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Degradation of AO Dye Wastewater by $Cu_{46}Zr_{45}Al_7Y_2$ Metallic Glasses Activated Sodium Persulfate

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Abstract: For the purpose of attenuating the environmental and public health hazards posed by azo dye wastewater, the degradation pathway and effect of Metallic glasses on printing and dyeing wastewater are explored. In this paper, $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses is used to activate sodium persulfate to achieve the purpose of degradation of acid orange dye. At the same time, the effects of different environmental factors on the degradation effect are explored, and the cycle stability and degradation principle of Cu-based Metallic glasses are preliminarily explored. The results show that when the starting concentration of acid orange solution is 50 mg/L, the dosage of $Cu_{46}Zr_{45}Al_7Y_2$ band is 2 g/L, the dosage of sodium persulfate is 4 g/L, the pH is 2, the temperature is 35 ℃, and the degradation reaction lasted for 15 min, the degradation rate reaches 95.01%. In addition, after six cycles of degradation, the Cu-based Metallic glasses band still has good stability and corrosion resistance.

Keywords: $Cu_{46}Zr_{45}Al_7Y_2$ Metallic Glasses; Activated Sodium Persulfate; Structural Stability; Degradation Efficiency

1. Introduction

At present, wastewater is very common in industrial and agricultural production. Among them, the problem of dye wastewater pollution in textile, printing and dyeing and other light industries is worth paying attention to. Highly coordinated unsaturated chemical bonds caused by highly disordered atomic arrangement of Metallic glassess make amorphous atoms in high energy state and have high surface activity [1]. In addition, as metastable materials, Metallic glassess have higher energy levels and more catalytic active centers than corresponding crystalline materials [2], so Metallic glassess have high catalytic activity.

Sodium persulfate ($Na_2S_2O_8$) shows unique chemical properties in aqueous media. It gradually releases active substances, sulfate ions (SO_4^{2-}) and sulfate radicals (SO_4^{-}•). Due to its high reactivity and strong oxidation performance, it converts complex organic pollutants into simpler and harmless elements, such as water molecules and carbon dioxide molecules [3], through oxidation. This process not only reduces the concentration of pollutants, but also realizes the effective purification of wastewater.

Numerous transition metals and alloys show high catalytic activity, since their partially filled d-orbitals are favorable for electron transfer in both directions. However, its high chemical activity and poor corrosion resistance limit its application as an electrocatalyst in acidic or alkaline electrolyte. A well-distributed amorphous structure can achieve concurrent promotion on the intrinsic catalytic activity and operational electrochemical stability of transition metal alloy-based electrocatalysts [4]. For example, under the conditions of pH =3.0 and temperature of 295 K, Rhodamine B can be completely degraded in only 10 minutes under the action of 1.6mm H_2O_2 and 0.5g/l Fe-Si-B catalyst [5]. The developed Fe-containing metallic glass systems possess superior degradation efficiency for target pollutants, as well as exceptional structural stability against performance attenuation [6-10]. Using Fe based Metallic glasses to treat pesticide wastewater by advanced oxidation method can achieve good results [11]. In this paper, $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses is used to study the degradation effect of advanced oxidation method on dye wastewater. Zhao et al [12]. Reveals the unique performance of $Cu_{46}Zr_{45}Al_7Y_2$ amorphous metal, which shows excellent catalytic activity in the treatment of dye wastewater. They observed that this material can completely convert azo dyes in just 30 mins. It has a wide range of applications, is easy to reuse, and shows strong corrosion resistance, which is one of its key findings. Li et

al. [13] studies $Cu_{46}Zr_{44.5}Al_{7.5}Gd_2$ Metallic glasses and found that the Cu-based Metallic glasses can be reused for up to 80 times, and the degradation efficiency will not be significantly reduced.

2. Experimental

In this experiment, Cu, Zr, Al and y with purity ≥ 99.99wt.% are selected to prepare the alloy according to the nominal composition of $Cu_{46}Zr_{45}Al_7Y_2$. The master alloy is melted in a vacuum induction furnace using a graphite crucible. Among them, the vacuum degree of the smelting furnace is not more than 5 Pa, and the recoil argon protection pressure is not more than 105 Pa. after grinding and crushing the master alloy, a strip with a width of 1~4 mm and a thickness of 25~30 μm is prepared by the single roll strip casting method under the same vacuum condition. The structure of the strip is characterized by X-ray scanner (XRD).

The prepared simulated AOII dye solution is used in the experiment. The specific process is as follows: accurately weigh 0.05 g of dry aoii solid powder, put it in a beaker, add 1 L of deionized water, and magnetic stirring at room temperature until it is completely dissolved to obtain the aoii dye solution with an starting concentration of 50 mg/L. During the experiment, adjust the pH with hydrochloric acid or sodium hydroxide to prepare the acid orange simulated dye wastewater. The concentration of the solution is determined by UV VIS spectrophotometer, and the morphology of the sample before and after reaction is analyzed by scanning electron microscope. Before the experiment, the Metallic glasses strip is cleaned with anhydrous ethanol to remove the impurities on its surface. Then measure 200 mlaoii dye solution and pour it into a beaker, and then add a quantitative Metallic glasses strip for degradation reaction. Then put the beaker on the magnetic stirrer and set the stirring speed to 100 rpm for degradation reaction. During the reaction process, take 4 ml of upper clear solution every 3 mins, and measure the absorbance of the solution with UV-vis spectrophotometer at the wavelength of 350-600 nm. After the reaction, the Metallic glasses strips are collected and recycled, and then used to study the recycling performance and structural stability of the catalytic material.

The specific experimental process is as follows:

(1) When the amount of amorphous ribbons is variable, 200 mL of Simulated Acid Orange Wastewater with starting concentration of 50 mg/L is put into the reaction device, and the amount of Metallic glasses ribbons is 1-4 g/L, respectively. The effects of different ribbons on the degradation effect are investigated, and the optimal amount of amorphous ribbons is determined.

(2) When the concentration of sodium persulfate is a variable, the amount of sodium persulfate is taken as 2 - 5 g/L respectively by using the determined amount of strip, pH value, reaction temperature and starting concentration of dye. The effect of different concentrations of sodium persulfate on the degradation effect of wastewater is investigated, and the optimal concentration of sodium persulfate is determined.

(3) When the pH of the solution is variable, the pH are 2, 7 and 9, respectively. The effect of different pH on the degradation effect of wastewater is investigated, and the optimal pH of the solution is determined.

(4) When the reaction temperature is variable, the temperatures are 25℃, 35℃, 45℃, 55℃, respectively. The effect of reaction temperature on the degradation effect of dyes is observed, and the optimal reaction temperature is obtained.

(5) When the starting concentration of dye is variable, the starting concentration is set to 25 mg/L, 50 mg/L, 75 mg/L and 100 mg/L with the determined amount of band, sodium persulfate concentration, pH value and reaction temperature, respectively, to determine the optimal starting concentration of dye.

3. Results and Discussion

3.1 Impact of pH Value on Degradation Effect

Figure 1 reveals the XRD pattern of $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses.

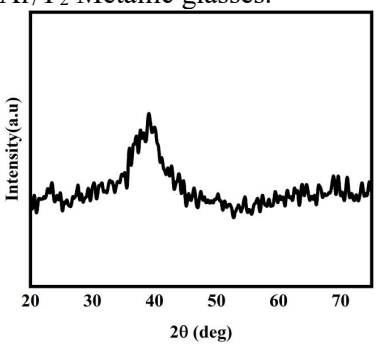


Figure 1. XRD Patterns of $Cu_{46}Zr_{45}Al_7Y_2$ Metallic Glasses Ribbons

The results reveal that $Cu_{46}Zr_{45}Al_7Y_2$ Metallic

glasses shows typical diffuse emission peak of Metallic glasses without sharp diffraction peak, and the prepared alloy strip has amorphous structure.

Figure 2 reveals the Impact of on the degradation effect. pH is a key factor in the treatment of printing and dyeing wastewater. pH will change the strength of sodium persulfate oxidation, and the actual wastewater environment will directly the pollutant treatment effect.

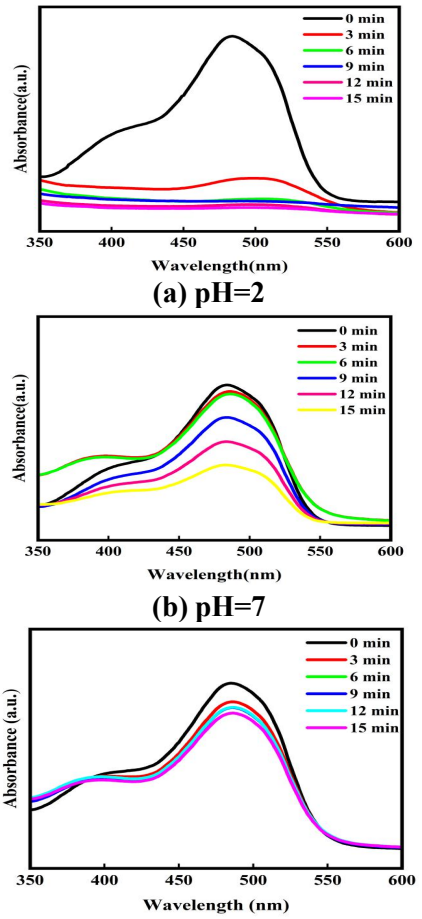


Figure 2. Effect of pH on Degradation Effect

Figure 2 shows the effect of pH on dye degradation. Under the strong acid condition of pH=2, the characteristic absorption peak of the dye decrease sharply with the extension of reaction time, and the degradation rate of the dye reaches 94.89% after 15 min ; Neutral environment with pH=7 and alkaline environment with pH=9 can significantly weaken the attenuation range of the characteristic peak. When the degradation time is 15 min, the degradation efficiency is significantly lower than that in acid environment, but azo dyes can also be degraded. This is because the characteristic absorption peak of acid orange comes from the azo bond (-N=N -) in the molecular structure [14]. When the reaction starts, the active sites distributed on the surface of the catalyst can efficiently promote the decomposition process of persulfate, thus generating SO_4^{-}• with strong oxidation activity, which can effectively break the -N=N- double bond inside the molecule, and then completely destroy the complete long-range conjugate structure of the dye molecule .

3.2 Impact of the Amount of Amorphous Ribbons on the Degradation Effect

Figure 3 reveals the effect of the amount of amorphous ribbons added on the degradation effect. When the addition amount of Metallic glasses strip is 1 g/L, the overall rise rate of the reaction curve is slow, and the degradation rate is 80.42% after 15 minutes of reaction. The results shows that the degradation efficiency increased significantly when the dosage increases to 2 g/L, and the degradation rate is 93.56% within 15min; With the further increase in the dosage of amorphous ribbons, the degradation efficiency is only slightly improved, while its growth rate gradually decreases.

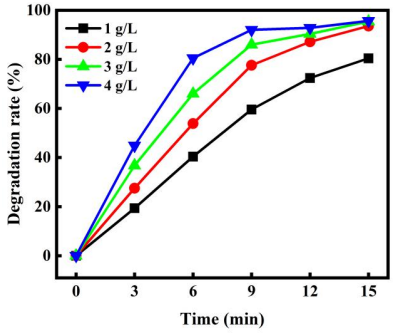


Figure 3. Effect of Addition Amount of Amorphous Ribbons on Degradation Effect

3.3 Impact of Temperature on Degradation Effect

Figure 4 reveals the effect of temperature on degradation effect. In this experiment, the initial pH=2, the dosage of sodium persulfate is 4 g/L, the dye concentration is 50 mg/L, and the dosage of amorphous ribbon is 2 g/L, and the temperatures are 25℃, 35℃, 45℃, and 55 ℃, respectively. The reaction process is slow at 25℃, and the dye degradation rate is 18.32% within 15 min; When the temperature increased to 35℃, the reaction speed is significantly accelerated, and the dye degradation rate reaches 94.36% within 15 mins, which basically realized the complete removal of dyes, and the rapid

reaction stage can be entered in 3~6 mins; The degradation rate decreased slightly when the temperature continued to rise to 45℃. Although the final degradation rate is still very high after 15 mins, the degradation effect is greatly reduced. This is because the increase of temperature can accelerate the generation of sulfate radical, increase the reactive activity of dye molecules and free radicals, and then accelerate the degradation rate. When the temperature exceeds 45℃, excessive heat will cause the invalid thermal decomposition of sodium persulfate, the loss of oxidant, and the concentration of effective components in the system will be reduced. High temperature will also accelerate the oxidation and passivation of the surface of Metallic glasses strips, and destroy the catalytic active sites [15].

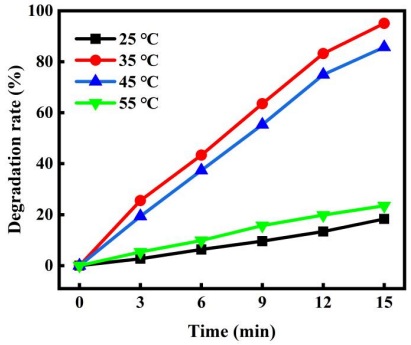


Figure 4. Impact of Temperature on Degradation Effect

3.4 Impact of Starting Concentration of Solution on Degradation Effect

Figure 5 reveals the effect of the starting concentration of the solution on the degradation rate. At a solution starting concentration is 25 mg/L, the degradation rate increased most rapidly and reaches 95.68% in 15 mins; when the starting concentration is adjusted to 50 mg/L, the degradation rate still maintained 95.03% within 15 mins; However, a distinct and significant drop in degradation performance can be detected when the starting concentration of the reaction system increased from 50 mg/L to 100 mg/L, and the degradation rates are only 47.52% in 15 mins. This is because the amount of reactive free radicals generated by sodium persulfate activation is less. When the dye concentration increases, the free radicals generated per unit time can not meet the needs of more dye molecule degradation, and high concentration dyes will also produce light shielding effect, and then inhibit the implementation of the reaction.

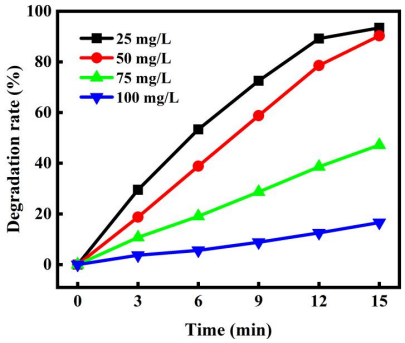


Figure 5. Impact of Starting Concentration of Solution on Degradation Rate

3.5 Impact of Sodium Persulfate Addition on Degradation Effect

Figure 6 reveals the impact of sodium persulfate addition on the degradation effect. In this experiment, the initial pH is 2, the temperature is 35 ℃, the dye concentration is 50 mg/L, and the amount of amorphous ribbon is 2 g/L. When sodium persulfate is added at 2 g/L, the degradation rate is significantly accelerated after increasing to 3 g/L, and the degradation rate is 89.58% in 15 mins; The degradation rate reaches 94.94% within 15 mins when it increased to 4 g/L; When the dosage increased to 5 g/L, the upward trend of the curve slowed down, and the final degradation rate is significantly lower than that of 4 g/L, showing a change rule of first increasing and then decreasing.

This is because the concentration of sodium persulfate has an effect on the generation and quenching of free radicals. When the dosage of sodium persulfate is 2-4 g/L, the generation of sulfate radical and oxidative degradation ability can be increased by adding sodium persulfate. However, when the concentration exceeds 4 g/L, excessive persulfate ions and free radicals will have quenching side reactions, consuming active species, and the reaction by-products will also contain the catalyst surface, inhibiting the degradation ability. 4 g/L is selected based on comprehensive consideration.

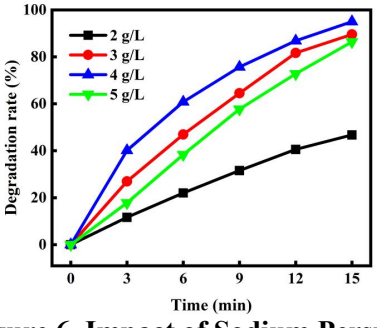


Figure 6. Impact of Sodium Persulfate Addition on Degradation Effect

3.6 Catalyst Circulation Stability Analysis

Figure 7 reveals the degradation effect of the cyclic experiment. In the practical application process of catalyst, its sustainability and reusable performance will constitute the key evaluation index. Metallic glasses has attracted attention in the field of catalysis because of its strong corrosion resistance. Based on this, the $Cu_{46}Zr_{45}Al_7Y_2$ amorphous bar after a degradation reaction is cleaned with ethanol, and then put into the wastewater under the same conditions to analyze its catalytic performance. The experimental conditions are the same: the starting concentration of acid orange is 50 mg/L, the amount of band is 2 g/L, the initial pH is 2, the reaction temperature is 35℃, the amount of sodium persulfate is 4 g/L.

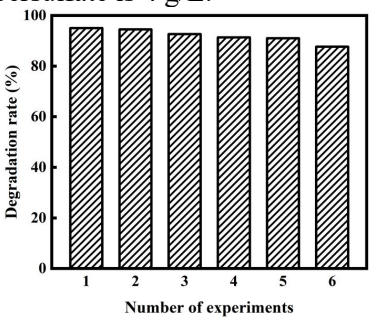
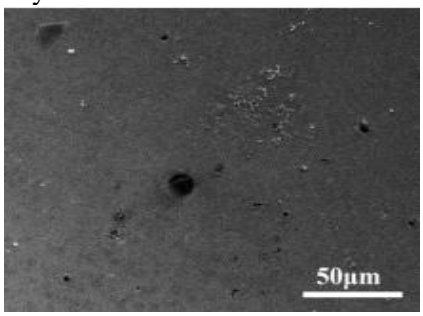


Figure 7. Degradation Effect of Six Cycles Experiment

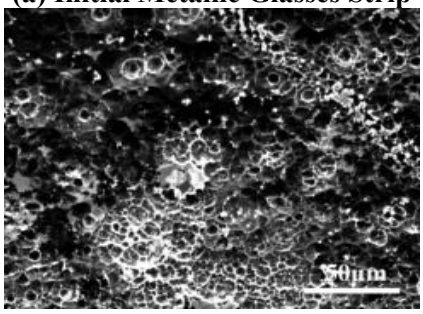
Figure 7 shows that after the amorphous ribbons are recycled for six times, the degradation rate of dyes can always remain above 85%, and there is no significant decline from the overall trend. This result shows that the amorphous ribbons can still maintain a high level of catalytic activity and good structural stability even after repeated cyclic reactions. In addition, its own corrosion resistance and passivation resistance will also significantly inhibit the loss of active sites, thus further ensuring the reuse effect of the catalyst.

In order to explore the stability of $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses in the process of multiple cycle degradation, the surface morphology before and after the reaction is analyzed, and the reuse performance is evaluated by comparing the changes of strip surface morphology before and after the cycle. The surface morphology of the Metallic glasses before and after degradation is shown in Figure 8.

Figure 8 reveals the initial $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses strip with smooth surface, no obvious defects, and only a small amount of pores and impurities. After six cycles, it can be observed that the porous structure is more obvious, the surface layer of the material forms interconnected dense pores, and the range of structural change is still controllable, which proves that $Cu_{46}Zr_{45}Al_7Y_2$ band is involved in the degradation of dyes. Repeated recycling will cause the alloy to form a small area of structural changes on the surface, will not cause large-area damage and performance failure, and the overall structure of the material is stable. After six cycles of degradation, the degradation rate of Acid Orange istewater reached more than 85%, and the catalytic performance did not decline significantly, indicating that the alloy has good reusability.



(a) Initial Metallic Glasses Strip



(b) Amorphous Ribbons Recycled Six Times

Figure 8. Surface Morphology of $Cu_{46}Zr_{45}Al_7Y_2$ Metallic Glasses Strips before and after Degradation

In conclusion, $Cu_{46}Zr_{45}Al_7Y_2$ activated sodium persulfate has a good effect on the degradation of methylene blue. Combined with the specific analysis, the principle analysis of related reactions is summarized. Figure 9 reveals the schematic diagram of the degradation principle of Acid Orange by activated sodium persulfate of Cu-based alloy.

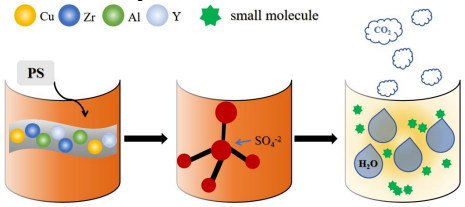


Figure 9. Activation of Sodium Persulfate by Cu based Alloy

The mechanism of Cu-based Metallic glasses activated sodium persulfate degradation of methylene blue mainly involves the process of electron transfer and free radical formation. Specifically, persulfate will rapidly adhere to the surface active sites of $Cu_{46}Zr_{45}Al_7Y_2$ Metallic glasses after being put into the reaction system. Relying on the unique disordered atomic structure and abundant surface active sites of the Metallic glasses, persulfate can effectively activate and generate highly reactive sulfate free radicals (SO_4^-•) which will continue to attack organic dye macromolecules and carry out oxidative cracking reaction on them, and can gradually mineralize complex organic pollutants into non-toxic small molecules [16,17], water (H_2O) and carbon dioxide (CO_2), so as to complete the efficient degradation and removal of organic dye pollutants in water .

4. Conclusion

In order to enhance the ability of Metallic glasses to degrade azo dyes, the catalytic performance of Cu based Metallic glasses in the degradation of acid orange fuel by activated sodium persulfate is studied. The results show that when the starting concentration of acid orange solution is 50 mg/L, the amount of $Cu_{46}Zr_{45}Al_7Y_2$ amorphous strip is 2 g/L, the amount of sodium persulfate is 4 g/L, the initial pH is 2, and the reaction temperature is 35 ℃, under these optimal conditions, the degradation rate of acid orange can reach 95.01% after only 15 mins of degradation reaction, which fully reflected the excellent catalytic performance of Cu-based Metallic glasses and the strong oxidative degradation ability of sulfate radical. Furthermore, the $Cu_{46}Zr_{45}Al_7Y_2$ catalyst demonstrates excellent structural durability after five cycling tests, and its degradation efficiency can still remain above 90% even after these repeated uses.

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