency for using this water. Furthermore, colored water is unsuitable for industrial usage. Color producing material may react with chlorine compounds and creates carcinogen disinfection by products (DBPs). The aim of this study was to investigate Acid Red 18 dye removal from aqueous solution by combined conventional coagulation and nanoparticle zero valent iron (nZVI) process. The experiments were conducted in jar test device and efficiency removal was evaluated by changing alum concentration, nZVI dosage, and pH. On the basis of the results, removal efficiency for integrated process was higher than only conventional coagulation. The color removal efficiency for integrated process and coagulation were 93 and 47 percent, respectively. Integrated process can be used effectively for color removal from water in regions with color problems. More studies are needed to investigate other aspects of this process.

Keywords: Coagulation, nZVI, Acid Red 18

Date of Submission: 26-09-2017

Date of Accepted: 06-10-2017

I. INTRODUCTION:

Colored waters cause aesthetical problems in water supply systems; so water resources polluted with color could not be applied in some industries such as paper, food processing, dairy, textile, and plastic. Furthermore, in some cases, color producing compounds is likely to be the precursors of carcinogenic disinfection by products (DBPs) which are made in reaction with chlorine. Even corrosion of metal and concrete pipes is also can attributed to existence of color in water [1-4].

Due to health problems related to colored waters, World Health Organization (WHO) recommends the guideline of 15 TCU for drinking water. Over this value the color is clearly visible in water [5]. Color in the water can be originated from many sources and composites such as iron and manganese ions, humic acid, fulvic acid, tannin, lignin, decayed plant materials, algae, and industrial wastewater [8-6]. There are two main types of color, depending on their origins, which are called appearance and true color. Appearance color is typically from colloidal and suspended solids and can be filtered easily. In contrast, true color is produced by soluble materials, which are not removed and deposited using unit operations like filtration. Therefore, it is required to use chemicals to degrade and remove this color [9, 6].

Several methods have been applied to remove color from aquatic solutions. The most common processes include coagulation, adsorption, chemical oxidation, nanofiltration and biological processes [13-10]. Advanced coagulation in which more can remove organic matter, color, and metals simultaneously. However, addition a coagulant over the typical dosage can cause large amount of sludge, extreme alkalinity consumption, decreasing in pH, corrosion, and high-cost operation in the water treatment facilities.

Zero-valent iron nanoparticle (nZVI) has been proved to be a potentially useful in removal of a wide range of contaminants and pollutants including pesticides, insecticides, chlorofluorocarbons (CFCs), chromium, lead, arsenic, and nitrate [16-14]. The most important advantages with iron particle are low cost, no toxicity, and more availability in comparison to other metals [17, 18]. Some previous studies have investigated dye removal from aqueous solution using iron nanoparticle [19-22]. The present study was intended to investigate integrated coagulation and nZVI process in color removal from aqueous solution.

II. MATERIALS AND METHODS

2.1. Synthesis of iron nanoparticle and characterization

In order to synthesize nZVI, 500 ml of 0.1 M ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) solution was prepared. Afterwards, a 0.25 molar sodium borohydride (NaBH_4) solution was added dropwise to the ferric chloride suspension for 60 minutes. After this reaction, the iron nanoparticles were deposited as black particles. The nZVI was separated by filtration and then dried at 180°C for 1 hour and stored in a refrigerator until the time of the experiment. The SEM image of synthesized nanoparticle was prepared with FESEM microscope (Hitachi model IS4160). Fourier transform infrared (FTIR) transmission spectra was obtained to characterize the surface groups on the synthesized iron nanoparticles

2.2. Coagulation experiments

Acid Red 18 dye powder was employed to prepare the synthetic dye solutions. The chemical structure of Acid Red 18 was shown in figure 1. Aluminum sulphate (alum) was used as main coagulant. The Jar test device was applied to conduct experiments. Experiments were done in three stages including rapid mixing (200 rpm and 2 min), flocculation (75 rpm and 15 min), and sedimentation (30 min). nZVI was added to the reactor in rapid mixing stage. After settling and deposition, the supernatant was extracted from Jar test container and residual dye solution was measured using UV-visible spectrophotometer.

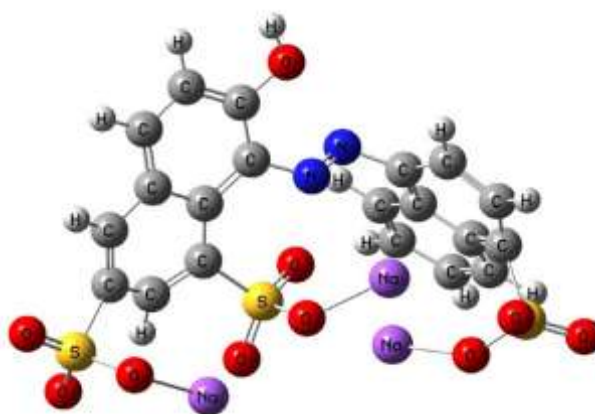


Fig.1-Molecular structure of Acid Red 18[23]

2.3. Process Optimization

To determine the optimum pH, experiments were conducted at different pH (1, 3, 5, 7, 9, and 11) and at fixed alum (50 mg/L) and dye (50 mg/L) concentrations. To specify the optimum concentration level of alum, dye solution with concentration of 50 mg/L at optimum pH obtained in previous stage was placed in contact with different concentrations of alum (10, 20, 30, 40, and 50). Combined coagulation and nZVI process was conducted at different dosage of nZVI (0.25, 0.50, 0.75, 1, 1.5 gr) and optimal value of pH and alum concentration and fixed concentration of dye (50 mg/L). After completing the process, removal efficiency (RE) was calculated as follows [24-26]:

$$RE (\%) = \left(\frac{c_0 - c_1}{c_0} \right) \times 100$$

Where c_0 and c_1 are initial and final concentration, respectively.

III. RESULTS AND DISCUSSION

3.1. Characterization of synthesized nanoparticles

As can be seen from the SEM image (Figure 2), synthesized nanoparticles are formed in rod and irregular shapes with different diameter less than 100 nm. This micrograph shows that the ZVI nanoparticles are not in the discrete particles form. The most probable explanation for this aggregation is the magnetic forces between iron particles. Figure 3 shows XRD diagram of nZVI. As can be seen from this figure, the most obvious peaks for synthesized nanoparticles are FeOOH and Fe⁰ groups.

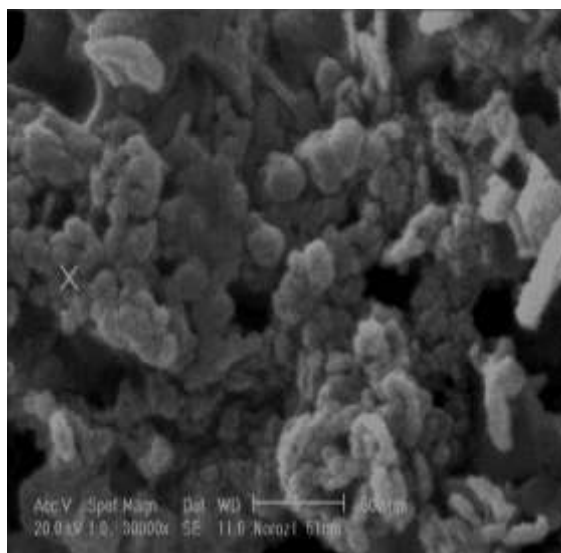


Figure 2: SEM image of synthesized nZVI

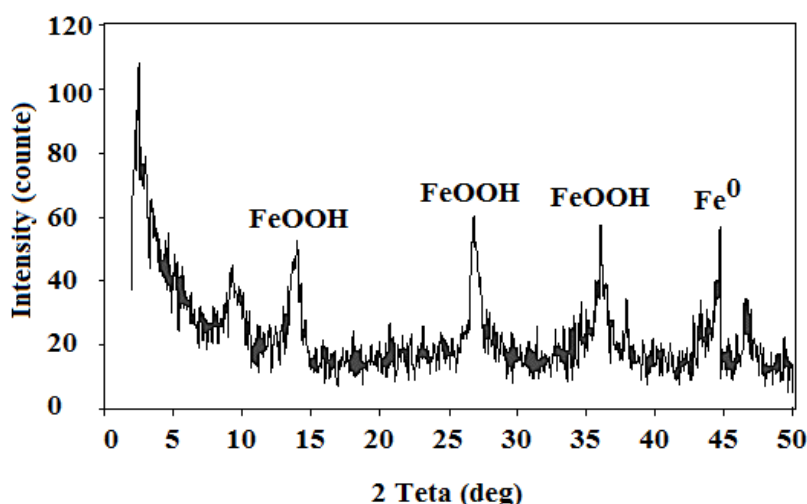


Figure 3: XRD pattern of Iron Nanoparticles

3.2. Effect of pH on the removal efficiency

In the present research, the effect of pH on dye removal was examined at fixed alum (50 mg/L) and dye concentration (mg/L). As can be seen from figure 4, increase in pH value result in decrease in removal efficiency. At acidic condition coagulant produce more positively hydrolyzated species and attractive force between these species and negative dye result in higher efficiency in comparison with alkaline condition. The results of this study is in agreement with Klimiuk et al. study in which the optimum pH for reactive dyes removal from aqueous solution using coagulation was found to be at acidic range [12].

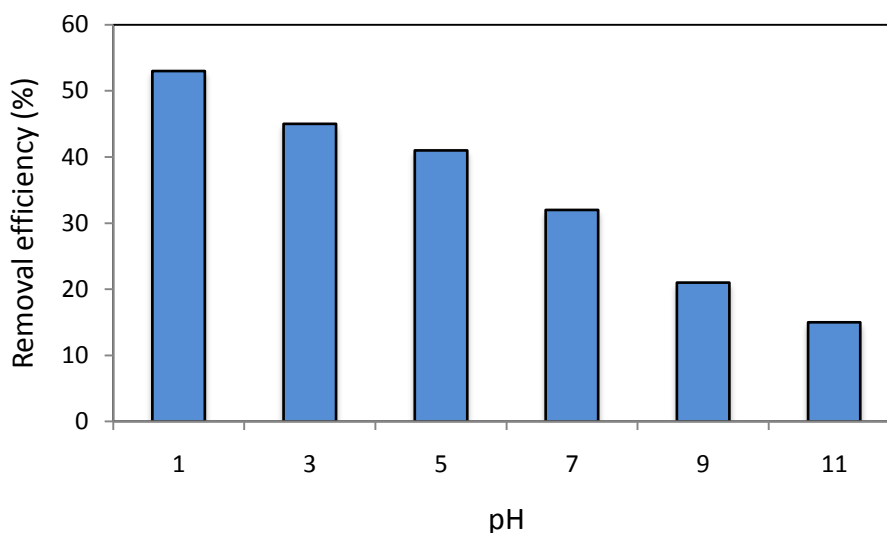


Figure 4: Effect of pH on Acid Red 18 removal

3.3. Effect of alum concentrations on the removal efficiency

Different initial alum concentrations (10, 20, 30, 40, and 50 g/L) were considered in the present work to determine the optimum alum concentration at fixed value of pH (3) and dye concentration (50 mg/L). The results of these experiments were shown in figure 5. As can be seen from this Figure, increase in alum concentration from 10 to 50 mg/L leads to increases in the removal efficiency approximately from 23 to 47 %. This results are in line with results of Malakootian et al. study. They showed that increase in coagulant dosage end in increase in efficiency of dye removal from aqueous solution. However, the level of efficiencies obtained in this study is different from those of our study [13].

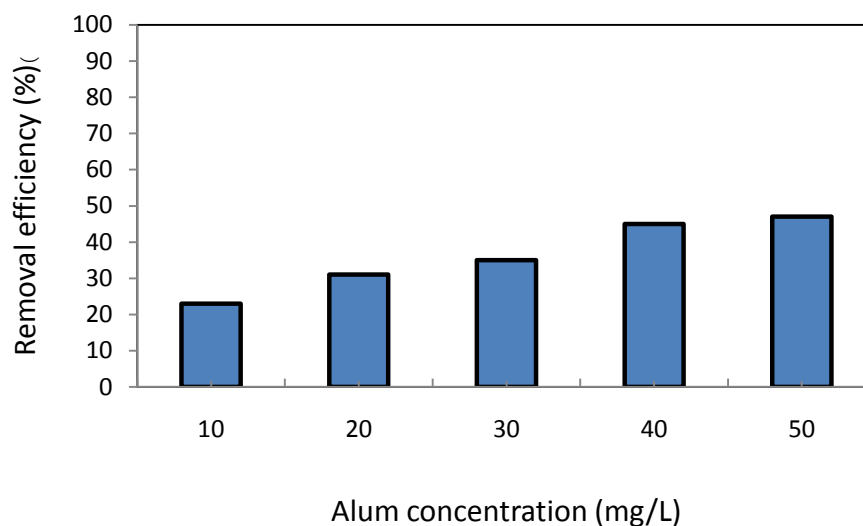


Figure 5: Effect of alum concentration on Acid Red 18 removal

3.4. Effect of combined nZVI-coagulation on the removal efficiency

In the present study in order to improve the removal efficiency of acid red 18 in coagulation process, iron nanoparticles in different dosages (0.25, 0.5, 0.75, 1, and 1.5 g/L) were added to the solution at rapid mixing stage with initial dye concentration of 50 mg/L at pH value of 4. Figure 6 shows the removal efficiencies obtained from simultaneous application of iron nanoparticles and alum in the coagulation process, as can be seen from figure 6, the removal efficiency is increased by adding iron nanoparticle so that removal efficiency is increased from 55 to 93% as nZVI dosage is increased from 0.25 to 1.5 g/L which is significantly higher than coagulation with alum as the only coagulant. At the same condition, removal efficiency of coagulation with

alum is 47%. The most probable mechanism for dye removal by nZVI is adsorption process in which negatively charged acid red 18 is adsorbed on nZVI byproducts produced in aqueous solution.

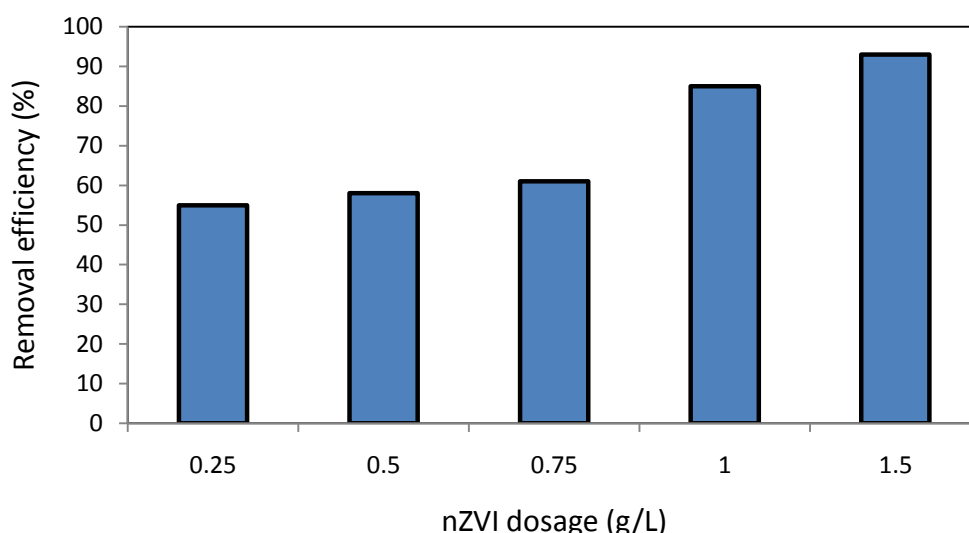


Figure 6: Effect of combined coagulation-nZVI process on Acid Red 18removal

IV. CONCLUSION

This study aimed to investigate the utility of iron nanoparticles as auxiliary coagulant for Acid Red 18removal from aqueous solution in the coagulation process. The results of this study indicated that the iron nanoparticles can be used as a suitable coagulant aid for color removal from aqueous solution. The main advantage with this process is that there is no need to add new equipment to the water treatment plant. In addition, iron nanoparticles are one of the most widely consumed nano-materials in water treatment due to the cheapness, safety, and easy availability. The main challenge in using the nanotechnology process in water treatment is the fate of nanoscale materials used in treatment process. Therefore, the use of this technology requires further studies.

ACKNOWLEDGMENT

Authors gratefully acknowledge the support received from Hormozgan University of Medical Sciences.

REFERENCE

- [1]. Fazli, M.M., et al., *Optimization of reactive blue 19 decolorization by ganoderma sp. using response surface methodology*. Iranian Journal of Environmental Health Science & Engineering, 2010. **7**(1): p. 35.
- [2]. Kim, S., et al., *COD reduction and decolorization of textile effluent using a combined process*. Journal of Bioscience and Bioengineering, 2003. **95**(1): p. 102-105.
- [3]. Rahmani, A., et al., *Degradation of Azo Dye Reactive Black 5 and acid orange 7 by Fenton-like mechanism*. Iranian Journal of Chemical Engineering, 2010. **7**(1): p. 87-94.
- [4]. Mirzaei, N., et al., *Survey of effective parameters (water sources, seasonal variation and residual chlorine) on presence of thermotolerant coliforms bacteria in different drinking water resources*. International Journal of Pharmacy and Technology, 2015. **7**(3): p. 9680-9689.
- [5]. Organization, W.H., *Guidelines for drinking-water quality*. Vol. 1. 2004: World Health Organization.
- [6]. Chermisinoff, N., *Handbook of water and wastewatertreatment technologies*. 2002: Butterworth – Heinemann.
- [7]. Kawamura, S., *Integrated design and operation of water treatment facilities*. Vol. 2. 2000: John Wiley and sons.

- [8]. Zarrabi, M., et al., *REMOVAL OF PHOSPHORUS BY ION-EXCHANGE RESINS: EQUILIBRIUM, KINETIC AND THERMODYNAMIC STUDIES*. Environmental Engineering & Management Journal (EEMJ), 2014. **13**(4).
- [9]. Degremont, O., *Water treatment handbook*. Vol. 6. 2002: Lavoisier Wiley.
- [10]. Nabi Bidhenhi, G.R., et al., *Evaluation of industrial dyeing wastewater treatment with coagulation and polyelectrolyte as a coagulant aid*. Iran. J. Environ. Health. Sci. Eng, 2007. **4**(1): p. 29-36.
- [11]. Naimabadi, A., H. Movahedian Attar, and A. Shahsavani, *Decolonization and biological degradation of azodye reactive red2 by anaerobic/aerobic sequential process*. Iran. J. Environ. Health. Sci. Eng, 2009. **6**(2): p. 67-72.
- [12]. Klimiuk, E., U. Filipkowska, and A. Korzeniowska, *Effects of pH and coagulant dosage on effectiveness of coagulation of reactive dyes from model wastewater by polyaluminium chloride (PAC)*. Polish Journal of Environmental Studies, 1999. **8**: p. 73-80.
- [13]. Malakootian, M. and A. Fatehizadeh, *Color removal from water by coagulation/caustic soda and lime*. Journal of Environmental Health Science & Engineering, 2010. **7**(3): p. 267-272.
- [14]. Rahmani, A.R., H.R. Ghaffari, and M.T. Samadi, *Removal of Arsenic (III) from Contaminated Water by Synthetic Nano Size Zerovalent Iron*. world academy of sciences, engineering and technology, 2010. **62**.
- [15]. Ponder, S.M., J.G. Darab, and T.E. Mallouk, *Remediation of Cr (VI) and Pb (II) aqueous solutions using supported, nanoscale zero-valent iron*. Environmental Science & Technology, 2000. **34**(12): p. 2564-2569.
- [16]. Xiong, Z., D. Zhao, and G. Pan, *Rapid and complete destruction of perchlorate in water and ion-exchange brine using stabilized zero-valent iron nanoparticles*. Water research, 2007. **41**(15): p. 3497-3505.
- [17]. Miri, M., et al., *Ecological risk assessment of heavy metal (HM) pollution in the ambient air using a new bio-indicator*. Environmental Science and Pollution Research, 2016. **23**(14): p. 14210-14220.
- [18]. Miri, M., et al., *Investigation of outdoor BTEX: Concentration, variations, sources, spatial distribution, and risk assessment*. Chemosphere, 2016. **163**: p. 601-609.
- [19]. Chompuchan, C., et al., *Decolorization of Reactive Black 5 and Reactive Red 198 using nanoscale zerovalent iron*. World Acad Sci Eng Technol, 2009. **49**: p. 130-134.
- [20]. Shih, Y.-H., C.-P. Tso, and L.-Y. Tung, *Rapid degradation of methyl orange with nanoscale zerovalent iron particles*. Nanotechnology, 2010. **7**: p. 16-7.
- [21]. Shu, H.-Y., et al., *Reduction of an azo dye Acid Black 24 solution using synthesized nanoscale zerovalent iron particles*. Journal of colloid and interface science, 2007. **314**(1): p. 89-97.
- [22]. Alahabadi, A., et al., *A comparative study on capability of different tree species in accumulating heavy metals from soil and ambient air*. Chemosphere, 2017. **172**: p. 459-467.
- [23]. Mirzaei, N., et al., *Modified natural zeolite using ammonium quaternary based material for Acid red 18 removal from aqueous solution*. Journal of Environmental Chemical Engineering, 2017.
- [24]. Pasalari, H., et al., *Activated carbon derived from date stone as natural adsorbent for phenol removal from aqueous solution*. DESALINATION AND WATER TREATMENT, 2017. **72**: p. 406-417.
- [25]. Sepehr, M.N., et al., *Potential of waste pumice and surface modified pumice for hexavalent chromium removal: Characterization, equilibrium, thermodynamic and kinetic study*. Journal of the Taiwan Institute of Chemical Engineers, 2014. **45**(2): p. 635-647.
- [26]. Rahmani, A., et al., *Synthesis of Zero Valent Iron Nanoparticles (nZVI) and its Efficiency in Arsenic Removal from Aqueous Solutions*. Water Wastewater, 2011. **1**: p. 35-41.

Babak Goudarzi. "Acid Red 18 Dye Removal from Aqueous Solution Using Combined Process of Coagulation and Nanoparticle Zero Valent Iron (nZVI) adsorption." The International Journal of Engineering and Science (IJES), vol. 6, no. 9, 2017, pp. 74-79