

Review of Study on Resin Dye-Fixatives on Cotton Fabrics

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Abstract

This paper introduced the study on preparation and properties of resin dye-fixatives on cotton fabrics in detail, and analysize the relations between their different structures, molecular weight and their fastness properties, from which some disciplines were obtained. Lastly, some suggestions on resolving the shortcomings of resin dye-fixatives on cotton fabrics were given according to the liturature results.

Keywords: Dye-fixatives, Molecular weight bigness and distribution, Structure characteristics, Fastness properties

Introduction

Cotton fabric is the most widely used textiles in the world, accounting for more than 50% of total consumption, which is mainly made up of cellulose (scheme 1)(Wang H F, Cheng Y Y, and Lin H., 2003). Several classes of dyes can be used to dye cellulosic fibres, namely vat, direct, reactive, sulphur colorants, the different classes varying in terms of factors such as their cost, ease of application, fastness properties, etc. Characteristically, reactive dyes furnish a wide gamut of shades of good light fastness and excellent wash fastness on cellulosic fibres. However, although reactive dyes are widely used for the exhaustion dyeing of cotton and other cellulosic fibres, the dyes suffer the disadvantage that dye-fibre reaction is not 100% efficient. This may be attributed to the incomplete exhaustion of the dye, to incomplete reaction of the adsorbed dye with the fibre and, more importantly, because the dyes can react not only with the fibre nucleophile (cellulosate anion) but also with nucleophiles (commonly hydroxyl ions) present in the dyebath, to dye hydrolysis(Zhang J. 2004). Therefore, dye fixing agents (dye-fixatives) should be used to improve the fastness properties of dyes on cotton fibrics. The resin fixtives on cotton fabrics consist of dicyano resin dye-fixatives, polyamine resin dye-fixatives, polycationic dye-fixatives, and reactive resin dye-fixatives, etc.

1. Dicyano resin dye-fixatives

Dicyano resin dye-fixatives were developed in 1950s, these resin dye-fixatives of which Dye-fixative Y was a classical example (Ye J X. 2003), which was synthesized as shown in scheme 2. The color fastness was in fact improved by a methylene bridge formed by reaction between the terminal methylol and the hydroxy group of cellulose. Extensive research work has shown that formaldehyde-based resin finished products release formaldehyde into the atmosphere directly or during processing, handling, garment manufacturing and subsequent wearing of textiles due to the hydrolysis of untreated or partically crosslinked N-methylol derivatives present on the fibre(Cui Y F, Zeng F S, and Ning P. 1994). Direct release of formaldehyde into the work environment causes severe irritation to eyes, nasal passages and respiratory tracth while an untreacted or partically crosslinked resin causes an allergenic response of the skin upon continuous handling of textiles. For reasons of these health problems associated with formaldehyde , there was an increasing demand for non-formaldehyde dye fixing agent(Huo Y. 2006).

2. Polyamine resin dye-fixatives

Polyamine resin dye-fixatives were developed in 1960s, which was the earliest non-formaldehyde dye fixing agent and was synthesized by polycondensation of diethylenetriamine and dicyandiamide as shown in scheme 3. These dye-fixatives were reported to improve overall fastness properties, without affecting the tone and depth of shades of dyes on cotton substrates. The results indicated that formaldehyde-based dye fixing agents would be replaced by developed polyamine resin fixatives(Huang M F. 2000).

In 1984, Juerg used the formaldehyde-free dve fixing agent SH-96, which was the polycondensate of diethylenetriamine and dicyandiamide, to treat the cotton fabric dved by reactive dves, the result showed that the fixtive SH-96 was a kind of effective fixing agent(Juerg H, Bruno K. 1984). Extensive study showed addition of the component which could be croolinked with the cellulose could improve the fastness properties of these dye-fixatives. Xu Wei added the polycondensate of dimethylamine and epichlorohydrin into the polycondensate of diethylenetriamine and dicyandiamide to obtain dye fixing agent AE, which was used for fixing experiment of reactive dyes on cotton fabric, the result showed that the dry rubbing fastness was improved from 2-3 grade when untreated to 4-5 grade, the wet rubbing fastness was improved from 1-2 grade when untreated to 4 grade(Xu W. 2001). Dye fixing agent ENF was synthesized by pre-polymerization of diethylenetriamine with dicyandiamide and reaction with dimethylamine and epichlorohydrin at 55-60 °C for 4 h, followed by adjustment of pH to 6-7 with glacial acetic acid and dilution with de-ionized water. When dye fixing agent ENF was used for fixing experiment of Direct Big Red 4BS and Direct Big Red EX on cotton fabric, the dry rubbing fastness reached both 4-5 grades, the wet rubbing fastness reached both 4 grades. When dye fixing agent ENF was used for fixing experiment of Reactive Black KE-GR and Reactive Green KE-4B on cotton fabric, the dry rubbing fastness reached both 4-5 grades, the wet rubbing fastness reached both 4-5 grade and 4 grade respectively (Yang J P. 2005). The good color fastness properties of these dve-fixatives may be that there were both cationic groups which could interact with anionic dyes and reactive groups which could be crosslinked with cellulose in the dye-fixatives' structures.

The fastness properties of polyamine resin dye-fixatives are not only related to their structure chacterics, but also related to their molecular weight bigness and distribution. In 2008, Deng Yaoming studied the difference of the fastness properties of polyamine resin dye-fixatives with different molecular weight bigness and distribution, the results showed that the fastness properties of the fixtives with higher molecular weight and narrow distribution were better than those with lower molecular weight and wide distribution, however, the molecular weight of these resin fixtives can't be so high that the gel or precipitate forming affect the fixing result(Deng Y M, Pi P, Wen X F. 2008). Therefore, the molecular weight should be controlled in a certain range. The dye fixing agents with a certain molecular weight bigness and distribution can be synthesized by controlling the preparation conditions, thus, preparation conditions of these dye-fixatives would affect their fastness properties.

However, these dye-fixatives would happen discoloration during processing, which may be due to the oxidation of primary amine, secondary amine, and tertiary amine in the structure (scheme 4).

3. Polycationic dye-fixatives

From the late of 1960s, many cationic polymers have been applied to cotton fabrics with a view to enhance the uptake of anionic dyes and the mechansim of inteeractions involved can be interpreted by the participation of electrostatic forces between the dyes and the basic cationic groups in the polymer. Poly (dimethyldiallylammonium chloride) (PDMDAAC) dye-fixative would be one of the most optimum polycationic fixtives, which is the polymer of monomer dimethyldiallylammonium chloride (DMDAAC)(Wandrey C, Hernandez B J,Hunkerler D. 1999), composed of five-membered pyrrolidone rings, six-membered piperdine rings, and little branched structures (shown in scheme 5). Electron spin resonance spectroscopy and ¹³C NMR spectroscopy have confirmed that PDMDAAC consists of a 5:1 ratio of five-membered pyrrolidone rings to six-membered piperdine rings(Rosunee S, Carre C M. 2003). Cellulose and dimethyldiallylammonium chloride have similar conformational structures, and this would be expected to contribute to strong interactions (scheme 6)(Blackburn R S, Burkinshaw S M. 2003), thus, PDMDAAC dye-fixative was thought to be applied in the fixing of direct dyes(Burkinshaw S M and Gotsopoulos A. 1999), sulphur dyes(Burkinshaw S M and Gotsopoulos A. 1996), phthalocyanine dyes(Shinbo K, Onishi K, Miyabayashi S, et al. 2003), azo dyes(Nizri G, Lagerge S, Kamyshny A. 2008), and reactive dyes on cotton fabrics(Burkinshaw S M and Katsarelias D. 1995).

In 1970s, PDMDAAC dye-fixative (Dye-fixative F) was firstly developed by Calgon and Halliburton company. In 1981, Nippon Senka Kogyo company used 0.1% (o.m.f) PDMDAAC to treat cotton fabric dyed with Remazol Black B, the results showed that wash fastnenss was improved from 1-2 grade when untreated to 4-5 grade(Kishioka H. 981). In 1994, Xu used 0.1-0.2% (o.m.f) PDMDAAC to treat cotton fabric dyed with Direct Black G, the results showed that the dry rubbing fastness reached 4-5 grade, the wet rubbing fastness reached 4 grade, and the use level of this fixtive was only 1/10-1/5 of Fixtive Y(Xu D P, Qian L Z, and Cong J L. 1994). In 2005, Zhang used 1% (o.m.f) PDMDAAC to treat cotton fabric dyed with Reactive Red 3BS and Reactive Red 3IS, the results showed that the dry rubbing fastness reached 4 grade, the wet rubbing fastness reached 3 grade, color fastness to soaping reached 5 grade, white fabric staining reached 5 grade, chloride fastness reached 5 grade, and no discoloration was found. However, the defect of these dye-fixatives is their bad rubbing fastness(Zhang X, Mao B, and Xiang M H. 2005).

The fastness properties of PDMDAAC were affected by the molecular weight bigness and distribution. It's reported in literature that the fastness of one certain distributed PDMDAAC fraction can reach 5 grade, but the fastness of undistributed PDMDAAC is only near to Dye-fixative Y(Zhang J G, 1992). Komiya Kaoru used the PDMDAAC with the viscosity of 1.23 dL/g and 0.61 dL/g to treat the cotton fabric dyed with Remazol Brilliant Blue R, the results

showed that the wash fastness of PDMDAAC with the viscosity of 1.23 dL/g reached 5 grade, while the wash fastness of PDMDAAC with the viscosity of 0.61 dL/g reached 4 grade, indicating that the molecular weight had effect on the fastness of the fixtive(Komiya K, Kanai S, Beppu K. 1988). PDMDAAC fixtives with a certain molecular weight bigness and distribution can be synthesized by controlling the preparation conditions, thus, it is very important for the fastness properties of PDMDAAC dye-fixatives to control their preparation conditions.

Addition of functional groups which can be reacted with dyes or cellulose into the PDMDAAC's chain would improve its fastness. Collins G W used the copolymer of allylamine and DMDAAC to treat cotton dyed with reactive dyes, color fastness to soaping and white fabric staining both reached 5 grade(Collins G W, Burkinshaw S M, Gordon R. 2001), the good fastness properties can be interpreted by reasons as follows(Iwata M, Saka T, Yamato H, et al. 1986): (1) the anionic dye molecules are attracted to the cationic sites in PDMDAAC, (2) the reaction between the amino-functional nucleophiles and the reactive group in the dye proceeds via a nucleophilic substitution mechanism. Daniher used the copolymer of DMDAAC and N-methylolacrylamide to treat the cotton fabric dyed with direct dyes, white fabric staining was improved from 3 grade when untreated to 4-5 grade(Daniher F A, Aspland J R. 1988). Cheng synthesized Fixative PAS by compolymerization of DMDAAC and SO₂ and used 0.5-1.5% (o.m.f) of PAS to treat the cotton fabric dyed with Reactive Green X-BR, the results showed that color fastness to soaping reached 4-5 grade, white fabric staining reached 4 grade, dry rubbing fastness and wet rubbing fastness reached both 4 grade, and no discoloration happened(Cheng H X, Yao Q C, and Yi X J. 2004). Yang synthesized the polyamine by the reaction of methylamine with epoxychloropropane at pH=6, 65 °C for 2 h, then mixed the product with poly (dimethyldiallylammonium chloride- acrylamine) (PDA) to give a complex Dye-fixative FHAD for reactive dyes, if the dye-fixative FHAD fixed at 50 °C for 20 min, the fixation of reactive dyes would have improved by 25-66%, and their color washing fastness was promoted from 3 grade to 5 grade(Yang J P. 2004). Wang synthesized the polymer CR by the compolymerization of DMDAAC and allylamine, then mixed the product with polyamine CS to give Fixative CRS, and used Fixtive CRS to treat the cotton dyed with reactive dyes, the results showed that chloride fastness was improved 1 grade(Wang Y J, Gong Z L, Liu X, et al. 2008).

4. Reactive resin dye-fixatives

These resin dye-fixatives with the groups which can react with dyes or cellulose is named as reactive resin dye-fixatives. reactive resin dye-fixatives consist of epichlorohydrin derived resin dye-fixatives, polyurethane dye-fixatives, and the reactive resin dye-fixatives containing organo-siloxsne units, and carbon-carbon double bond, etc. epichlorohydrin derived resin dye-fixatives contain the reactive groups which derive from epichlorohydrin, which are most widely used reactive resin dye-fixatives, the reactive groups derived from epichlorohydrin can be crosslinked with cotton fabrics (cellulose), resulting in the enhance of fastness properties. In 1999, Wang used the product which was synthesized by the reaction of polycondensate of diethylenetriamine and dicyandiamide with epichlorohydrin to treat the cotton fabrics dyed with Direct Big Red 4BS and Reactive Green X-BR, the results showed that dry rubbing fastness reached 4-5 grade and 4 grade respectively, wet rubbing fastness reached both 3 grade, and no discoloration happened (Wang C M, Hu X L. 1999). In 2001, Zhuang synthesized formaldehyde-free dye fixing agent CS by polycondensation of diethylenetriamine and epichlorohydrin, and used the product to treat the cotton fabric dyed with Reactive Green B-2GLN, the result showed that dry rubbing fastness reached 4-5 grade, wet rubbing fastness reached 4 grade, and no discoloration happened(Zhuang B L. 2001). In 2002, Guo polymerize the monomer which was synthesized by diethylenetriamine, epichlorohydrin, and acrylchloride to give Dye-fixative YSZ, and used the product to treat the cotton fabric dyed with Direct Lake Green 6B and Direct Big Red 4BS, the results showed that the dry rubbing fastness reached both 4-5 grades, the wet rubbing fastness reached both 4 grades(Guo Z L, Meng Y F, and Guo X K. 2002). In 2006, Jia synthesized the Fixative JXH by polycondensation of diethylenetriamine, dicyandiamide, and epichlorohydrin, and used the product to treat the cotton fabric dyed with Direct Cu Green 2, the result showed that the dry rubbing fastness reached 4-5 grade, the wet rubbing fastness reached 4-5 grade(Jia S T, Ju X H, Cao D X, et al. 2006). In 2007, Zhang synthesized reactive dye fixing agent X by polycondensation of triethyltetraamine, epichlorohydrin, and butylacrylate, and used the product to treat the cotton fabric dyed with reactive dyes and direct dyes, the result showed that the dry rubbing fastness and wet rubbing fastness were both improved 1-2 grades, wash fastness was also improved 1 grade(Zhang X L, Zhao Z H. 2007).

Polyurethane dye-fixatives contain isocyanic ester units which can react with the hydroxy group of cellulose very easily, resulting in enhance of fastness. In 2002, He synthesized the pre-polyurethane by polycondensation of polypropylene glycol (PPG), polyoxyethylene glycol (PEG), dihydroxypropionic acid, and TDI, then reacted with diethylenetriamine and epichlorohydrin, when the product was used to treat the cotton fabric dyed with Direct Lake Green SB, the wash fastness was improved from 3-4 grade to 4 grade, the dry rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 3-4 grade to over 4 grade, when the product was used to treat the cotton fabric dyed with Reactive Green EF-2G, the wash fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the wet rubbing fastness was improved from 4 grade to 5 grade, the W, Xu H Y, and Hua Z W. 2002). In 2009, Hu synthesized the polyurethane dye-fixative by polycondensation of polyoxyethylene

glycol (PEG), dihydroxypropionic acid, and TDI, when the product was used to treat the cotton fabric dyed with Reactive Big Red 4BS, the dry rubbing fastness reached 5 grade, the wet rubbing fastness reached 4 grade, when the product was used to treat the cotton fabric dyed with Reactive Green EF22G, the dry rubbing fastness reached 4 grade, the wet rubbing fastness reached 3 grade(Hu Y Y, Guo R, and Jia S T. 2009).

Recently, some novel reactive resin dye-fixatives containing organo-siloxsne units, and carbon-carbon double bond have been successfully developed. In 2004, novel cationic and reactive resin dye-fixatives with organo-siloxsne units were synthesized by polycondensation of silicol, dimethyldichlorosilane, amine, siloxane, and quaternized reagent. There were both cationic groups which could interact with anionic dyes and organo-siloxsne units which could crosslinked with the hydroxy group of cellulose in their structures, therefore, these fixtives have good fastness properties(Wang L M, Fan Z Y, and Lin J. 2004). In 2005, Liu synthesized novel reactive dye-fixatives with carbon-carbon double bonds in their structures could be self-polymerized into film on cotton fabrics, resulting in the enhance of fastness(Liu X, Ding W W, Zhang Z, et al. 2005).

The above-mentioned studies showed that addition of reactive groups into the dye-fixatives could improve their fastness, in addition, the fastness of reactive resin dye-fixatives are also related to their molecular weight bigness and distribution. In 2008, Li studied the difference of these fixtives with different inherent viscosity which were synthesized with triethyltetramine, butylacrylate and epichlorohydrin, etc, the results showed that the dye-fixatives had better effect when the limiting inherent viscosity was 1.7575 dL/g, and had bad effevt when the inherent viscosity was more or less than 1.7575 dL/g(Li W X, Hong H X, and Zhang H. 2008). Therefore, in order to gain better fastness of these dye-fixatives, their molecular weight bigness and distribution should be also controlled.

5. Conclusions

By now, the fastness properties of resin dye-fixatives on cotton fabrics have been made a certain improvement, but the rubbing fastness, light fastness, and chloride fastness are not still optimum. Extensive studies show that the fastness properties of resin dye-fixatives on cotton fabrics are not only related to their structure chacterics, but also related to their molecular weight bigness and distribution. Therefore, in order to gain better fastness of these dye-fixatives, some suggestions are given as follows:

(1) Addition of functional groups which can be reacted with dyes or cellulose into these dye-fixatives would improve their fastness.

(2) The molecular weight of these dye-fixatives should be also controlled.

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Scheme 1. structure of main component of cotton fabric (cellulose)



 $X = Cl^-, CH_3COO^-, et al$





Scheme 3. synthesis of polyamine dye-fixative



Scheme 4. discoloration mechansim of polyamine dye-fixative



I = initiator Scheme 5. Synthesis and structures of PDMDAAC dye-fixative



R = dyeScheme 6. fixing mechansim of PDMDAAC dye-fixative on cotton fabric