Effects of gamma irradiation on color removal from reactive red 24 aqueous solutions

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ABSTRACT

Decolorization from synthetic wastewater containing Reactive Red 24 (RR24) using gamma Cobalt-60 ray was investigated. The influence of the irradiation dose, initial pH, initial dye and hydrogen peroxide (H_2O_2) concentration were evaluated. The pH effect has proved that lower decolorization efficiency was observed in alkaline medium as compared to the neutral and acid medium. While color removal efficiency increased with increasing absorbed dose. A synergistic effect of gamma radiation with H_2O_2 was applied and the results showed that H_2O_2 accelerated decolorization process, however when the dosage of H_2O_2 exceeded the optimal concentration, the color removal efficiency attained saturation and even reduced. The color removal efficiencies achieved over 99% at the dose of 4 kGy, initial pH 7, initial dye concentrationof 100 mg/L and H_2O_2 concentration of 3mM. These results highlighted the potential of radiation technology for dye removal from textile wastewater.

Keywords: *Decolorization, gamma irradiation, reactive red 24, textile wastewater.*

1. INTRODUCTION

Reactive dye-containing wastewater is one of the most difficult wastewater to treat, because most of reactive dye has complex chemical structures with high water-solubility. Moreover, the dye structures are commonly highly resistant to biodegradation process.

One of the greatest concerns in wastewater treatment of a textile effluent is the color removal. Various types of techniques have been used to eliminate color including adsorption, chemical coagulation, photodegradation, membrane, ozonation, sonolysis, etc. [1-3]. Nonetheless, most of these techniques are limited by technology, cost or difficulties in operation. Hence, they could not be employed to treat real dyeing wastewater.

Recently, ionizing radiation methods such as gamma radiation and electron beam, a kind of oxidation method which could induce an amounts of oxidizing species ($OH^{\bullet}$, H_2O_2, $HO_2^{\bullet}$…) and reducing species (e^-_{aq} and $H^\bullet$) through water radiolysis [4], received great attention for the effective treatment of toxic pollutants.

The gamma radiation has been successfully to treat slaughterhouse wastewater [5], coking wastewater [6], pesticide production wastewater [7], pharmaceutical wastewater [8] ,and also drinking water [9]. However, the ionizing radiation process normally required high irradiation dose (or irradiation time) to degrade the complex organic compounds [10-12]. In this way, there is a need for an effective and non-hazarsous method could combine with the ionizing radiation process. Hydrogen peroxide (H_2O_2) could be the key to the problem. Some reports [8, 13, 14] indicate that adding small amounts of hydrogen peroxide could improve the irradiation capacity through increasing the formation of OH radicals as follows:

$$e^-_{aq} + H_2O_2 \rightarrow {}^\bullet OH + OH^- \quad (1)$$

$$H^\bullet + H_2O_2 \rightarrow {}^\bullet OH + H_2O \quad (2)$$

Yulin et al. [15] had also demonstrated that the gamma irradiation/H_2O_2 process was more cost-effective than sole H_2O_2 or irradiation. Nonetheless, as the concentrations of H_2O_2 are exceeded the optimal value, the degradation rates are reduced due to the scavenging of OH radical formation as follows [14]:

$$H_2O_2 + {}^\bullet OH \rightarrow HO_2^\bullet + H_2O \quad (3)$$

$$HO_2^\bullet + {}^\bullet OH \rightarrow H_2O + O_2 \quad (4)$$

The inconsistency of the reports implies that synergic effects of gamma irradiation and H_2O_2 in wastewater treatment technology should be continuingly explored.

To the best of our knowledge, there has been no research on the treatment of Suncion Red P-2B (RR24) aqueous solution by gamma irradiation method reported so far. The objective of this study is to investigate the decolorization of RR24 aqueous solution using gamma Co-60 radiation in the presence and absence of H_2O_2 with dose up to 12 kGy.

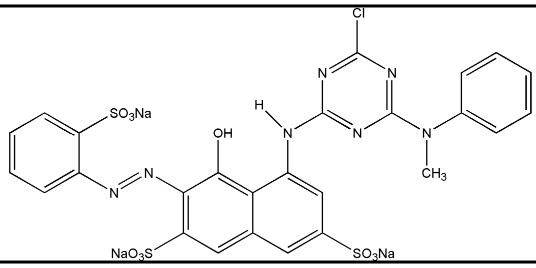
2. MATERIALS AND METHODS

Reagents and apparatus

All chemicals were reagent grade, Sigma-Aldrich and hydrogen peroxide (H_2O_2) obtained from Merck, Germany.

The commercial Suncion Red P-2B (RR24) was supplied by Oh-Young (a Korean company) with a molecular structure and characteristics as summarized in Table 1.

Table 1. Characteristics of RR24

Chemical structure	
CAS No.	70210-20-7
Chromophore	Monoazo
M_w (g/mole)	788.07
$\lambda_{max\ (nm)}$	534

The dye stock solution (1000 mg/L) was achieved by completely dissolving 1 gram of dye powder into 1 liter of distilled hot water at pH 11 for an hour to get the dye stock in the “hydrolyzed” form, and the solution was diluted to appropriate concentrations (30-200 mg/L) before being used.

Procedures and analysis methods

Firstly, 1000 mL beaker containing 500 mL of the dye solution with specific concentration (30, 50, 80, 120, 150, 180, and 200 mg/L) were adjusted to the different pH values (1, 2, 3, 5, 7, 9, and 11) by NaOH or HCl (0.05 N) solutions. Secondly, an amount of H_2O_2 was added to reach the solution desired H_2O_2 concentration (0, 1, 2, 3, 4, and 5 mM). Then 400 mL of the solution were poured into 500 mL glass bottle (Schott, Germany), packed with a dichromate dosimeter and irradiated with different doses of gamma rays (1 - 12 kGy) at room temperature (25 ± 1°C) on a gamma Co-60 irradiator (Gamma chamber GC-5000, BRIT, India) at Nuclear Research Institute, Dalat , Viet Nam.

The irradiated water samples were then settled for two hours; then filtered and determined the absorbance at maximum absorption wavelength (λ_{max}) 541 nm using spectrophotometer UV-VIS GENESYS 10 (Thermo Fisher, Germany). The degree of decolorization was calculated from the decrement of absorbance at this maxium wavelength. pH was measured with a SevenEasy pH-meter (Mettler-Toledo, Switzerland). While the absorbed dose of the gamma rays were validated using the dichromate dosimetry method [16]. All analyses were conducted in triplicate and results presented here are the mean values ± standard deviations.

3. RESULTS AND DISCUSSION

Influence of rrradiation dose

In the gamma irradiation process, irradiation dose determines the hydroxyl radical formation rate hence affects the decolorization efficiency [4]. In order to examine the effect of irradiation dose on decolorization, the experiment was carried out using various irradiation doses from 0 to 12 kGy at pH 9, 100 mg/L of dye concentration with no adding H_2O_2.

As expected, the increasing irradiation dose had a dramatic impact on the color removal, there was approximately a linear relationship between irradiation dose and color removal of dye (*Figure 1*). Also, high rate of color removal efficiencies were obtained when irradiation dosage increment from 0 to 4 kGy. It may due to the hydroxyl radical increased in dye solution with the increase of irradiation dose [4] and finally attains saturation when equilibrium (between the dye and radical) was reached [8]. These results are similar to the irradiation dose recommended by Guo and Shen [6]. So, the suitable irradiation dose of 4 kGy was selected for the next experiment.

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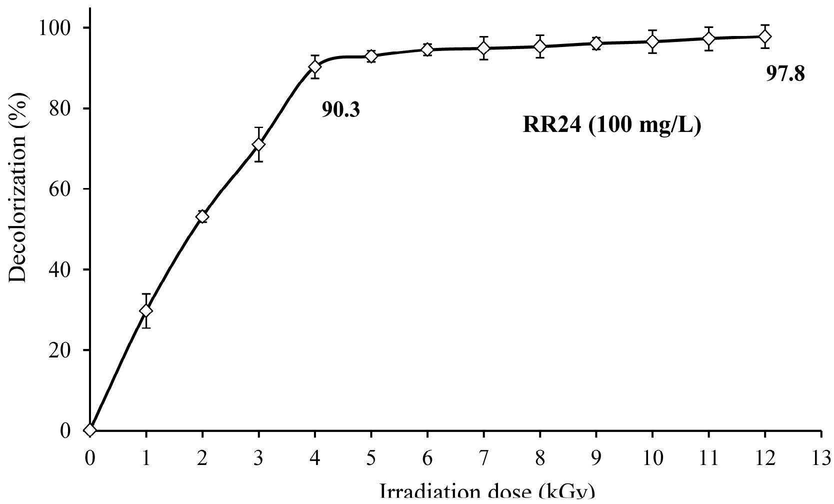

Figure 1. Effect of irradiation dose on removal efficiency at pH 9 without H_2O_2

Influence of initial dye concentration

The effect of initial dye concentration (IDC) on the color removal of the irradiation experiment was investigated. IDC was from 30 to 200 mg/L and irradiated with the fixed irradiation dose 4 kGy at pH 9 without adding H_2O_2. The results of color removal efficiency in *Figure 2* indicated that the IDC upped to 100 mg/L, the irradiation capacity was not exhausted and the degree of color removal was slightly reduced from 98.6 ± 1.0 % (30 mg/L) to 90.8 ± 1.7 % (100 mg/L). However, beyond 100 mg/L, the degree of the color removal was sharply decreased. The reason may be due to the IDC is rather high.

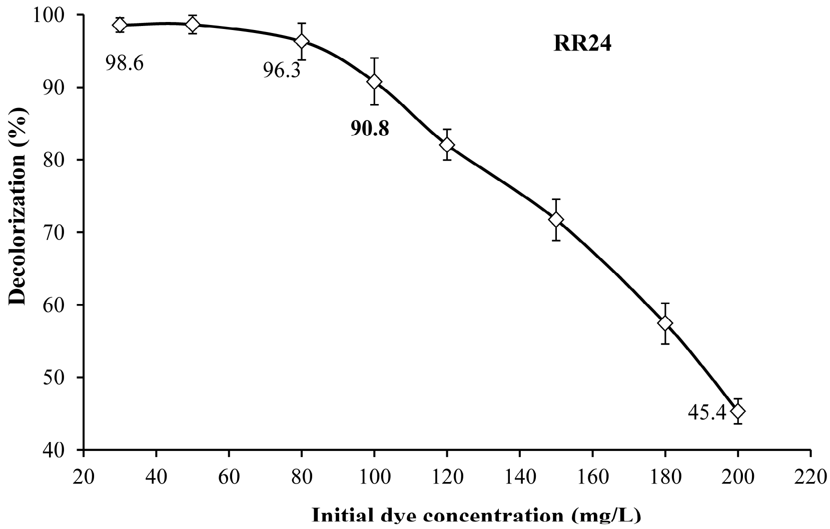

Figure 2. Effects of IDC on removal efficiency at pH 9 and dose 4 kGy without H_2O_2

Effect of pH

It has been established that pH plays an important role in impacting on the performance of dye irradiation process because it could affect both the specialization of dye [17] and the formation of radicals which could react with pollutants [4]. A series of experiments were carried out by varying the pH from 1 to 11 by using 0.05 N NaOH (or HCl), with IDC of 100 mg/L and irradiation dose of 4 kGy. The color removal of the dye solution was influenced by pH in *Figure 3* showed that the decolorization efficiency decreased with the increasing of initial pH from 1 (99.6 ± 0.4%) to 11 (89.4± 0.6%). This may be due to the dissociation ability of dyeing substances at organic phase is increased at low pH [17] and the dye could react efficiently with hydroxyl radical at lower pH value [4]. Similar trend was also reported in the research of Dessouki et al. [7] for eliminating of pesticides.

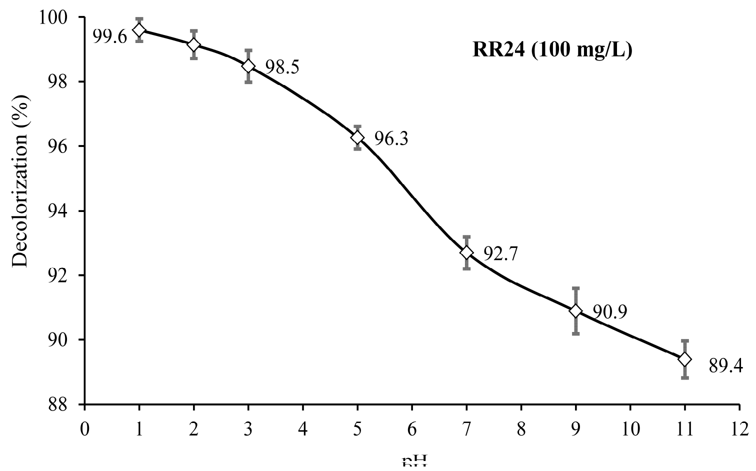

Figure 3. Effect of pH on color removal efficiency at 4 kGy of irradiation dose without H_2O_2

Although the lower pH was, the higher color removal efficiency achieved, but in fact, it needs a great amount of neutralizing acids consumption to justify pH from 9 (normal value of waste water source after the reactive dyeing processes) down to pH of 1 or 2. Furthermore, in the acidic environment, the corrosion likelihood of irradiation and related facility often occurs. Therefore, pH 7 was typical for further investigation.

Effect of hydrogen peroxide concentration

Prerious studies [7, 8] reported that the presence of H_2O_2 in the solution could lead the formation of hydroxyl radical which contribute the treatment efficiency. In order to investigate the synergistic decolorization of gamma Co-60 and H_2O_2, a series of experiments were perfromed at different concentration of H_2O_2 from 0 to 5 mM. The results obtained were presented in *Figure 4*.

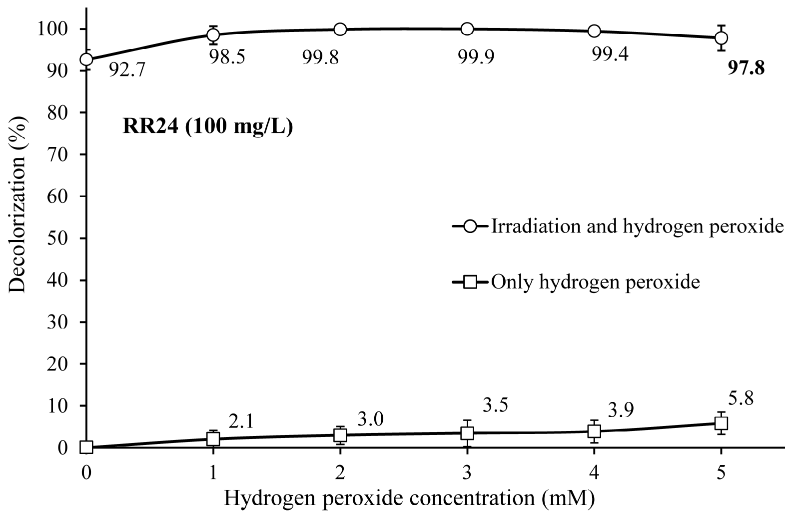

Figure 4. Effects of IDC on removal efficiency at pH 7 and 4 kGy of irradiation dose

Results in *Figure 4* illustrated that color removal efficiency depended remarkably on H_2O_2 concentration. The decolorization percentage for the H_2O_2 alone increased only to 5.8 ± 2.7% at the highest H_2O_2 concentration (5 mM). Whereas, as combining irradiation with H_2O_2, color removal degree increased from 92.7 ± 2.4% (without H_2O_2) to 99.9 ± 0.1% (3 mM H_2O_2). The combined effect of H_2O_2 and radiation was much higher than the effect of either component, or even of the sum of the individual effect. These results mean that irradiation and H_2O_2 induced the decomposition of dye efficienly because of the abundant hydroxyl radical produced from irradiation of dye solution in the presence of H_2O_2 as described in equations 1and 2 [4]. Nevertheless, when H_2O_2, concentration was in the range of 3-5 mM, the color removal degree dropped slightly from 99.9 ± 0.1% (3 mM) to 97.8 ± 3.0% (5 mM). This phenomenon may be due to an excessive H_2O_2 promotes an inhibitory effect (hydroxyl radical scavenging) and the formation of another radical ($HO_2^•$), having an oxidation potential considerably smaller than $HO^•$ as described in equations 3 and 4 [4].

4. CONCLUSIONS

Gamma Co-60 irradiation proved to be an effective method for decolorization of reactive red 24 (RR24) dye solution. The color removal of the dye solution was almost 99.9 ± 0.1% at the initial dye concentration of 100 mg/L and absorbed dose of 4 kGy. The synergistic effect of gamma Co-60 irradiation and H_2O_2 was found out at H_2O_2 concentration of 3 mM as suitable one. Thus, radiation technology is considered as a new method for decolorization of textile wastewater.

Nghiên cứu loại màu dung dịch nhuộm hoạt tính Red 24 bằng bức xạ Gamma Co-60

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TÓM TẮT

Nghiên cứu giảm màu trong dung dịch nhuộm hoạt tính reactive red 24 bằng bức xạ gamma Co-60 được thực hiện. Ảnh hưởng của liều xạ, pH, nồng độ màu và nồng độ H_2O_2 được khảo sát. Kết quả cho thấy hiệu suất khử màu tăng khi tăng liều xạ, pH môi trường acid hoặc trung tính có hiệu quả xử lý cao hơn trong môi trường kiềm. Hiệu ứng đồng vận của bức xạ với H_2O_2 cũng cho thấy H_2O_2 thúc đẩy quá trình khử màu bằng chiếu xạ, tuy nhiên khi nồng độ H_2O_2 vượt quá nồng độ tối ưu (3 mM) lại làm giảm hiệu quả khử màu. Hiệu quả khử màu bằng chiếu xạ đạt 99,9 ± 0,1% tại pH 7, nồng độ H_2O_2 3 mM, nồng độ màu nhuộm 100 mg/l và liều xạ 4 kGy. Kết quả nghiên cứu cho thấy phương pháp chiếu xạ rất có hiệu quả trong việc giảm màu trong nước thải nhuộm hoạt tính.

Từ khóa: *H_2O_2, bức xạ gamma Co-60, màu nhuộm red 24, nước thải nhuộm hoạt tính.*

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