

Contents lists available at ScienceDirect

Journal of Molecular Liquids



Role of maze like structure and Y₂O₃ on Al-based amorphous ribbon surface in MO solution degradation



Qi Chen^a, Zhicheng Yan^a, Lingyu Guo^a, Hao Zhang^a, Lai-Chang Zhang^b, Weimin Wang^{a,*}

^a Key Laboratory for Liquid-Solid Structural Evolution and Processing of Materials (Ministry of Education), School of Materials Science and Engineering, Shandong University, Jinan 250061, China ^b School of Engineering, Edith Cowan University, 270 Joondalup Drive, Joondalup, Perth WA6027, Australia

ARTICLE INFO

Article history: Received 30 June 2020 Received in revised form 9 September 2020 Accepted 12 September 2020 Available online 14 September 2020

Keywords: Al-based alloys Methyl orange dye Maze like structure Y₂O₃

ABSTRACT

The $Al_{85}Co_{10}Y_5$ (Co10) amorphous ribbon exhibits an excellent degradation performance in polluted water treatment for the first time by degrading methyl orange (MO) dye. Compared with the $Al_{85}Fe_{10}Y_5$ (Fe10) amorphous ribbon for degradation MO solution, the Co10 ribbon has a higher degradation efficiency, a higher TOC removal rate, a lower reaction activation energy, a stronger reusability and a higher corrosion resistance. The Co10 ribbon is not only applicable for acidic MO solution, but also has high degradation efficiency in alkaline MO solution. Compared with the spherical particles on the surface of Fe10 ribbon, the maze like structure on the surface of Co10 ribbon can increase the reaction surface area and improve the degradation efficiency of MO solution. And more $\bullet O_2^-$ radicals by supplying photo catalytic Y_2O_3 under visible light irradiation and then enhance the degradation of MO solution, but also provides a new method with high efficiency and low cost for the degradation of MO solution, but also provides a new research direction for the degradation mechanism of Al-based alloys.

© 2020 Elsevier B.V. All rights reserved.

1. Introduction

Azo dyes have been widely used in textile industry. However, they have teratogenic and carcinogenic properties, chemical stability and non-decomposition [1–3]. The treatment of sewage containing this synthetic dye has caused serious environmental problems and has become one of the major environmental problems in the world [4–7]. In the past few decades, people have made great efforts to reduce their harmful effects, including physical adsorption using activated carbon and clay [8,9], biodegradation using microorganisms [10], chemical degradation using advanced oxidation processes [11,12] and degradation by specific alloy [13–17], etc. However, these methods have obvious defects such as low efficiency, high cost and short service life. Thus, we should actively explore advanced materials for better degradation of synthetic dyes in polluted water [18].

Amorphous alloys have attracted more and more attention due to their unique metastable structure, excellent corrosion resistance and oxidation resistance [19,20]. At present, there are many researches on the degradation of azo dyes by amorphous alloy, including Fe-based [21–29], Mg-based [30–33], Co-based [34–36] and Al-based [37–39] ribbons, powders and nanoporous structures, have been proved to have good degradation performance in the wastewater for removing synthetic dyes and organic pollutants. It is generally believed that the thermodynamic instability, high residual stress and a large number of unsaturated sites on the surface of amorphous alloy are the reasons for its excellent degradation performance [40].

At present, the amorphous alloys used to catalyze the degradation of synthetic dyes are mostly ribbon [41–43] and powder shapes [13,44–47]. Among all the amorphous alloys used for dye degradation, Al-based amorphous alloys have attracted much attention due to their low material cost, corrosion resistance, good repeatability and high decomposition efficiency. Wang et al. studied the degradation of Al₉₁. $_xNi_9Y_x$ (x = 0, 3, 6 and 9 at.%) metallic ribbons on azo dyes at different pH values [38]. The results showed that the reactivity of Al-based metallic glass in alkaline and acidic azo dye solutions was about 1.5 and 189 times higher than that in neutral solutions, respectively. The low activation energy and network structure on the surface of Al-based amorphous ribbon make it have higher reactivity in acidic and alkaline solutions, which shows that Al-based amorphous ribbons have good potential in degrading azo dye solutions.

In recent years, some scholars have studied the ability of Y₂O₃-based materials to degrade organic compounds under visible light irradiation [48–52]. Karunakaran et al. conducted photocatalytic degradation of formic, oxalic, acetic and citric acids on the surface of Y₂O₃ and found that the order of photodegradation was as follows: formic acid > oxalic acid > acetic acid > citric acid, indicating that Y₂O₃ has a good degradation performance for organic compounds [53]. Magdalane et al. synthesized CeO₂/Y₂O₃ materials with layered nanostructures by chemical precipitation assisted hydrothermal method, and generated reactive

^{*} Corresponding author. E-mail address: weiminw@sdu.edu.cn (W. Wang).

oxygen species with organic dye degradation performance under visible light. As CeO_2/Y_2O_3 binary metal oxide nanostructure has hierarchical one-dimensional nanostructures that can increase the reaction surface areas, the CeO_2/Y_2O_3 shows higher photocatalytic performance in the degradation of Rhodamine-B dye [54]. The role of transition metals of Al-based glasses in the glass formation and dye degradation is still unclear and valuable to be further studied.

In this paper, we report the degradation of synthetic dyes by using $AI_{85}Co_{10}Y_5$ (Co10) amorphous ribbon for the first time, and the degradation processes using $AI_{85}Fe_{10}Y_5$ (Fe10) amorphous ribbon are investigated for comparison. Methyl orange (MO) is a common synthetic dye used for acid-base titration indicator and dyeing textile, and is used as degradation object in this paper. Through cyclic tests, it is revealed that Co10 ribbon has a longer service life than Fe10 ribbon. The Co10 amorphous ribbons have high degradation pathways under acidic and alkaline conditions have been investigated. Our research not only provides a new research idea for polluted water treatment, but also extends the application field of Al-based amorphous alloy.

2. Experimental

2.1. Materials and reagent

The alloy ingots with nominal compositions of Al₈₅Fe₁₀Y₅ (Fe10, at. %) and $Al_{85}Co_{10}Y_5$ (Co10, at.%) were prepared by arc melting of highpurity (99.5 wt%-99.9 wt%) Al, Fe, Co and Y metals in an arc melting system, which was vacuumed to 5×10^{-3} Pa first and then filled with purified argon (99.999%). Ribbons with a thickness of ~20 µm and a width of ~2 mm were prepared in a single roller melt-spinning system in the air, with a speed of 44 m s^{-1} . The ribbons were cut into 5 cm long strips for degradation tests. The ribbons were then annealed at 579 K (Fe10) and 550 K (Co10) in Ar atmosphere. Commercially available methyl orange (MO, C14H14N3NaO3S, AR grade) was purchased from Tianjin Tianxin Fine Chemical Development Center. Hydrochloric acid (HCl, AR grade) was purchased from Sinopharm Chemical Reagent Co., Ltd. Sodium hydroxide (NaOH, AR grade) was purchased from Tianjin Hengxing Chemical Reagent Manufacturing Co., Ltd. 1,4-benzoquinone $(BO, C_6H_4O_2, CP grade)$ was purchased from Shanghai dingfen Chemical Technology Co., Ltd. Tertiary butanol (TBA, C₄H₁₀O, CP grade) was purchased from Sinopharm Chemical Reagent Co., Ltd.

2.2. Characterization

The amorphous structure of the as-spun ribbons was characterized by X-ray diffraction (XRD, Bruker D8 Discover) with Cu-K α radiation and transmission electron microscopy (TEM, JEM-2100). The amorphous structure of the ribbons was also certified by differential scanning calorimetry (DSC, NETZSCH-404) at a heating rate of 20 K/min. The surface morphology of the ribbons was observed using a scanning electron microscope (SEM, JSM-7800F) equipped with an energy dispersive Xray spectrometer (EDS). The binding states of elements on the surfaces of the as-spun and reacted Fe10 and Co10 ribbons were investigated by X-ray photoelectron spectroscopy (XPS, AXIS Supra) with a monochromatic Al K α X-ray source (hv = 1486.6 eV). The total organic carbon (TOC) of the MO solution was determined with a TOC-L analyzer. The specific surface area (SSA) of the as-spun and reacted Fe10 and Co10 ribbons were measured by surface area and porosimetry analyzer (V-Sorb 2800P).

2.3. Degradation tests

First, 500 mL MO solution (10 mg L^{-1} MO if not noted) has been prepared in a volumetric flask using deionized water (DW) and MO powders. Then, 50 mL MO solution was poured in a 100 mL beaker in preparation of the degradation test. A specific amount of ribbons

 $(1.0 \text{ g L}^{-1} \text{ if not noted})$ was added to the solution, upon visible light irradiation, the solution was stirred at a fixed speed (200 r min⁻¹) during the degradation process, and the temperature (298 K if not noted) of the solution was maintained using a water bath. The initial pH (pH = 1 if not noted) of the solution was adjusted using 12 mol L⁻¹ HCl, as well as 1 M NaOH. At selected time intervals, 3 mL of the solution was extracted using a syringe and filtered with a 0.45 µm membrane, and the concentration of real-time MO solution was monitored using a UV-Vis spectrophotometer (UV-4802) to obtain the absorbance spectrum of the solution. For cyclic tests, the ribbons were extracted from the solution after each degradation test and stir washed with deionized water for 60 s before putting them into the next reaction batch.

2.4. Electrochemical tests

The electrochemical properties including polarization curves and impedance spectra (EIS) were measured using an electrochemical measuring instrument (CHI 660E) in the 25 mL DW or MO solutions (10 mg L⁻¹ MO) at 298 K. The three-electrode cell was used for measurement, with saturated calomel electrode (SCE) as reference electrode, platinum as counter electrode and as-spun ribbon as working electrode. The polarization curves were recorded at a potential sweep speed of 1 mV s⁻¹ after the open circuit potentials were stabilized. EIS was conducted under static states with scanning frequencies from 100 kHz to 0.01 Hz and the amplitude was \pm 10 mV.

3. Results

3.1. Microstructure of $Al_{85}(Fe/Co)_{10}Y_5$ amorphous ribbons

A. XRD and DSC analysis

Fig. 1 shows the XRD patterns and DSC curves of as-spun Al₈₅Fe₁₀Y₅ (Fe10) and Al₈₅Co₁₀Y₅ (Co10) ribbons. For comparison, the XRD patterns of the annealed Fe10 and Co10 ribbons at 579 K and 550 K for 5 min are added. Here the annealed temperature is 30 K below the crystallization peak T_{P1} in DSC curves. As shown in Fig. 1(a) the XRD pattern of as-spun Fe10 ribbon has only a typical diffuse scattering peak at about $2\theta = 40$ degree, indicating that the as-spun Fe10 ribbon owns a fully amorphous structure. This diffuse scattering peak can be decomposed into P1 and P2 peaks and the area fraction $A_{P2}/(A_{P1} + A_{P2})$ of P2 is 40%. The crystalline peaks in the XRD pattern of annealed Fe10 ribbon can be identified as α -Al, Al₂Fe and Fe₂Y crystalline phases, indicating the ribbon experiences the crystallization transformation in annealing. Apparently, the P1 is locating at the position of Al (111) peak and P2 at the position of Fe₂Y (222) peak. Based on the heredity between the amorphous state and the crystalline state [55], the decomposed P1 in Fig. 1(a) is associated with Al—Al clusters, and P2 with Fe—Y clusters. The as-spun Co10 ribbon also has a typical diffuse scattering peak, which can also be decomposed into P1 and P2 peaks (Fig. 1(b)). In addition, the area fraction $A_{P2}/(A_{P1} + A_{P2})$ of P2 is 27.6%, which is lower than 40% for as-spun Fe10 ribbon. Moreover, P1 stands at the left side of Al (111), while P2 locates at the same position of P2 in as-spun Fe10 ribbon XRD pattern, i.e., the position of Co₂Y (222) peak. Hence, the P1 represents Al—Al and Al—Y clusters, and P2 represents Co—Y clusters. The XRD pattern of annealed Co10 ribbon still has diffuse scattering peak at 38 degree, indicating that a fully amorphous structure after annealing. Moreover, the position of main peak in annealed Co10 ribbon is corresponding to Al (111) peak and the corresponding atomic distance is shorter than is as-spun state, which may be ascribed to the oxidation of the abundant Y atoms in Al-Y clusters in annealing. Nevertheless, at the temperature below crystallization, the as-spun Co10 ribbon is more stable than the as-spun Fe10 ribbon.

Fig. 1(c) shows the DSC curves of as-spun Fe10 and Co10 ribbons. In the DSC curve of Fe10 ribbon, there are three crystallization peaks T_{P1} ,



Fig. 1. XRD patterns of the as-spun and annealed (a) Al₈₅Fe₁₀Y₅ (Fe10) and (b) Al₈₅Co₁₀Y₅ (Co10) ribbons, (c) the DSC curves of the as-spun Fe10 and Co10 ribbons.

 T_{P2} and T_{P3} in the heating scan, the T_{P3} - T_{P1} is 217 K, indicates a wide crystallization temperature range and a heterogeneous structure in the as-spun Fe10 ribbon, this is consistent with the high P2 area fraction decomposed from the main peak in its XRD pattern. The crystallization peaks in the XRD pattern of Fe10 ribbon annealed at 30 K below crystallization peak temperature also confirm the heterogeneous structure of Fe10 ribbon. In the DSC curve of Co10 ribbon, there are two crystallization peaks T_{P1} and T_{P2} , the crystallization range is much shorter than Fe10 ribbon. The exothermic heat ΔH_C of the crystallization of Co10 ribbon is higher than the ΔH_C of Fe10 ribbon, indicating that the as-spun Co10 ribbon has the glass formability, stability and homogeneity, which are higher than Fe10 ribbon and these characteristics are consistent with the XRD patterns (Fig. 1(a) and (b)).

B. TEM investigation

In order to further characterize the microstructure and morphology of the as-spun Fe10 and Co10 ribbons, we carried out TEM investigations, which are shown in Fig. 2. There is mainly maze shape pattern without crystallites in the high resolution bright field images of the as-spun Fe10 and Co10 ribbons (Fig. 2(a) and (b)). Here, the amount of the ordered zone in diameter of 20 Å in Fe10 ribbon inside the circle is higher than in Co10 ribbon. The corresponding SAED patterns have two typical diffraction halos (Fig. 2(c) and (d)), confirming that the as-spun Fe10 and Co10 ribbons own a fully amorphous structure. And the light intensity of the inner diffraction halo in the as-spun Fe10 and Co10 ribbons can be decomposed into P1 and P2 peaks (inset in Fig. 2 (c) and (d)). Here, the P2 area fraction of the as-spun Fe10 ribbon is higher than that of Co ribbon. Thus, the results of TEM are agreeing with the XRD patterns (Fig. 1(a) and (b)). Hence, the P1 and P2 peaks in SAED of Fe10 ribbon represent Al-Al and Fe-Y clusters, respectively. The P1 of Co10 ribbon represents Al—Al and Al—Y clusters and P2 stands for Co-Y clusters. The TEM bright field images and light intensities of SAED patterns also indicate that the homogenous degree of the as-spun Co10 ribbon is higher than Fe10 ribbon.

3.2. Degradation performance of $Al_{85}(Fe/Co)_{10}Y_5$ amorphous ribbons

A. UV-Vis absorbance analysis and degradation performance

The UV–Vis absorbance spectra of the filtered methyl orange (MO) solution with pH = 1 after processing with as-spun Fe10 and Co10 ribbons for a series of time intervals ($t_r = 0-45$ min) are presented in Fig. 3 (a) and (b). The spectra of MO solution have a major absorption peak at about 508 nm, which represents MO chromophore group, i.e., nitrogen-nitrogen double bond (-N=N-) [56,57]. The normalized concentration of the MO solution is obtained with the peak values at

508 nm, as shown in Fig. 3(c). In reacting with both ribbons, the solution concentration stays almost unchanged during the first 5 min, and then declines rapidly. With increasing t_r , the absorption peak at 508 nm decays gradually, suggesting that the chromophore groups of MO disappear gradually. The degradation kinetics after the first 5 min are usually described by the pseudo-first-order equation as follows [58]:

$$C_{\rm t} = C_0 \, \exp(-kt_{\rm r}) \tag{1}$$

where k is the reaction rate constant (min⁻¹), C_0 is the initial concentration of MO solution (mg L⁻¹), and C_t is the instant concentration of MO solution (mg L⁻¹) at t_r . Then the degradation reaction rate constant can be derived as follows:

$$k = \ln\left(\frac{C_0}{C_t}\right)/t_r \tag{2}$$

According to the ln (C_0/C_t) – t_r curves shown in Fig. 3(d), the reaction rate constant of Co10 ribbon in this degradation reaction is 0.069 min⁻¹, which is larger than 0.049 min⁻¹ for Fe10 ribbon, with the goodness of fit R^2 being 0.96 and 0.97, respectively. Thus, the asspun Co10 ribbon exhibit a higher degradation ability for MO solution with pH = 1 than the as-spun Fe10 ribbon.

In order to further analyze the mineralization of MO solution during degradation with as-spun Fe10 and Co10 ribbons, their TOC removal rates are shown in Fig. 3(e). It is observed that the mineralization rate of as-spun Co10 (82.4%) ribbon for MO solution is higher than that of as-spun Fe10 (75.1%) ribbon at 45 min, which is corresponding to the degradation efficiency of as-spun Co10 (94.4%) and Fe10 (86.7%) ribbons for MO solution in Fig. 3(c). However, the mineralization rate of as-spun Fe10 and Co10 ribbons for MO solution is lower than the corresponding degradation efficiency, which is due to the fact that in the process of degradation and mineralization, the degrading radicals first attack the chromophoric groups and make the MO solution decolorize quickly. Afterward, the initial degradation products are generated, which are further mineralized into the final products as H₂O, CO₂, NO₃⁻ and SO₄²⁻. Thus, completing mineralization process takes longer time than degradation [24].

Fig. 3(f) shows the MO dye equilibrium dark adsorption on as-spun Fe10 and Co10 ribbons at pH = 7 and MO solution concentration of 10 mg L⁻¹. The quantity of adsorption q (mg/g) of the MO dye is shown as the following equation [26]:

$$q = (C_0 - C_t)V/M \tag{3}$$

where C_0 is the initial concentration of MO solution $(mg L^{-1})$, C_t is the instant concentration of MO solution $(mg L^{-1})$ at t_r , V is the volume of the MO solution (L) and M is the mass of as-spun Fe10 and Co10 ribbons (g). Fast MO dye adsorption is observed on as-spun Fe10 and Co10



Fig. 2. The TEM images of the as-spun (a) Al₈₅Fe₁₀Y₅ (Fe10) and (b) Al₈₅Co₁₀Y₅ (Co10) ribbons, the SAED patterns of the as-spun (c) Fe10 and (d) Co10 ribbons. The insets in (c) and (d): light intensity of SAED patterns along AB line.

ribbons from initial q = 0 to q = 0.31 and 0.40 mg/g at $t_r = 15$ min followed by slower equilibrium adsorption with q = 0.47 and 0.56 mg/g at $t_r = 45$ min, respectively. In the degrading process, the MO dye molecules are initially adsorbed on the as-spun Fe10 and Co10 ribbon surfaces, and then are further degraded and mineralized. The q of the as-spun Co10 ribbon for MO dye is higher than that of asspun Fe10 ribbon. Thus, from the q, we think that as-spun Co10 ribbon is more conducive to degradate and mineralize the MO solution than the as-spun Fe10 ribbon.

B. SEM images of surface morphology

To understand the MO degradation process with Al-based amorphous ribbons, it is very important to study the structure evolution of the ribbon surfaces after the reaction. The SEM images on the surface of the as-spun and reacted Fe10 and Co10 ribbons are displayed in Fig. 4, the EDS and specific surface area (SSA) results are listed in Table 1. A typical smooth surface of amorphous ribbons is observed on the as-spun Fe10 and Co10 ribbons (Fig. 4(a) and (b)). After reacting with MO solution the Fe10 ribbon surface has many round corroding spots, which are related to the Al—Al cluster (Fig. 4(c)). After the reaction, the Co10 ribbon surface has a maze like network structure, shows a spinodal decomposed characteristic (Fig. 4(d)). After reaction for 45 min, the Fe10 and Co10 ribbons have decreased c_{Al} and c_{Y} , and an increased c_{Fe} and c_{Co} (Table 1). After degradation, the c_{O} increment in Fe10 ribbon surface is apparently larger than that in Co10 ribbon surface. Moreover, the c_0 of Fe10 ribbon before and after degradation is higher than that of Co10 ribbon, indicating a lower anti-oxidation ability than

Co10 ribbon. The specific surface areas (SSA) of the as-spun Fe10 and Co10 ribbons are basically the same (Table 1), which corresponds to the typical smooth surface in SEM images (Fig. 4(a) and (b)). After the reaction, the specific surface areas of Fe10 and Co10 ribbons increased to 0.131 and 0.172 m^2/g , which was related to the round corroding spots formed on the surface of Fe10 ribbon and the maze like network structure formed on the surface of Co10 ribbon (Fig. 4(c) and (d)), respectively. In other words, the network structure on the surface of Co10 ribbon greatly increased its specific surface area and provided more reactive sites for the degradation of MO solution, and thus improving the degradation efficiency.

C. XPS analysis

It is known that the surface elements especially their electronic structure of an amorphous ribbon have a great influence on the degradation efficiency. Figs. 5 and 6 show the XPS analysis on the surface of the as-spun and reacted Fe10 and Co10 ribbons to measure Al 2p, Fe 2p_{3/2}, Co 2p_{3/2}, Y 3d and O 1s, and the XPS parameters are listed in Table 2.

In Fig. 5(a), the Al 2p spectra of as-spun Fe10 ribbon can be deconvoluted into 3 peaks P1, P2 and P3 at 72.8, 74.9 and 76.0 eV, which are assigned to the metallic Al⁰, Al₂O₃ and Al(OH)₃, respectively [59]. Similar to the as-spun Fe10 ribbon, the Al⁰, Al₂O₃ and Al(OH)₃ peaks of the reacted Fe10 ribbon locate at 72.6, 75.1 and 76.1 eV, respectively. Generally, the area of each peak is related to the relative content of element in the sample. The fraction of Al⁰ peak on the surface of the as-spun Fe10 ribbon is about 45.8% of the total Al 2p spectrum, and



Fig. 3. UV–Vis absorbance spectra of the as-spun (a) $Al_{85}Fe_{10}Y_5$ (Fe10) and (b) $Al_{85}Co_{10}Y_5$ (Co10) ribbons after reacted with MO solution with time $t_r = 0-45$ min, (c) normalized concentration change of MO solutions during the degradation process, (d) the ln (C_0/C_t)- t_r curves for the as-spun Fe10 and Co10 ribbons, (e) TOC removal rate of MO solution during the degradation process, (f) quantity of MO dye adsorption on as-spun Fe10 and Co10 ribbons.

decreases to 29.7% after degradation, indicating that α -Al phase precipitates from the amorphous matrix and dissolves in the degradation of MO solution. In Fig. 5(b) both of the Fe 2p_{3/2} spectra from the as-spun Fe10 ribbons consist of 3 peaks P1, P2 and P3 at 707.3, 709.5 and 712.0 eV, with P1, P2 and P3 from the reacted Fe10 ribbon at 707.5, 709.3 and 711.3 eV, respectively. According to Ref [60], P1, P2 and P3 are assigned to the metallic state Fe⁰, oxidized Fe²⁺ and Fe³⁺ ions, respectively. The Fe⁰/Fe_{total} area ratio of the as-spun ribbon decreases from 67.2% to 58.4% after degradation. Meanwhile, the absolute area of Fe⁰ in reacted Fe10 ribbon is much higher than as-spun Fe10 ribbon.

The Y 3d spectrum exhibits a doublet due to the spin-orbit splitting into Y $3d_{5/2}$ and Y $3d_{3/2}$, and both Y⁰ and Y₂O₃ have $3d_{5/2}$ and $3d_{3/2}$ [61,62]. In Fig. 5(c) both of the Y 3d spectra from the as-spun Fe10 ribbons consist of 4 peaks P1, P2, P3 and P4 at 154.7, 156.9, 157.9 and 159.7 eV, with P1, P2, P3 and P4 from reacted Fe10 ribbon at 154.6, 156.8, 157.7 and 159.7 eV, respectively. And the P1, P2, P3 and P4 are assigned to Y⁰ $3d_{5/2}$, Y⁰ $3d_{3/2}$, Y₂O₃ $3d_{5/2}$ and Y₂O₃ $3d_{3/2}$, respectively. The area fraction of Y⁰ peaks for as-spun ribbon is 3.7% and increases to 44.8% after reaction. This is because the Y₂O₃ is consumed by H⁺ ions in the MO solution, which exposes the Y⁰ on the ribbon surface. In Fig. 5(d), the O 1s spectrum of the as-spun Fe10 ribbon surface consists of 2 peaks P1 and P2 at 529.8 and 531.1 eV, respectively. After reaction, P1 and P2 locate at 529.8 and 531.1 eV. Here, P1 and P2 are assigned to the Y_2O_3 and Al_2O_3 , respectively [63]. The fraction of P1 area of the as-spun Fe10 ribbon is about 35.6%, and decreases to 27.5% after degradation. This result indicates that Y_2O_3 is continuously consumed during degradation, which is consistent with the result in Fig. 5 (c).

The electronic structure of the as-spun and reacted Co10 ribbons is also obtained using XPS for comparison as shown in Fig. 6. Contrast to the as-spun and reacted Fe10 ribbons, in Fig. 6(a) the Al^0/Al_{total} area ratio of the as-spun Co10 ribbon is 37.5%, and decreases to 37.0% after reaction. It indicates that the oxide film on the surface of the Co10 ribbon has been thinned in the reaction of degrading MO solution. Similar to the Fe⁰ peak in the as-spun and reacted Fe10 ribbon, the absolute area of Co⁰ of Co10 ribbon increases apparently after reaction (Fig. 6(b)), confirming the thinning of oxide film in the reaction.

In Fig. 6(c), similar to the Fe10 ribbon, the content of Y_2O_3 on Co10 ribbon surface decreased from 95.7% to 47.3% in the degradation of MO solution, while the content of Y^0 increased from 4.3% to 52.7%. Compared with the Fe10 ribbon, Co10 ribbon has a higher consumption of Y_2O_3 exposed during the reaction with MO solution. In Fig. 6(d), unlike



Fig. 4. SEM micrographs of (a) the as-spun Al₈₅Fe₁₀Y₅ (Fe10) ribbon, (b) the as-spun Al₈₅Co₁₀Y₅ (Co10) ribbon, (c) reacted Fe10 ribbon and (d) reacted Co10 ribbon. The insets in (c) and (d): the high-magnification images.

Fe10 ribbon, the area fraction of Y_2O_3 peak of the as-spun Co10 ribbon is 29.8%, and increases to 38.1% after degradation. Meanwhile, the fraction of Al_2O_3 peak decreases from 70.2% to 61.9%. However, according to the peak of Al 2p spectra of Co10 ribbon decomposition before and after the reaction, the content of Al_2O_3 did not change much. The increase of Y_2O_3 content was probably due to the increase of surface area supplied by the network structure of Co10 ribbon surface.

D. Electrochemical analysis.

The polarization curves and electrochemical impedance spectra (EIS) of the as-spun Fe10 and Co10 ribbons in DW and MO solutions with pH = 1 were shown in Fig. 7. In DW, the corrosion potential (E_{corr}) of the as-spun Co10 ribbon is -0.87 V (Fig. 7(a)), which is higher than Fe10 ribbon (-0.97 V). Besides, the corrosion current densities $(i_{\rm corr})$ of Co10 ribbon is 1.76×10^{-6} A cm⁻², being lower than Fe10 ribbon $(2.64 \times 10^{-6} \text{ A cm}^{-2})$. In MO solution, the E_{corr} value of the as-spun Co10 ribbon is -0.54 V (Fig. 7(b)), being higher than Fe10 ribbon (-0.70 V); in addition, the i_{corr} of Co10 ribbon is $1.55 \times 10^{-5} \text{ A cm}^{-2}$, which is lower than Fe10 ribbon $(1.81 \times 10^{-5} \text{ A cm}^{-2})$. In addition, as E = 0 V vs SCE, the pitting curve of Fe10 ribbon in DW can be divided into two stages (denoted E_P^1 and E_P^2), while that of Co10 ribbon has one stage started at E_P^1 . Similar phenomenon occurs in MO solution. These polarization curves indicate that the as-spun Co10 ribbon has a higher corrosion resistance than as-spun Fe10 ribbon in DW and MO solution, and confirm that the as-spun Co10 ribbon has a more stable and homogenous structure than Fe10 ribbon.

Table 1

EDS and specific surface area (SSA) analysis of the $Al_{85}Fe_{10}Y_5$ (Fe10) and $Al_{85}Co_{10}Y_5$ (Co10) ribbons before and after degradation.

A	lloy	Before degradation						After degradation					
		C _{Al}	C _{Fe}	C _{Co}	CY	C _O	SSA (m ² /g)	C _{Al}	C _{Fe}	C _{Co}	CY	c _o	SSA (m²/g)
Fe	e10 o10	77.7 78.3	14.9 -	- 14.3	5.4 5.5	2.0 1.9	0.111 0.117	76.6 78.1	15.5 -	- 14.6	5.0 5.1	2.9 2.2	0.131 0.172

In both DW and MO solutions (Fig. 7(c) and (d)), the Nyquist semicircle diameter of the as-spun Co10 ribbon is larger than that of Fe10 ribbon. The equivalent circuit consisting of R(C(R(Q(R(CR))))) is used to fit the EIS data in DW and MO solution. In the equivalent circuit, the constant phase element (CPE) Q is defined as [64]:

$$\mathbf{Q} = (j\mathbf{w})^{-n} / \mathbf{Y}_0 \tag{4}$$

The fitting results like resistances of solution (R_s), charge transfer (R_t), film (R_f), reaction (R_a) and total circuit (R_{total}), capacities of electric double layer (C_{dl}) and reaction (C_a), and CPE parameters (Y_f and N_f) are summarized in Table 3. Both ribbons in MO solution have lower R_s and R_t , and higher C_{dl} , Y_f , R_f , C_a and R_{total} than in DW, which may be due to a certain amount of methyl orange molecules and H⁺ in MO solution. Moreover, the R_{total} of Co10 ribbon is higher than that of Fe10 ribbon in both DW and MO solution. Thus, the EIS results are also in good agreement with the results of polarization curves (Fig. 7(a) and (b)). In MO solution, the Co10 ribbon surface area is larger than Fe10 ribbon surface.

3.3. Effect of temperature, pH on ribbon's degradation and reusability

A. Effect of temperature

In order to further compare the MO solution degradation efficiency of the as-spun Fe10 and Co10 ribbons, both ribbons reacted with MO solution at different temperatures (T_r), as shown in Fig. 8. And we deduced k and thermal activation energy E_r of MO solution degradation. The reaction conditions are set as follows: initial pH = 1, ribbon dosage 0.5 g L⁻¹, and $C_{MO} = 10$ mg L⁻¹. The normalized MO solution concentration C_t/C_0 shows that temperature has a positive effect on the degradation process, as the degradation process takes less time with a increasing T_r (Fig. 8(a) and (b)). Meanwhile, the k of the Fe10 and Co10 ribbons increases with T_r (Fig. 8(c)). Since k (T_r) is dependent on T_{r} , the thermal activation energy E_r of the MO solution degradation



Fig. 5. XPS spectra of (a) Al 2p, (b) Fe 2p_{3/2}, (c) Y 3d and (d) O 1s in binding energy regions for the Al₈₅Fe₁₀Y₅ (Fe10) ribbons before and after degradation.

reaction with Fe10 and Co10 ribbons can be deduced through the Arrhenius-type equation [22]:

$$\ln k(T_r) = -\frac{E_r}{RT_r} + \ln A \tag{5}$$

where *R* is the gas constant and *A* is a constant. According to $\ln k(T_r)$ vs $- 1/RT_r$ curves (inset in Fig. 8(c)), the E_r values of Fe10 and Co10 ribbons are 23.8 and 21.3 KJ mol⁻¹, respectively, being opposite to the varying tendency of their *k* at 298 K (Fig. 8(d)) and indicating that Co10 ribbon has a better performance in the MO degradation reaction.

B. Effect of pH

Fig. 9 shows the pH effect on the degradation ability of the as-spun Fe10 and Co10 ribbons against MO solution, with other reaction conditions unchanged. When pH increases from 1 to 5, the degraded MO concentration C_t/C_0 by the as-spun Fe10 ribbon drastically decreases. However, as pH rises from 5 to 9, the C_t/C_0 of MO solution is basically unchanged, i.e., it is basically not degraded. As pH rise from 9 to 13, the C_t / C_0 of MO solution drastically increases, indicating that the degradation effect of Fe10 ribbon is enhanced again (Fig. 9(a)). Meanwhile, similar to the as-spun Fe10 ribbon, the as-spun Co10 ribbon has a similar MO solution degrading behavior as pH increases from 1 to 13 (Fig. 9(b)). The degradation efficiency ($\eta = (1 - C_t/C_0 \times 100\%, t_r = 45 \text{ min})$ of the as-spun Fe10 and Co10 ribbons with $T_r = 298$ K at different pH values are shown in Fig. 9(c) and (d). As pH = 1, the η of the as-spun Fe10 (86.7%) and Co10 (94.4%) ribbons is maximum. As pH rises from 5 to 9, the η of the as-spun Fe10 and Co10 ribbons is basically zero, respectively. As pH rises from 9 to 13, the η of the as-spun Fe10 and Co10 ribbons is drastically increases. As pH = 13, the η of the as-spun Fe10 (84.3%) and Co10 (91.5%) ribbons is still lower than the η of their counterparts at pH =1 (Fig. 9(c) and (d)). The deduced η values of both ribbons at various pH also suggest that the degradation efficiency of Co10 ribbon is higher than Fe10 ribbon.

C. Reusability and stability of Fe10 and Co10 in degradation MO solution

The reusability and stability of amorphous ribbon are important indexes to evaluate the potential of polluted water remediation. We tested the degradation efficiency η of the Fe10 and Co10 ribbons in MO solution with pH = 1 by reusability test as shown in Fig. 10. The η of the Fe10 ribbon in MO solution, increases gradually from the 1st to 3rd cycle, and decreases gradually from the 4th to 7th cycle (Fig. 10 (a) and (b)). The η variation of Co10 ribbon from the 1st to 3rd cycle is similar to that of Fe10 ribbon, but the η in the 4th cycle decreased to the lowest value, and then increased gradually from the 5th to 7th cycle, showing a better reusability than Fe10 ribbon (Fig. 10(c) and (d)). It can be noted that the η of the Fe10 and Co10 ribbons in the 2nd and 3rd degradation cycle is higher than in the 1st cycle (Fig. 10 (b) and (d)). And normalized concentration C_t/C_0 of the solution reacting with both kinds of ribbons stays almost unchanged during the first 5 min (Fig. 10(a) and (c)). This may be because it takes certain time to consume the thin oxide layer on the ribbon during the 1st test, while the 2nd and 3rd runs can react with the truly fresh surfaces of the Fe10 and Co10 ribbons.

4. Discussion

Under different pH conditions, the degradation efficiency and degradation mechanism of the as-spun $Al_{85}Fe_{10}Y_5$ (Fe10) and $Al_{85}Co_{10}Y_5$



Fig. 6. XPS spectra of (a) Al 2p, (b) Co 2p_{3/2}, (c) Y 3d and (d) O 1s in binding energy regions for the Al₈₅Co₁₀Y₅ (Co10) ribbons before and after degradation.

(Co10) ribbons in MO solution are different. As pH \leq 3, hydrogen ions are dominant in MO solution. The H⁺ ions can react with Al or other metals on the ribbon surface to produce the reduced initial hydrogen [H] radicals, which have certain reducibility and can degrade MO solution [65–69]. As pH \geq 11, the dominant OH⁻ ions can react with Al and other metals on the surface of the ribbon to produce hydroxyl •OH radicals, which have strong oxidibility and can degrade MO solution [69,70]. However, in the condition where 5 \leq pH \leq 9, MO solution is more stable and does not undergo any degradation, which may be due to the fact that the concentration of H⁺ or OH⁻ in MO solution is too

low to effectively react and produce [H] or •OH radicals. When pH = 1, there exists a transient platform before degradation of MO solution; while in the condition of pH = 13, there doesn't exist any transient platform (Fig. 9(a) and (b)). It is due to the fact that oxides on the surface of the as-spun Fe10 and Co10 ribbons are easier to react with OH⁻, which can accelerate the degradation rate. However, when the reaction had lasted for 30 min, the degradation rate of pH = 13 was gradually decreased and became lower than that of pH = 1 (Fig. 9(a) and (b)), which should be due to the decrease of OH⁻ in the solution and the decrease of the rate of generating •OH radical.

Table 2

KPS analysis of the Al 2p, Fe 2p _{3/2} , Co 2p	$_{1/2}$, Y 3d and O 1s of the Al ₈₅ Fe ₁₀ Y ₅ (F	e10) and Al ₈₅ Co ₁₀ Y ₅ (Co10) ribbons	s before degradation (BD) and after	degradation (AD).
---	---	--	-------------------------------------	-------------------

Alloy		Spectrum	P1		P2		Р3		P4	
			A	%	A	%	A	%	A	%
Fe10	BD	Al 2p	13,522	45.8	11,638	39.4	4378	14.8	_	-
		Fe 2p _{3/2}	5577	67.2	1803	21.7	921	11.1	-	-
		Y 3d	2209	1.9	2023	1.8	61,910	54.6	47,210	41.7
		O 1s	110,723	35.6	200,070	64.4	-	-	-	-
	AD	Al 2p	9198	29.7	15,857	51.2	5912	19.1	-	-
		Fe 2p _{3/2}	14,339	58.4	6607	26.9	3619	14.7	-	-
		Y 3d	2757	22.8	2654	22.0	4377	36.2	2294	19.0
		O 1s	80,048	27.5	210,635	72.5	-	-	-	-
Co10	BD	Al 2p	10,983	37.5	12,329	42.1	5997	20.4	-	-
		$Co 2p_{3/2}$	5520	73.1	1438	19.0	596	7.9	-	-
		Y 3d	2800	2.4	2223	1.9	63,133	54.3	48,035	41.4
		O 1s	90,251	29.8	212,441	70.2	-	-	-	-
	AD	Al 2p	14,921	37.0	18,253	45.2	7190	17.8	-	-
		Co 2p _{3/2}	32,800	70.6	8847	19.1	4783	10.3	-	-
		Y 3d	4149	27.7	3744	25.0	4305	28.8	2759	18.5
		O 1s	120,586	38.1	195,558	61.9	-	-	-	-



Fig. 7. Polarization curves of the as-spun Al₈₅Fe₁₀Y₅ (Fe10) and Al₈₅Co₁₀Y₅ (co10) ribbons in (a) DW and (b) MO solutions and their Nyquist curves in (c) DW and (d) MO solutions. The upper insets in (c) and (d): the general fitted circuit. Symbols show the experimental data while solid lines are fitting results.

Through XPS results of the as-spun and reacted surface elements of Fe10 and Co10 ribbons, we found that consumption of Al⁰ and Fe⁰ on the surface of Fe10 ribbon during reaction was higher than the loss of Al⁰ and Co⁰ on the surface of Co10 ribbon. Compared with Fe₂O₃ and Co₂O₃, the Y₂O₃ occupy a higher fraction on the Fe10 and Co10 ribbons surface in as-spun and reacted states (Figs. 5 and 6). Upon visible light irradiation, the electron from the valence band of Y₂O₃ can be excited to conduction band react with O₂ to generate \cdot O₂⁻ and then accelerate the degradation of MO solution [49,54]. Through XPS analysis, we know that the relative content of Y₂O₃ on the surface of Co10 ribbon is higher than that of Fe10 ribbon, leading to the fast degradation MO solution, which explains the better degradation performance of the Co10 amorphous ribbon.

In order to explore the degradation mechanism of MO solution under different pH conditions and clarify the contribution of reactive oxygen species during the degradation process, we investigated the degradation efficiency η change of MO solution after adding quenching agents of 1,4-benzoquinone (BQ 0.1 mol L⁻¹) and tertiary butanol (TBA 0.1 mol L⁻¹), which are normally used for quenching the produced •O₂⁻ and •OH radicals, respectively [71,72]. As shown in Fig. 11, the η of MB solution decreased to different degrees after adding BQ and TBA, indicating the produced $\bullet O_2^-$ and $\bullet OH$ radicals play an important role in the degradation of the MO dye molecules. When pH = 1, the addition of BQ resulted the η of Co10 ribbon on MO solution decreased to 75.5%, being 18.9% lower than the original result, and confirming that the added BQ can quench the produced $\bullet O_2^-$ (Fig. 11(a)). However, the present η is still high (75.5%), owing to the [H] generated in the reaction, which plays a dominant role in the degradation of MO solution under acid conditions [38,69]. When pH = 13, the addition of BQ and TBA reduced the η of Co10 ribbon to 70.1% and 41.6%, respectively, indicating that the addition of BQ and TBA could respectively quench the $\bullet O_2^-$ and $\bullet OH$ generated in the reaction, and the $\bullet OH$ was the dominant species for degradation of alkaline MO solution (Fig. 11(b)). Moreover, the simultaneous addition of BQ and TBA significantly reduced the MO solution degradation efficiency from 91.5 to 26.4%.

The degradation efficiency η of MO solution is increased from the 1st to 3rd cycle. This is due to the fact that various metal oxide films are removed in the 1st cycle, so that the degradation of MO solution can begin directly in the 2nd and 3rd cycles, which can be confirmed by the non-transient platform of the 2nd and 3rd cycles in Fig. 10(a) and (c). The η

Table 3

Parameters from EIS measurements: R_s , solution resistance; C_{dl} , resistance of electric double layer; R_t , resistance of transfer charge; Q_f and R_b resistance of passivation film; C_a and R_a , resistance of electrochemical reaction; R_{totab} total resistance.

Solution	Alloy	$R_{\rm s} \left(\Omega \cdot {\rm cm}^2 \right)$	$C_{\rm dl} (10^{-7} \Omega^{-1} \cdot {\rm cm}^{-2})$	$R_{\rm t} \left(\Omega \cdot {\rm cm}^2 \right)$	Q _f		$R_{\rm f}$ (10 ⁴ $\Omega \cdot { m cm}^2$)	$C_{\rm a} (10^{-6} \Omega^{-1} \cdot {\rm cm}^{-2})$	$R_{\rm a} \left(\Omega \cdot {\rm cm}^2 \right)$	$R_{\rm total} \left(\Omega \cdot { m cm}^2 \right)$
					$Y_{\rm f} (10^{-6} \ \Omega^{-1} {\rm s}^{-n} \cdot {\rm cm}^{-2})$	N_{f}				
DW	Fe10	1.63	7.0×10^{-3}	566	4.6×10^{-3}	0.97	0.69	$9.4 imes 10^{-3}$	0.05	7467
	Co10	1.29	6.2×10^{-3}	521	8.1×10^{-3}	0.93	0.85	$2.4 imes 10^{-3}$	1.3×10^3	10,322
MO	Fe10	0.07	1.74	21.56	4.69	0.93	1.86	4.85	0.07	18,621
	Co10	0.03	5.34	31.93	3.46	0.95	2.08	4.39	1.4×10^3	22,232



Fig. 8. The normalized concentration C_t/C_0 change of MO solution during the degradation process of the as-spun (a) $Al_{85}Fe_{10}Y_5$ (Fe10) and (b) $Al_{85}Co_{10}Y_5$ (Co10) ribbons at different temperatures (T_r). (c) Derived reaction rate constants k at different reaction temperatures, the bottom right inset in (c): $lnk(T_r)$ vs $- 1/RT_r$ curves for the as-spun Fe10 and Co10 ribbons. (d) The reaction activation energy E_r and reaction rate constants k for the as-spun Fe10 and Co10 ribbons.

of Fe10 ribbon dropped sharply from the 4th cycle, while Co10 ribbon increased (Fig. 10(b) and (d)). After three degradation cycles, the amount of available zero-valent metals on the ribbon surface decreases and the degradation efficiency decreases [68]. However, since the iron element in the Fe10 ribbon forms iron oxide on the surface to prevent zero-valent metals from participating in degradation reactive [22], the

cobalt element in the Co10 ribbon is easy to combine with other metal elements to form a network structure and increase the specific surface area of the reaction, thus slowing down the degradation rate [73].

The homogeneity of the Co10 ribbon is higher than Fe10 ribbon (Figs. 1 and 2). According to free energy with concentration is binary $A_x B_{1-x}$ system, the spinodal occurs in the x = 0.5 adjacent and the



Fig. 9. The normalized concentration C_t/C_0 change of MO solution during the degradation process of (a) $A_{185}Fe_{10}Y_5$ (Fe10) and (b) $A_{185}Co_{10}Y_5$ (Co10) ribbons at different pH values. The degradation efficiency ($\eta = (1 - C_t/C_0 \times 100\%, t_r = 45 \text{ min})$ of the degradation process vs. pH values for (c) Fe10 and (d) Co10 ribbons.



Fig. 10. The normalized concentration C_t/C_0 change of MO solution during the degradation process of (a) $Al_{85}Fe_{10}Y_5$ (Fe10) and (b) $Al_{85}Co_{10}Y_5$ (Co10) ribbons from the 1st to the 7th degradation cycles. The degradation efficiency ($\eta = (1 - C_t/C_0 \times 100\%, t_r = 45 \text{ min})$ of the degradation process vs. reaction cycles for (c) Fe10 and (d) Co10 ribbons.

deviated zone from x = 0.5 can occurs metastable decomposition (nucleation and growth) [55,74]. Hence, the maze like structure and spinodal characteristic structure formed on Co10 ribbon surface (Fig. 4 (d)), while the spherical particles formed on the Fe10 ribbon surface (Fig. 4(c)). Apparently the depth of the groove in reacted Co10 ribbon surface is deeper than Fe10 ribbon surface, and the specific surface area of Co10 ribbon to join reaction is larger than Fe10 ribbon (Table 1). Hence, the stability and degradation efficiency of Co10 ribbon are higher than Fe10 ribbon.

Based on the thorough analyses of the elemental information, surface morphology and electronic structure of the Fe10 and Co10 ribbons during MO solution degradation, we have drawn the schematic diagram is illustrated in Fig. 12. The as-spun Fe10 ribbon in the acidic and alkaline MO solutions showed some spheroid particles on the surface of the ribbon, while the as-spun Co10 ribbon formed a developed maze like network structure on the surface, and through XPS analysis, the as-spun and reacted surfaces consist of Y and Y_2O_3 . Upon visible light irradiation, after adding as-spun Fe10 and Co10 ribbons into the acidic MO solution, metallic elements are ionized and generate electrons, which combine with H⁺ on the ribbon surface to generate [H]. And in the alkaline MO solution, the electrons produced by the ionization of the metal element combine with the OH⁻ to form a strong oxidizing •OH radical. Upon visible light irradiation, the Y_2O_3 on the ribbon surface combines with O_2 and electron to generate $\bullet O_2^-$ and then accelerate the degradation of MO solution. And the surface of Co10 ribbon has a maze like network structure, which can increase the specific surface area of reaction and provide more Y_2O_3 to generate $\bullet O_2^-$. By magnetic stirring force, the [H] or $\bullet OH$ or $\bullet O_2^-$ radicals are dispersed into the MO solution, and MO dye molecules are then degraded and mineralized into small molecules, including H₂O, CO₂, NO₃⁻ and SO₄²⁻. According to Ref [74], the higher homogeneity and stability of Co10 ribbon can form a maze like surface to increase the specific surface area of the reaction, and may improve the MO solution degradation efficiency and reusability in MO solution.

5. Conclusions

In this work, we have prepared $Al_{85}Fe_{10}Y_5$ (Fe10) and $Al_{85}Co_{10}Y_5$ (Co10) amorphous ribbons with melt spun method and we studied the microstructure, MO degradation behavior with various techniques.



Fig. 11. Comparable results of degradation efficiency of as-spun Co10 ribbon with and without adding quenching agents of 1,4-benzoquinone (BQ) and tertiary butanol (TBA) at (a) pH = 1 and (b) pH = 13.



Fig. 12. Schematic illustration of the degradation mechanism of MO dyes using the Al₈₅Fe₁₀Y₅ (Fe10) and Al₈₅Co₁₀Y₅ (Co10) amorphous ribbons.

We have found: the Fe10 ribbon has lower glass formability, stability and homogeneity than Co10 ribbon, the Co10 ribbon has a higher degradation efficiency in MO solution at various temperatures and rather lower/higher pH values. Co10 ribbon has a higher TOC removal rate in MO solution than Fe10 ribbon at pH = 1. The maze like structure on Co10 surface and spherical particles on the Fe10 surface can improve the degradation efficiency of MO solution especially at pH = 1. The Y₂O₃ on the as-spun and reacted ribbon surfaces can also catalyze the MO solution under visible light irradiation. The higher degradation efficiency of Co10 ribbon than Fe10 ribbon can be ascribed to the larger specific surface area. This work provides a scheme that can efficiently degrade MO solution, significantly improving the degradation rate of metallic catalysts in polluted water treatment, thus expanding the application field of Al-based alloys.

CRediT authorship contribution statement

Qi Chen wrote the article. Zhicheng Yan, Lingyu Guo and Hao Zhang did the help in the experiments. Laichang Zhang gave the key advices in organizing the article. Weimin Wang designed the experiment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was financially supported by the National Key Research Program of China (2016YFB0300501), National Natural Science Foundation of China (51471099, 51571132, 51511140291 and 51771103).

References

- [1] T.A. Saleh, I.B. Rachman, S.A. Ali, Sci. Rep. 7 (2017) 4573.
- [2] H. Lachheb, E. Puzenat, A. Houas, M. Ksibi, E. Elaloui, C. Guillard, J.M. Herrmann, Appl. Catal., B 39 (2002) 75.
- [3] F.L. Fu, D.D. Dionysiou, H. Liu, J. Hazard. Mater. 267 (2014) 194.
- [4] I.K. Konstantinou, T.A. Albanis, Appl. Catal., B 49 (2004) 1.
- [5] T. Robinson, G. McMullan, R. Marchant, P. Nigam, Bioresour. Technol. 77 (2001) 247.

- [6] H. Park, W. Choi, J. Photochem. Photobiol., A 159 (2003) 241.
- [7] S. Jayaraman, A.R. Warrier, J. Mol. Liq. 301 (2020) 112360.
 [8] C. Djilani, R. Zaghdoudi, F. Djazi, B. Bouchekima, A. Lallam, A. Modarressi, M. Rogalski, J. Taiwan Inst. Chem. Eng. 53 (2015) 112.
- [9] R.R. Pawar, Lalhmunsiama, P. Gupta, S.Y. Sawant, B. Shahmoradi, S.M. Lee, Int. J. Biol. Macromol. 114 (2018) 1315
- [10] P. Nigam, I.M. Banat, D. Singh, R. Marchant, Process Biochem. 31 (1996) 435.
- [11] M. Lucas, J. Peres, Dyes Pigments 71 (2006) 236.
- [12] A.G. Shende, C.S. Tiwari, T.H. Bhoyar, D. Vidyasagar, S.S. Umare, J. Mol. Liq. 296 (2019) 111771.
- [13] M. Ramya, M. Karthika, R. Selvakumar, B. Raj, K.R. Ravi, J. Alloys Compd. 696 (2017) 185.
- [14] W.J. Shen, H.L. Kang, Z.H. Ai, J. Hazard. Mater. 357 (2018) 408.
- [15] M.F. Khan, L. Yu, G. Achari, J.H. Tay, Chemosphere 222 (2019) 1.
- [16] N.K. Gupta, Y. Ghaffari, J. Bae, K.S. Kim, J. Mol. Liq. 301 (2020) 112473.
- [17] C.S. Bhatt, B. Nagaraj, A.K. Suresh, J. Mol. Liq. 242 (2017) 958.
- [18] N. Tara, M. Arslan, Z. Hussain, M. Iqbal, Q.M. Khan, M. Afzal, J. Clean. Prod. 217 (2019) 541.
- [19] S. Singh, M.D. Ediger, J.J. de Pablo, Nat. Mater. 12 (2013) 139.
- [20] H.B. Yu, Y. Luo, K. Samwer, Adv. Mater. 25 (2013) 5904.
- [21] C.Q. Zhang, H.F. Zhang, M.Q. Lv, Z.Q. Hu, J. Non-Cryst. Solids 356 (2010) 1703.
 [22] Q.Q. Wang, M.X. Chen, P.H. Lin, Z.Q. Cui, C.G. Chu, B.L. Shen, J. Mater. Chem. A 6 (2018) 10686.
- [23] C.Q. Zhang, Z.W. Zhu, H.F. Zhang, Q.L. Sun, K.G. Liu, Results Phys. 10 (2018) 1.
- [24] Z. Jia, J. Kang, W.C. Zhang, W.M. Wang, C. Yang, H. Sun, D. Habibi, L.C. Zhang, Appl. Catal., B 204 (2017) 537.
- [25] Z. Jia, S.X. Liang, W.C. Zhang, W.M. Wang, C. Yang, L.C. Zhang, J. Taiwan Inst. Chem. Eng. 71 (2017) 128.
- [26] Z. Jia, W.C. Zhang, W.M. Wang, D. Habibi, L.C. Zhang, Appl. Catal., B 192 (2016) 46.
 [27] J.Q. Wang, Y.H. Liu, M.W. Chen, G.Q. Xie, D.V. Louzguine Luzgin, A. Inoue, J.H.
- Perepezko, Adv. Funct. Mater. 22 (2012) 2567. [28] P. Liu, J.L. Zhang, M.Q. Zha, C.H. Shek, ACS Appl. Mater. Interfaces 6 (2014) 5500.
- [29] Y.J. Li, Y.G. Wang, B. An, H. Xu, Y. Liu, LC. Zhang, H.Y. Ma, W.M. Wang, PLoS One 11 (2016), e0146421.
- [30] J.Q. Wang, Y.H. Liu, M.W. Chen, D.V. Louzguine-Luzgin, A. Inoue, J.H. Perepezko, Sci. Rep. 2 (2012) 418.
- [31] Y.F. Zhao, J.J. Si, J.G. Song, Q. Yang, X.D. Hui, Mater. Sci. Eng. B 181 (2014) 46.
- [32] Q. Chen, J. Pang, Z.C. Yan, Y.H. Hu, L.Y. Guo, H. Zhang, L.C. Zhang, W.M. Wang, J. Non-Cryst. Solids 529 (2020), 119802, .
- [33] Q. Chen, Z.C. Yan, L.Y. Guo, H. Zhang, L.C. Zhang, K. Kim, X.Y. Li, W.M. Wang, J. Alloys Compd. 831 (2020). 154817.
- [34] A. Mondal, B. Adhikary, D. Mukherjee, Colloids Surf. A Physicochem. Eng. Asp. 482 (2015) 248.
- [35] P. Taneja, S. Sharma, A. Umar, S.K. Mehta, A.O. Ibhadon, S.K. Kansal, Mater. Chem. Phys. 211 (2018) 335.
- [36] Y.Y. Sha, I. Mathew, Q.Z. Cui, M. Clay, F. Gao, X.J. Zhang, Z.Y. Gu, Chemosphere 144 (2016) 1530.
- [37] S. Das, S. Garrison, S. Mukherjee, Adv. Eng. Mater. 18 (2016) 214.
- [38] P.P. Wang, J.Q. Wang, H. Li, H. Yang, J.T. Huo, J.G. Wang, C.T. Chang, X.M. Wang, R.W. Li, G. Wang, J. Alloys Compd. 701 (2017) 759.
- [39] K. Shaheen, H. Suo, Z. Shah, L. Khush, T. Árshad, S.B. Khan, M. Siddique, L. Ma, M. Liu, J. Cui, Y.T. Ji, Y. Wang, Mater. Chem. Phys. 244 (2020), 122748, .
- [40] B. Lin, X.F. Bian, P. Wang, G.P. Luo, J. Mater. Sci. Eng. B 177 (2012) 92.

O. Chen. Z. Yan. L. Guo et al.

- [41] S.X. Liang, Z. Iia, Y.J. Liu, W.C. Zhang, W.M. Wang, J. Lu, L.C. Zhang, Adv. Mater, 30 (2018), e1802764,
- [42] L.C. Zhang, Z. Jia, F. Lyu, S.X. Liang, J. Lu, Prog. Mater. Sci. 105 (2019) 100576.
- [43] Q. Chen, Z.C. Yan, H. Zhang, L.C. Zhang, H.J. Ma, W.L. Wang, W.M. Wang, Materials. 13 (2020) 3694
- [44] X.L. Li, Y.Y. Wu, S.T. Yang, X.J. Cha, P.C. Shao, L. Wang, J. Non-Cryst. Solids 503-504 (2019) 284
- [45] Z.W. Lv, Y.Q. Yan, C.C. Yuan, B. Huang, C. Yang, J. Ma, J.Q. Wang, L.S. Huo, Z.Q. Cui, X.L. Wang, W.H. Wang, B.L. Shen, Mater. Des. 194 (2020) 108876.
- [46] F. Wang, H. Wang, H.F. Zhang, Z.H. Dan, N. Weng, W.Y. Tang, F.X. Qin, J. Non-Cryst. Solids 491 (2018) 34.
- [47] S.X. Liang, X.Q. Wang, W.C. Zhang, Y.J. Liu, W.M. Wang, L.C. Zhang, Appl. Mater. Today 19 (2020), 100543,.
- [48] C. Li, L. Xu, Y.R. Zhao, J.L. Sun, D.P. He, H. Jiao, J. Alloys Compd. 627 (2015) 31.
 [49] T. Munawar, F. Mukhtar, M.S. Nadeem, K. Mahmood, A. Hussain, A. Ali, M.I. Arshad, M. Ajaz un Nabi, F. Iqbal, Solid State Sci. 106 (2020), 106307, .
- [50] C. Karunakaran, R. Dhanalakshmi, P. Anilkumar, J. Hazard. Mater. 167 (2009) 664.
- [51] J.B. Prasanna kumar, G. Ramgopal, Y.S. Vidya, K.S. Anantharaju, B. Daruka Prasad, S.C. Sharma, S.C. Prashantha, H.B. Premkumar, H. Nagabhushana, Spectrochimica acta. Part A, Spectrochim. Acta, Part A 141 (2015) 149.
- [52] X.N. Zhao, P. Wu, M. Liu, D.Z. Lu, J.L. Ming, C.H. Li, J.Q. Ding, Q.Y. Yan, P.F. Fang, Appl. Surf. Sci. 410 (2017) 134.
- [53] C. Karunakaran, R. Dhanalakshmi, P. Anilkumar, Radiat. Phys. Chem. 78 (2009) 173.
- [54] C.M. Magdalane, K. Kaviyarasu, J.J. Vijaya, B. Siddhardha, B. Jeyaraj, J. Kennedy, M. Maaza, J. Alloys Compd. 727 (2017) 1324.
- [55] Y.G. Wang, Y. Liu, Y.J. Li, B. An, G.H. Cao, S.F. Jin, Y.M. Sun, W.M. Wang, J. Mater. Sci. Technol. 30 (2014) 1262.
- [56] P.R. Gogate, A.B. Pandit, Adv. Environ. Res. 8 (2004) 501.

- [57] A. Deomartins, V. Canalli, C. Azevedo, M. Pires, Dves Pigments 68 (2006) 227.
- [58] S. Nam, P.G. Tratnyek, Water Res. 34 (2000) 1837.
- [59] L.M. Zhang, S.D. Zhang, A.L. Ma, H.X. Hu, Y.G. Zheng, B.J. Yang, J.Q. Wang, Corros. Sci. 144 (2018) 172.
- [60] C.L. Qin, Q.F. Hu, Y.Y. Li, Z.F. Wang, W.M. Zhao, D.V. Louzguine-Luzgin, A. Inoue, Mater. Sci. Eng. C Mater. Biol. Appl. 69 (2016) 513.
- [61] X.W. Yu, S.J. Shen, B. Jiang, Z.T. Jiang, H. Yang, F.S. Pan, Appl. Surf. Sci. 370 (2016) 357.
- [62] C.H. Lee, H.N. Park, Y.K. Lee, Y.S. Chung, S. Lee, H.I. Joh, Electrochem. Commun. 106 (2019) 106516.
- [63] F. Zhao, O. Amnuayphol, K.Y. Cheong, Y.H. Wong, J.Y. Jiang, C.F. Huang, Mater. Lett. 245 (2019) 174
- [64] C.G. Jia, J. Pang, S.P. Pan, Y.J. Zhang, K.B. Kim, J.Y. Qin, W.M. Wang, Corros. Sci. 147 (2019) 94.
- [65] B. Zberg, P.J. Uggowitzer, J.F. Loffler, Nat. Mater. 8 (2009) 887.
- [66] D. Monti, M. Meneghini, C. De Santi, A. Bojarska, P. Perlin, G. Meneghesso, E. Zanoni, Microelectron. Reliab. 88-90 (2018) 864.
- [67] S. Cho, M. Lee, Y. Jun, Res. Commun. 430 (2013) 787.
- [68] C.Q. Zhang, Z.W. Zhu, H.F. Zhang, Results Phys. 7 (2017) 2054.
- [69] B.W. Zhao, Z.W. Zhu, X.D. Qin, Z.K. Li, H.F. Zhang, J. Mater. Sci. Technol. 46 (2020) 88. [70] G.W. Pan, X.H. Jing, X.Y. Ding, Y.J. Shen, S.J. Xu, W.J. Miao, J. Alloys Compd. 809
- (2019), 151749, . [71] Z. Jia, X.G. Duan, W.C. Zhang, W.M. Wang, H.Q. Sun, S.B. Wang, L.C. Zhang, Sci. Rep. 6
- (2016) 38520.
- [72] Z. Jia, F. Lyu, L.C. Zhang, S. Zeng, S.X. Liang, Y.Y. Li, J. Lu, Sci. Rep. 9 (2019) 7636.
- [73] H.J. Chang, E.S. Park, W. Yook, D.H. Kim, Intermetallics 18 (2010) 1846.
- [74] J. Antonowicz, J. Mater. Sci. 45 (2010) 5040.