Applied Surface Science 466 (2019) 715–723

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

Preparation of 4,4′-diaminostilbene-2,2′-disulfonic acid derivative/ PVA/ LDHs composite fluorescent brightener and performances on paper surface

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A R T I C L E I N F O

Keywords:
Composite fluorescent brighter
DSD acid derivative
LDH
Anti-aging agent
Surface sizing agent

A B S T R A C T

In order to improve the anti-aging property and surface strength of high-yield-paper (HYP), a novel composite fluorescent brighter (PFB/PVA/LDH) was successfully synthesized via hydrothermal reaction with 4,4′-diaminostilbene-2,2′-disulfonic acid derivative (PFB), polyvinyl alcohol (PVA) and layered double hydroxides (LDH) as raw materials in this study. The introduction of LDH can effectively shield the ultraviolet to improve the anti-aging performance on one hand, and PVA will enhance the strength of paper though formation of thin film on the other hand. The anti-yellowing property of the as-prepared PFB/PVA/LDH was remarkably excellent than that of pure PFB for the HYP paper. After 70 h aging, the brightness of paper sheets coated with PFB/PVA/LDH is still reached to 91.24 ISO%. Moreover, the composite brighter could form a thin film in the surface of paper sheets to improve its surface strength and smoothness. Compared with blank paper sheets, the surface strength and smoothness of paper sheets treated with 0.4 $g\,m^{-2}$ of PFB/PVA/LDH were increased 2.11 $m\,s^{-1}$ and 56.8 s, respectively. Based on the characterization and results analysis, it can be inferred that the PFB/PVA/LDH composite can not only improve the paper's brightness as a fluorescent brightener and anti-aging agent, but also increase its surface strength as a surface sizing agent.

1. Introduction

To expand the use of high-yield pulp (HYP) is an ideal way to make full use of wood fiber resources in the paper industry, which is an effective project to treat the shortage of wood resources [1,2], alleviate the deterioration of ecological environment [3] and reduce the waste of by-production [4]. However, the most obstacle for the application of HYP is the color will be changed from white to yellow while irradiated by the nature light, which are caused by the lignin contained in HYP was oxidized by the ultraviolet light in the nature [5,6]. Cockram and co-workers revealed that the consumption of HYP will be 2–6 times higher than that in before if the whiteness of HYP is still higher than 97% (compared with the original HYP) in the future 3 to 12 months [7]. Therefore, to inhibit the color reversion of HYP is an effective strategy to solve the low consumption of HYP, which was expected by the papermakers over the years [8–10].

For many projects, to inhibit the color reversion through the introduction of additives in the HYP is considered as the most economically ones and can be obtained by industrialization [8,11]. The UV absorbers and radical scavenger are reported and demonstrated as the most effective additives on the suppressing of color reversion [12–14]. The 4,4′-diaminostilbene-2,2′-disulfonic acid (DSD acid) is a typical and common used ultraviolet absorber because it performed a good absorption property in the near ultraviolet region and re-emitted the blue light in visible region with longer wavelength [15,16]. Meantime, the yellow light, emitted from the high-yield pulp, is hybridized with the blue light for the emission of white light [17]. Therefore, the DSD acid and its derivatives are widely used to the color reversion of high-yield pulp in paper industry [16,18,19]. Maqbool reported that the series stilbene-triazine compounds exhibited a high degree of whiteness with moderate UV blocking properties and fairly good fastness properties [20]. Lee revealed the various derivatives of disodium 4,4′-bis(1,3,5-triazin-6-yl) diaminostilbene-2,2′-disulfonate showed a higher dyeing property than that of CI86 [21]. However, the above reported triazine-stilbene fluorescent brightening agent (FBA) compounds still had some drawbacks on the practical application, such as the single-function, poor performance on paper fiber surface and poor light stability. Based on above issues, our team had done a lot of work to achieve the multi-functionalization of FBA and improve FBA's performance on light stability and mechanical properties of paper. Liu synthesized a new style asymmetrical polymeric fluorescent brightener and confirmed that the polymeric fluorescent brightener was a type of high photo-stabile fluorophores [22]. Zheng prepared a polymeric fluorescent brightener agent based on DSD acid-triazine structure, which not only has higher

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https://doi.org/10.1016/j.apsusc.2018.10.088
Received 1 June 2018; Received in revised form 8 September 2018; Accepted 10 October 2018
Available online 11 October 2018

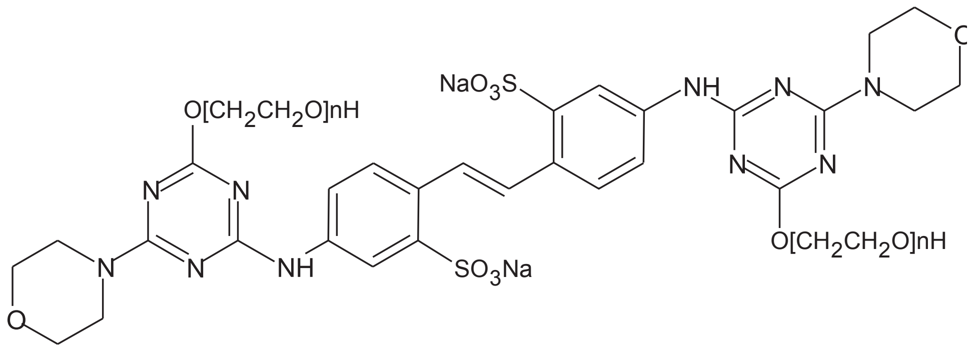

Fig. 1. The structure model of PFB.

hydrophilicity, light stability and brightness than those of FBA as light stabilizer and fluorescent brightener agent, but also enhances the surface strength and smoothness of paper as surface sizing agent [23]. Nonetheless, the above polymer fluorescent whitening agent still has the following: initial poor brightness problems, poor weather resistance, toxicity and complex operability.

In order to solve these problems, this experiment proposes to introduce inorganic UV shielding material into the polymeric brightener fluorescent (PFB) system. Layered double hydroxides (LDHs) as commonly used inorganic UV shielding material have been used in a variety of organic dyes [15,24]. Due to its structural characteristics, LDHs have better adsorption properties, UV shielding performance, flame retardant properties and bactericidal properties compared with other inorganic UV shielding materials (carbon black, titanium dioxide, zinc oxide and cerium oxide, etc) [25,26]. Moreover, LDHs has organic intercalation property, which can effectively avoid the human body directly contacted with organic matter to reduce the damage on human body [27]. Therefore, the zinc-aluminum layered double hydroxide (Zn_2Al-LDH) was introduced into the PFB system, which could produce a synergistic effect, to improve the initial whiteness and weather resistance of the paper. In addition, PVA was also combined with the above system to prepare a multi-functional inorganic-organic composite fluorescent whitening agent which could improve the surface strength of paper.

2. Experiment

2.1. Materials and equipment

All chemicals such as $Zn(NO_3)_2{\cdot}6H_2O$, $Al(NO_3)_3{\cdot}9H_2O$, $NaNO_3$, cyanuric chloride, 4,4′-diaminostilbene-2,2′-disulfonic acid (DSD acid), sodium hydroxide, absolute ethyl alcohol, acetone, ethanolamine, polyving akohol (PVA) and so on, were used as purchased from commercial sources from Xi'an, and were of industrial or analytical grade and used without further purification. All the aqueous solutions were prepared with deionized and CO_2-free water.

The FT-IR (KBr, σ/cm^{-1}) spectra was carried out by using VECTOR-22 (Bruker Corporation, Germany). The UV–Vis spectra was obtained on a Cary 100 UV–Visible spectrophotometer with an integrating sphere detector, and $BaSO_4$ was used as the reflectance standard material. Fluorescence spectra were measured on a Fluorolog spectrofluorometer (Horiba, France). The brightness was recorded on a ZB-A colorimeter (Hangzhou Zhibang Instrument Co. Ltd, China). UV accelerated aging test was completed by using ZN-100 N desktop UV light resistance climate chamber (Xi'an Tongsheng Instrument Manufacturing Co. Ltd, China). Patterns used in the experiment was obtained by paper machine (Shaanxi University of Science and Mechanical Equipment Factory, China). The strength of paper was measured by the J-IGT350 printability tester (Sichuan Changjiang Instrument Co. Ltd, China).

2.2. Synthesis of the compounds

Zn-Al-NO_3-LDH (referred to as LDH) was synthesized using a traditional coprecipitation method [28]. Distilled and deionized water was used in all preparations. The water was boiled immediately before use and all synthesis experiments were carried out under a stream of N_2 in order to minimize carbonate contamination. A metal precursor solution containing 7.465 g $Zn(NO_3)_2{\cdot}6H_2O$ and 4.700 g $Al(NO_3)_3{\cdot}9H_2O$ in 50 ml H_2O was added drop-wise into the anion solution containing 2.125 g $NaNO_3$ in 50 ml H_2O, while the pH was kept constant at *ca.* 10 using a NaOH (4 M) solution. The mixture was aged at room temperature for 24 h, followed by centrifugation and washing with water until the pH was close to 7. The obtained LDH was dried at 70 °C for 24 h.

The 4,4′-bis (1,3,5-triazinyl)-diaminostilbene-2,2′-disulfonic acid derivatives (PFB), previously reported by our group, was selected as one of raw materials in this paper and its structure was shown in Fig. 1 [18]. Typically, in a 100 ml beaker, 0.05 g PFB and different amount of LDH (0.00, 0.02, 0.04, 0.05, 0.06, 0.08 g) was added in 50 ml of DI water and dispersed with ultrasonic dispersion instrument. Then, 2.0 g PVA-1788 was added in above mixture and heated at 100 °C. After the PVA dissolved completely, the mixture was transferred to a Teflon-sealed autoclave and heated at 110 °C for 10 h. After cooling to room temperature, the obtained resultant milk white liquid was target product (PFB/PVA/LDH).

2.3. Applied experiment of paper coating

The poplar chemi-mechanical pulp (Yueyang forest & paper Co., LTD, China) was dispersed into the distilled water and the percentage of pulp was adjusted to 10% in mass fraction. Then the H_2O_2 (1%), EDTA (0.05%), and sodium silicate ($Na_2SiO_3{\cdot}9H_2O$, 0.5%) were introduced into the solution in sequence and the pH was adjusted to 9 to 10. After that, the above solution was kept in a constant temperature (70 °C) with 90 min for the reaction and then washed until neutral with distilled water. The pulp solution was used to make the handsheets (100 $g\,m^{-2}$) and cut it into pieces (68 mm × 73 mm). The synthesized compounds were directly coated on the handsheets used the coating machine with an opaque background and the samples were dried at room temperature [29].

2.4. UV accelerated aging test

The brightness of the hand sheets was measured by a ZB-A colorimeter (Hangzhou Zhibang Instrument Co., Ltd., Hangzhou, China) at λ = 457 nm. The brightness numerical value was the average value of 5 times the determination. Paper anti-UV aging testing was performed using a ZN-100 N UV light resistant climate chamber (tube of UV-340). The test conditions were as follows: 25 °C, wavelength of the UV lamp was 340 nm, output power was 5.3 $mW\,cm^{-2}$, radiation for 36 h, and the distance between the samples and the lamp was 30 cm. The