

Effects of Polycarboxylic Acids on Untreated Cotton and Solubilised Sulfur Dyed Cotton

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ABSTRACT

A number of polycarboxylic acids (butane tetracarboxylic acid (BTCA), polyacrylic-co-maleic acid (PAMA), citric acid, maleic-itaconic (MA-IA)) were examined in combination with sodium hypophosphite (SHP) in order to give a similar high fabric crease recovery performance and whiteness index to traditional formaldehyde based crease agents. BTCA showed to be the best crease resist agent for crosslinking cotton fabric with SHP. In addition, BTCA treatment offers higher tear/ tensile strength compared to fabrics treated with dimethylol dihydroxyethylene urea (DMDHEU). FTIR/FT-RAMAN spectroscopies showed the presence of intermolecular ester crosslinks in cotton fabrics treated with BTCA. Treatment of Jarosol solubilised sulfur dyed cotton fabrics with PAMA/BTCA and SHP resulted in a color change of dyed fabrics because of the reductive nature of SHP. Jarosol solubilised sulfur dyed cotton fabric treated with BTCA/PAMA improved wash fastness due to dye locking inside the fibres, but light fastness did not change. Prog. Color Colorants Coat. 7(2014), 1-9. © Institute for Color Science and Technology.

1. Introduction

Because of concern for occupational hazards from formaldehyde in the work place, the textile industry has maintained an interest in formaldehyde-free crosslinking agents for producing durable press properties in cellulosic materials [1,2,3]. Polycarboxylic acids (formaldehyde-free) can be used as an alternative to the traditionally used formaldehyde based cellulose crosslinking agents, i.e. dimethylol dihydroxyethylene urea (DMDHEU) [4,5,6,7]. Sulfur dyes are one of the most popular of all dye classes on cellulosic fibres and their blends [8]. Very limited information is available

on the effects of polycarboxylic acid finishing on sulfur-dyed fabrics. This paper focuses on the color change and color fastness of a cotton fabric after application of soluble sulfur dyes and subsequent finishing with BTCA or Poly acrylic-co-maleic acid. This paper has also investigated the mechanical properties of crosslinked cotton fabric and the use of FT-IR and FT-Raman to study the BTCA-cross linked cotton.

2. Experimental

2.1. Treatment of polycarboxylic acid as zero-

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formaldehyde crosslinking agent on cotton fabric

The fabric was plain cotton bleached/mercerised fabric, 135 g/m², 60/60 warp/weft threads with a 20' carded weft supplied by Phoenix Calico Limited, UK. 1, 2, 3, 4-Butane tetracarboxylic acid (BTCA, MW=234.16), Citric acid (CA, MW=192.12), Maleic (MA, MW= 116.07), Itaconic acid (IA, MW =130.10), Polyacrylic-co-maleic acid (PAMA, MW=3000), and Sodium hypophosphite hydrate (SHP, MW=105.9) were supplied from Aldrich.

A conventional dimethylol dihydroxyethylene urea (DMDHEU) type easy-care crosslinking finish (Permafresh LF, having an active value of 50%) and its catalyst LO based on magnesium chloride were supplied from Warwick Chemicals plc.

Cotton fabric was wetted by immersion in the treatment bath (containing crosslinking agent and catalyst), and then passed between the nip rollers. The pressure of the nip rollers was sufficient to give a wet pick up of 75-80% based on the original weight of fabric sample (o.w.f). The fabric was then subjected to drying and heat treatment in a stenter at a specified time mentioned later.

For the highest wrinkle angle, the concentration of each crosslinking agent and catalyst was:

8% BTCA (g/100ml), mole ratio BTCA/SHP=0.6;

10% CA (g/100ml), mole ratio CA/SHP=0.8;

13% MA-IA (g/100ml), mole ratio MA/IA=1, mole ratio acid/SHP=1.2;

25% PAMA (g/100ml), mole ratio PAM/SHP=0.05;

Typically with polycarboxylic acids, the drying time was 5 min at 85°C and the curing was done at 180°C for 90 sec.

For comparison, the fabric was treated with conventional crosslinking agents (12% Permafresh LF (g/100ml)) and 30% Catalyst LO on weight of Permafresh LF). The recommended conditions by the supplier were drying at 85°C for 5 min and curing at 160°C for 2/3 min.

2.2. Jarofast Dyeing system

Jarofast Dyeing system was developed by James Robinson Ltd. and allows quick process time using water soluble sulfur dyes (Jarosol) on a substrate pre-treated with Jarofix 391, a cationic fixing auxiliary. As no reducing or oxidising agents are required, the system has less environmental impact [9].

Chemicals and Dyes used in dyeing: Jarosol

Bordeaux JB (C.I. Sol.Sulfur Red 6), Jarosol Orange JG (C.I.Sol. Sulfur Orange 5), Jarosol Yellow JR (C.I.Sol. Sulfur Yellow 23), Jarofix 391 (cationic fixing agent). Pre-treatment and dyeing were carried out in a Roaches Rotohose model 50 L dyeing machine. All the processes (pre-treatment and dyeing) were carried out separately for each three sulfur dyes.

• Pre-treatment with Jarofix 391

The Pre-treatment bath containing 0.5% o.w.f. non-ionic wetting agent was set at 30°C. The fabrics were added and circulated for 5-10 min. 2% o.w.f. Jarofix 391 was added and circulated for 5-10 min. The temperature was raised to 60°C over a minimum of 10 min and maintained at 60 °C for 15 min. The bath was dropped, refilled with clean water, rinsed for 5 min and finally the fabric was dried in tumble dryer.

• Jarosol Dyeing Method

The pre-treated material was added to the bath, run for 10 min and temperature raised to 30°C. 2% o.w.f. Jarosol dye dissolved in water, added to the dyebath and circulated for 10 min, at PH 7. The temperature was raised to 95° C at 2°C/min and maintained at 95°C for 30 min. 1% acetic acid was added and the dyebath cooled down to 70°C. The fabric was rinsed for 15 min in warm running tap water and tumble-dried prior to fabric evaluation.

(Calculations were based on a liquor ratio 40:1 o.w.f.= on weight of fabric.)

2.3. BTCA/PAMA crosslinking Treatments on Solubilised Sulfur Dyed Cotton Fabrics

The aim of the experiment was to study the effects of BTCA and polyacrylic-co-maleic acid (PAMA) with sulfur dyed cotton fabrics on shade changes, wash fastness and light fastness. Each sulfur dyed fabric was divided into three pieces;

(1) The first piece of the sulfur dyed cotton fabrics was used as reference sample;

(2) The second piece of the sulfur dyed fabric was treated with 8% BTCA, SHP (mole ratio BTCA/SHP= 0.6). The treated dyed samples were dried at 85°C for 5 min and cured at 180° C for 90 seconds. The dyed crease resist samples were rinsed in running water and tumble-dried

(3) The third piece of the sulfur dyed fabric was treated with 25% PAMA, SHP (mole ratio

PAMA/SHP=0.05). The drying, curing and rinsing conditions were the same as the second piece.

2. 4. Equipment and Tests

- **Martindale abrasion test**

The flat abrasion of fabric was performed using the Martindale abrasion tester according to BS 5690: 1991.

- **Instron Tensile Testing**

Fabric testing was carried out according to method BS 2576: 1986 on an Instron 1122 (constant rate of elongation) under standard conditions.

- **Shirley Crease Recovery Test**

Crease recovery angle was performed using Shirley crease recovery tester according to BS 3086 (1972).

- **Whiteness measurement**

Whiteness was measured on a MVE-P Macbeth reflectance spectrophotometer with 10° standard observer and illuminant D65.

- **Attenuated Total Reflectance (ATR) spectroscopy**

A Nicolet Magna-IR system 750 spectrometer with associated ATR was used for all the infrared spectra. Resolution was 4cm⁻¹ and there were 500 scans for each spectrum. Using the technique, a sample finished fabric was placed on each side of a thallium /iodide (KRS-5) crystal to maximize the number of reflections at the sample interface.

- **Fourier Transform Raman Spectroscopy (Ft-Raman)**

The spectrometer was a standard Nicolet system 950 with a neodymium YAG laser as a light source operating at 1064 nm (near infrared). The fabric sample (approximately 2cm × 2cm) was directly mounted on stainless steel post with double sided tape.

2.4.1. Fastness to washing

Fastness to washing tests, according to BS 1006:C06(1990), were carried out in the Washtec-A2 series machine. Each sample (10cm × 4cm) was sewn to a piece of SDC multi-fibre adjacent fabric along the shorter edge. Washtec-A2 containing a standard detergent powder of 4 g per litre was heated to 60° C. In C06/C2S,

1g/l sodium perborate was also added. The composite was put in a pot, 25 steel balls was added and maintained at 60°C for 30 min. The composite was rinsed and stitches were opened. The sample and multi-fibre swatch dried at room temperature.

The staining of adjacent fabric was assessed against the grey scale, and the sample was assessed for color change by grey scale and color difference values.

2.4.2. Assessment of wash fastness

- **The color change**

Two different methods were used to examine the color change of wash fastness treated and control of dyed fabric however there are other methods reported [10, 11]:

(1) Grey scale: For the assessments, a rating scale of 1 (poor) to 5 (excellent) was used.

(2) Color difference values (ΔE): ΔE was measured by using MVE-P Macbeth reflectance spectrophotometer using CIE CMC 2:1 color difference formula under illuminant D65, using a 10° standard observer.

- **Staining**

A different "grey scale" from that above is required to assess the degree of staining. The scale consists of one pair of white and four pairs of grey and white color panels. The white pair is rated No. 5 and is equivalent to no staining have occurred and No. 1 being the most heavily stained.

2.4.3. Fastness to light and its assessment

Light fastness was measured according to BS 1006: B02 (1990) in a Xenotest machine. The dyed sample was exposed to artificial light along with Blue Wool fabric References. Light fastness was assessed by comparing the color change of the sample with the Blue Scale 1-8. No. 1 has low light fastness and No. 8 has high light fastness.

3. Results and discussion

3.1 Comparative study of the properties of fabrics treated with polycarboxylic acids (formaldehyde-free crosslinking agent) and DMDHEU

An improvement in dry/wet crease recovery angle values of fabrics treated with all crosslinking agents was evident, Table 1 shows the greatest improvement in

fabric crease recovery angle with BTCA and DMDHEU while citric acid had the lowest degree. Crosslinked cotton fabrics imparted higher dry crease recovery angle than the wet one. This difference is probably due to the breakdown of the accessible hydrogen bonds, which normally operate in combination with the chemical bonds in the treated fabrics. When the fabric sample becomes wet during wet crease angle, the fibre molecules swell and cause the crosslinks to be subject to strain, resulting in decreasing the wet crease recovery angle [12].

Comparison of whiteness index showed that BTCA, DMDHEU and PAMA maintained higher WI than citric acid and MA-IA. Citric acid is an alpha-hydroxy acid and the low whiteness index of the citric acid treated fabric at high temperature presumably is from dehydration of citric acid residues and/or from trace amounts of sugars as impurities [4]. The MA/IA system exhibited lower whiteness possibly due to oxidation of the unsaturated group to carbonyl chromophores.

The tensile/tear strength retention and elongation at break of the fabrics treated with the four polycarboxylic acid, applied with sodium hypophosphite catalyst, and conventional crosslinking agent, DMDHEU, are shown in Table 1. All crosslinked cotton fabrics show decreased tensile/tear strength and elongation at break. This decrease is caused by the restriction of stress distribution within fibres due to their rigid crosslinking [13].

Tensile strength retention in hypophosphite-catalysed finishing of cotton with BTCA, PAMA, citric acid and MA-IA were 60-69% and tearing strength retention was

54-59% compared with 53% and 50%, respectively, for DMDHEU finishing catalysed by magnesium chloride. Tensile strength retention and tear strength retention percentages in polycarboxylic treated fabrics were higher than in DMDHEU treatment catalysed by magnesium chloride. The advantage of relative strength retention for polycarboxylic acid finishes is attributed to buffering action of sodium hypophosphite catalyst and the absence of Lewis acid catalysts [3].

Low tensile strength in DMDHEU treated fabric may rise from chemical degradation of cellulose by acid-producing catalyst during curing of dried fabrics at high temperature [6].

BTCA-treated fabric showed lower retention of fabric strength than the other polycarboxylic acid treated sample, particularly the citric acid treated one, (see Table 1). The higher number of crosslinks formed by BTCA is likely to be probable cause of the lower retention of fabric tensile strength compared with that of citric acid treated fabrics.

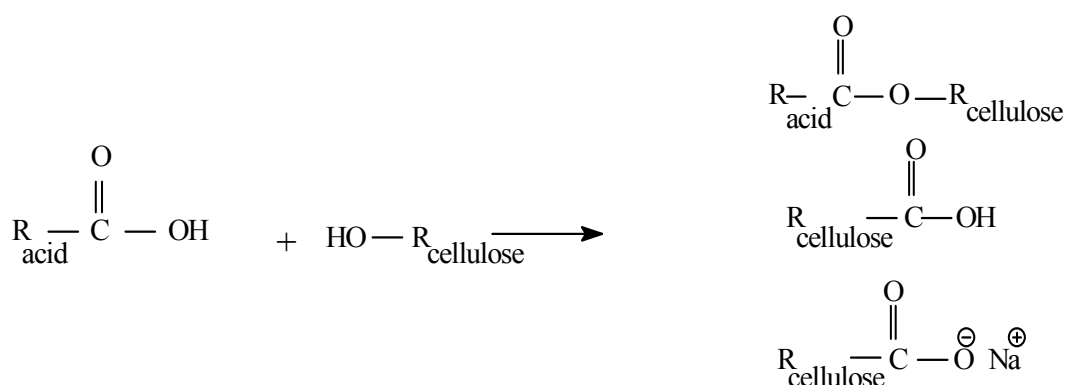
3.2. FT-IR and FT-Raman analysis of BTCA ester crosslinked cotton fabrics

When esterification occurs between polycarboxylic acid and cotton cellulose, the carbonyl in the fabric can exist in three forms: ester linkage, carboxylic acid, or carboxylate anion. The generalized reaction for the formation of the three species is shown in scheme 1 [6,14]:

Table 1: Dry /wet crease recovery angle, whiteness index, tensile/tear strength and elongation at break of crosslinked cotton.

Crosslinking agent	WRA*(dry)	WRA*(wet)	CIE Whiteness Index	Tensile Strength Retention%	Elongation at break (%)	Tear Strength Retention (%)
Untreated	135	129	68	100	9.5±0.1	100
BTCA	252	233	64	60	6.2±0.1	54
DMDHEU	249	230	66	53	5.8±0.2	50
PAMA	232	216	65	65	6.4±0.2	58
Citric acid	216	202	43	69	6.9±0.3	59
MA-IA	230	213	51	62	6.3±0.2	54

*wrinkle recovery angle (in degree)



Scheme1: Reaction cotton cellulose with polycarboxylic acid.

Figure 1 shows ATR infrared spectrum of cotton fabric treated in a solution containing 6.3% BTCA and 6.5% SHP (pH=2), dried at 85°C for 5 min and cured at 180°C /200°C for 90 seconds. The peak at 1725cm⁻¹ in the treated sample represents the total amount of carbonyl ester groups formed between BTCA and cellulose, plus carbonyl groups in the unesterified free carboxylic groups of BTCA. Identifying the ester and acid peaks through conversion of all free carboxylic acid

groups (carbonyl peak at 1725cm⁻¹) to the carboxylate (carbonyl peak at 1585cm⁻¹) by rinsing the fabrics in a base prior to analysis allows quantification of the esterified fraction of finishing [15]. In Figure 2, the carbonyl band at 1730 cm⁻¹ is similarly shown in the FT-Raman spectrum of BTCA finished fabrics though the carboxylate peak at 1585cm⁻¹ is better defined than the FT-IR signal. This may allow better quantification of relative proportions if necessary.

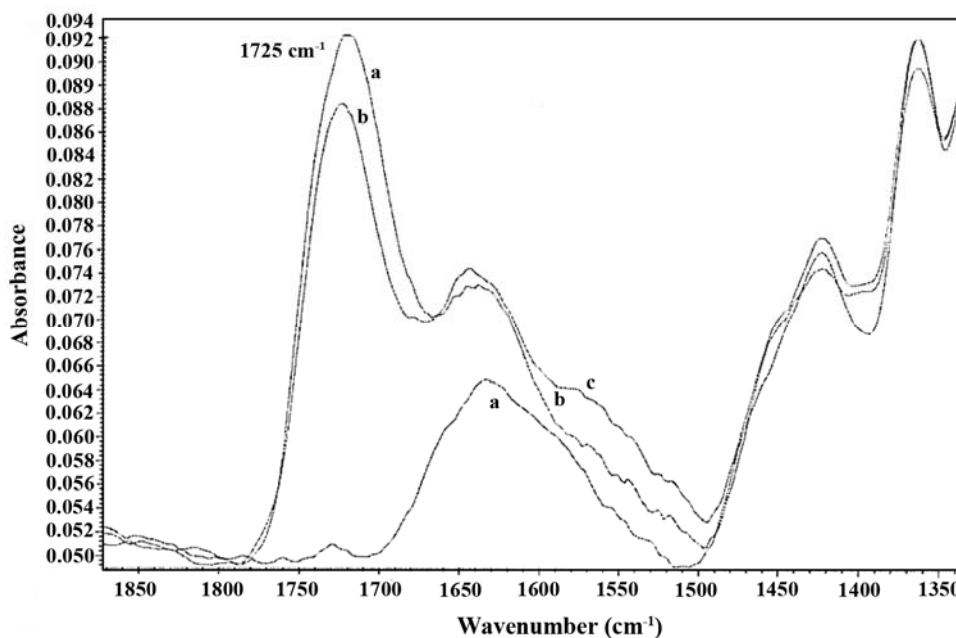


Figure 1: ATR infrared spectra of cotton treated with BTCA and SHP: (a) Untreated, (b)Cured at 180°C, and (c)Cured at 200°C.

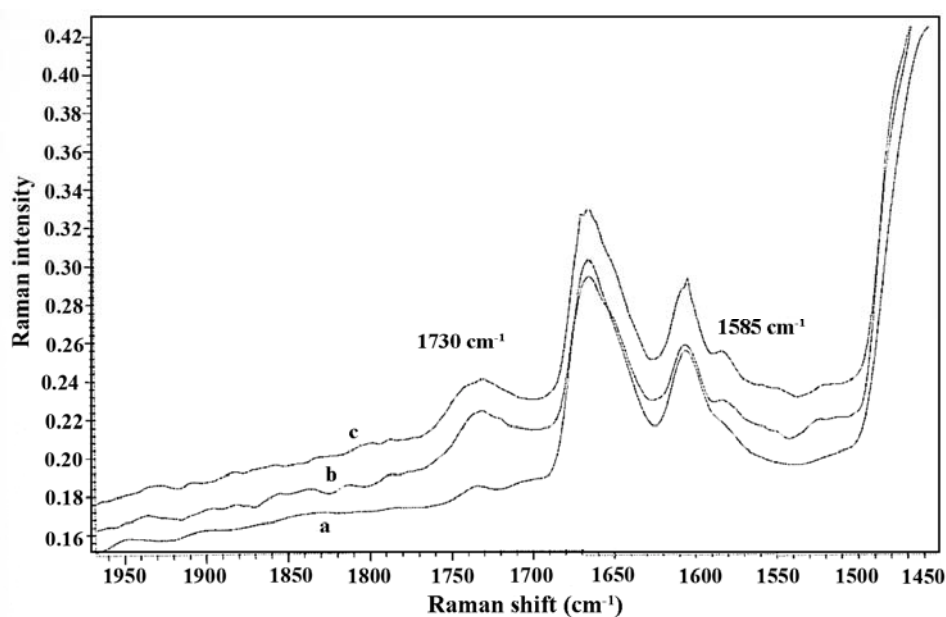


Figure 2: FT-Raman spectra of cotton treated with BTCA and SHP: (a) Untreated, (b) Cured at 180°C, and (c) Cured at 200°C.

3.3. BTCA/PAMA crosslinking treatments on Jarosol sulfur dyed cotton fabrics

3.3.1. Shade changes

In Table 2, ΔE was used to determine the effect of two crosslinking agent (BTCA and PAMA) on shade changes of the three Jarosol sulfur dyed cotton fabrics. Color difference values of the treated fabrics was compared with those of dyed as reference. The treatment with BTCA/PAMA and SHP resulted in color changes of the dyed fabrics; the color change

being Bordeaux > orange > yellow, respectively. These color changes produced by SHP were believed to be due to its reductive nature, which would affect the redox property of the sulfur dyes.

Unrinsed BTCA/PAMA crosslinked cotton fabrics showed less color change compared to rinsed samples (see Table 2). It may be due to the unbound crease resist agent masking the underlying color change to the fabric.

Table 2: Color difference values (ΔE) of Jarosol sulfur dyed cotton fabric treated with BTCA/PAMA.

Sample	Color difference value (ΔE)		
	Bordeaux JB	Orange JG	Yellow JR
Unrinsed -BTCA	3.46	0.77	0.56
Rinsed-BTCA	3.75	1.95	0.94
Unrinsed-PAMA	2.03	2.67	0.45
Rinsed-PAMA	3.71	2.67	0.93

3.3.2. Wash fastness

The results of C06/C1S and C06/C2S wash fastness tests of BTCA/PAMA crosslinked sulfur dyed fabrics shows that the shade changes occur after washing as judged visually and objectively by the color difference values (Table 3 and 4).

The wash fastness rating (C06/C1S) of the three Jarosol sulfur dyed fabric (reference samples) was 3-4 (see Table 3). The wash fastness can be attributed partly to the relatively large molecule and partly to the relatively low aqueous solubility of the dye/cationic fixing agent complex. In general, compared to the reference sample, BTCA/PAMA crosslinked sulfur dyed fabrics exhibited improved wash fastness (less color

change) both with sodium per borate and without sodium per borate (Table 4). BTCA treatment of cotton produces ester crosslinked cellulose fabric; the crosslinking treatment serves to "lock" the dye in fibre, so increasing the wash fastness. However, it is evident that per borate significantly increases the color change in both non-crosslinked and crosslinked samples (see Tables 3 and 4). The bleach sensitivity of sulfur dyes is usually attributed to the cleavage of some of the S-S bonds within the complex heterocyclic macromolecules [16].

Table 5 shows the results of C06/C1S and C06/C2S wash fastness on staining on cotton from crosslinked treated sulfur dyed fabric and untreated sulfur dyed (reference) fabric.

Table 3: Wash fastness of crosslinked Jarosol sulfur dyed cotton fabrics (grey scale).

Sample	Grey scale values					
	Bordeaux JB		Orange JG		Yellow JR	
	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²
Untreated	4	1	3	2	3-4	1
BTCA	3	2	3	3	4	2
PAMA	3-4	2	4-5	3-4	4	2

¹ without sodium per borate, ² with sodium per borate

Table 4: Wash fastness of crosslinked Jarosol sulfur dyed cotton fabrics (ΔE).

Sample	Color difference values(ΔE)					
	Bordeaux JB		Orange JG		Yellow JR	
	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²
Untreated	2.1	6.8	2.1	3.2	1.7	5.9
BTCA	3.5	4.4	1.3	2.2	1.6	4.2
PAMA	3.5	4.7	1.1	1.9	1.4	5.2

¹ without sodium per borate, ² with sodium per borate

Table 5: Cross staining performance crosslinked of Jarosol sulfur dyed cotton fabrics (cotton swatch).

Sample	Grey scale values					
	Bordeaux JB		Orange JG		Yellow JR	
	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²	C06/C1S ¹	C06/C2S ²
Untreated	1-2	1-2	3	3	3	2
BTCA	3	3	3-4	3-4	3-4	3-4
PAMA	3	3	3-4	3-4	3-4	3-4

Table 6: Light fastness of the crosslinked Jarosol sulfur dyed cotton fabrics.

Sample	Blue scale values		
	Bordeaux JB	Orange JG	Yellow JR
Original dyeing	2	1	1
Unrinsed -BTCA	3	2	2
Rinsed-BTCA	2-3	1	1
Unrinsed-PAMA	3	2	2
Rinsed-PAMA	2-3	1	1

There was no staining on polyester, acrylic and wool (grey scale=5). Nylon and acetate has little staining, but most staining occurs on cotton. BTCA/PAMA treatment of Jarosol sulfur dyed cotton improved staining relative to the reference samples. The wash fastness performance (staining) for BTCA and PAMA on sulfur dyed cotton fabric was the same. Staining on untreated and treated sulfur dyed fabrics appears to be independent of sodium per borate. Per borate does not destroy sufficient dye to affect the cross staining.

3.3.3. Light fastness

The light fastness performance of BTCA/PAMA crosslinked sulfur dyed cotton fabrics shows that the reference samples show poor light fastness especially with yellow and orange (Table 6). The low light fastness of the sulfur dyed cotton fabrics may be due to the lack of stability of the dye molecule in the presence of light and cationic fixing agent which accelerates the degradation of the chromophore of the sulfur molecule.

Unrinsed BTCA/PAMA crosslinked fabrics have better light fastness by one grey scale unit than rinsed crosslinked sulfur dyes which exhibit little improvement relative to the reference sample.

4. Conclusions

*BTCA crosslinked cotton showed similar high wrinkle recovery angle and whiteness index to fabric treated with DMDHEU. In addition, polycarboxylic acid as crosslinking agents produced better retention of fabric tensile/tear strength than that obtained by applying DMDHEU system due to lower crosslinking or less damaging application conditions. FTIR/FT-Raman spectroscopy showed formation of ester linkage on BTCA crosslinked cotton fabrics.

*Jarosol sulfur dyed cotton fabric treated with BTCA/PAMA showed better wash fastness (less color change) and staining compared to reference samples (Jarosol sulfur dyed fabric) in either the C06/C1S or C06/C2S fastness tests. This is attributed to the treatment producing ester crosslinked cellulose fabrics. The crosslinking treatment serves to "lock" the dye in the fibre, so increases the wash fastness.

*The wash fastness rating C06/C1S (without sodium per borate) of BTCA/PAMA crosslinked Jarosol sulfur dyed fabrics and reference samples showed less color change than C06/C2S test (with sodium per borate). This is may be related to the cleavage of some S-S bonds of the sulfur dyes in the presence of perborate.

* BTCA/PAMA crosslinked Jarosol sulfur dyed cotton showed poor light fastness, so the effect of UV-absorber on crosslinked cotton fabrics needs to be studied.

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