ELSEVIER

Contents lists available at ScienceDirect

Carbohydrate Polymers

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



Utilization of hydroxypropyl cellulose and poly (acrylic acid) – Hydroxypropyl cellulose composite as thickeners for textile printing

E.S. Abdel-Halim*, H.E. Emam, M.H. El-Rafie

Textile Research Division, National Research Center, El-bohoos Street, Giza, Egypt

ARTICLE INFO

Article history: Received 5 May 2008 Received in revised form 14 May 2008 Accepted 20 May 2008 Available online 28 May 2008

Keywords: Hydroxypropyl cellulose Poly (acrylic acid)-hydroxypropyl cellulose composite Reactive printing and pigment printing

ABSTRACT

Trials were carried out to partially replace kerosene oil with hydroxypropyl cellulose (HPC), poly (acrylic acid)-hydroxypropyl cellulose composite (poly (AA)-HPC) and their mixture in pigment printing paste. Partial replacement was carried out under a variety of conditions. Variables studied include type and concentration of the aqueous thickening agent and type of pigment dyes. In addition to that, poly (AA)-HPC composite was tried to replace sodium alginate in reactive printing. The fastness properties, color strength and stiffness were measured for the reactive and pigment printed samples.

© 2008 Elsevier Ltd. All rights reserved.

1. Introduction

Different processes have been developed for textile printing, depending on the kind of the fabric used (cellulosic, polyester, acrylic, protein...), on the nature of the dyestuff applied (reactive dye, vat dye...) and on the expected quality of the final product. The printing paste mainly contains the colorants and the thickeners (Saffour, Viallier, & Dupuis, 2006). The printing paste undergoes high deformation rates and stresses and, consequently, marked changes in the rheological parameters occur such as viscosity drop and increased elasticity, which all together give the printing paste the capability to penetrate into the fabric (Oblosek, Sostar-Turk, & Lapasin, 2003).

The printing process is a sequence of several operations in order to obtain the penetration and the fixation of the dye molecule in the fabric. During the first step, the printing paste is deposited on the fabric through a rotating screen, and then, it is dried. The second step mainly consists of fixing (padding and steaming) the dye and finishing the fabric (Saffour et al., 2006).

Recently, there has been an increasing research for preparation of thickeners from polysaccharides and application of these thickeners in textile printing (Barba, Montane, Rinaudo, & Farriol, 2002; El-Sherbiny, 2004; Oblosek et al., 2000; Samah, 2004; Sostar & Sehneider, 2000; Zahran, Mohmoud, & El-Rafie, 2007).

Thickeners are used in textile printing to modulate the rheological properties of the printing pastes during application and to obtain sharp and clean drawing patterns by preventing dye

migration. Alginates, guar gum and its derivatives, methyl and carboxymethyl cellulose, some exudates gums and xanthan are excellent thickeners for this application, since they can impart high viscosities at low concentrations and possess the adequate rheological behavior (Baranov, Dymnikova, & Il'in, 2002; Fijan, Sostar-Turk, & Lapasin, 2007).

Alginates are the most widely used thickeners in cellulosic fabrics printing. However, their unstable price and quality as well as their unreliable supply have made other polysaccharides, in particular guar gum and their derivatives quite interesting alternative thickeners (Oblosek et al., 2003).

The pigment printing makes use of kerosene or mineral turpentine which is involved in making emulsion thickeners. In this system, the kerosene in the emulsion gets evaporated to the atmosphere at the time of curing of the pigment printed fabric. It is almost impossible to reclaim this kerosene. In spite of the superior thickening properties of kerosene/water emulsion which also contribute towards a soft hand of the print and good fastness properties, environmental and economical factors have compelled the search for a replacement for kerosene (El-Molla & Schneider, 2006).

Application of pigment is performed through a suitable synthetic binder. Binders are synthetic compounds which have a low degree of polymerization at the application stage. But they polymerize subsequently in the presence of a catalyst at a high temperature to form a transparent film on the fiber surface. Binders play a paramount role in pigment printing, as they are responsible for blooding the non-substantive pigment on the fabric (Zahran et al., 2007).

In the present work HPC, poly (AA)-HPC composite and their co-mixture were used as partial substituents for kerosene oil emulsion in pigment printing pastes. Neutralized poly (AA)-HPC com-

^{*} Corresponding author. Tel.: +20 108113477. E-mail address: essamya@yahoo.com (E.S. Abdel-Halim).

posite was also used for replacement of sodium alginate in printing pastes containing reactive dyes.

2. Experimental

2.1. Materials

Mill scoured, bleached and mercerized plain weave 100% cotton fabric (323 g/m²) and mill scoured, bleached and mercerized plain weave (65/35) cotton/polyester fabric (300 g/m²) were supplied by El-Nasr Co. for Spinning, Weaving and Dying, Mehalla El-Kubra, Egypt. Drimarine Blue P, Printofix Red HB were supplied by Clarient. Ismative Red SHB was supplied by Ismadye. Indofix Golden Yellow 3RN was supplied by Jacofix. Miner Daico Blue SRF was supplied by Miner Diaco. Pigma Color G Yellow KRE was supplied by Pigma Color. Sodium hydroxide, glacial acetic acid, hydrogen peroxide, non-ionic detergent, isopropyl alcohol, propylene oxide, acetone, acrylic acid, thiourea, potassium bromate, potassium bromide, potassium iodide, sulfuric acid, sodium thiosulfate and hydroquinone were all laboratory grade reagents. Sodium alginate, kerosene, sodium carbonate, urea, resist salt, sequestering agent, binder, emulsifier and diammonium phosphate were of technical grade.

2.2. Preparation of hydroxypropyl cellulose

Flax wastes were subjected to alkali treatment followed by bleaching with peracetic acid. The so obtained purified flax waste was used as a starting material for the preparation of HPC according to reported method (El-Rafie, Abdel-Halim, & Emam, 2007).

2.3. Preparation of poly (acrylic acid)-hydroxypropyl cellulose composite

Acrylic acid was grafted onto HPC using (potassium bromate/ thiourea) initiation system to prepare poly (AA)-HPC composite. Details of the grafting reaction are described elsewhere (El-Rafie et al. (2008)).

2.4. Preparation of the printing pastes

2.4.1. Preparation of reactive printing paste

Printing pastes containing reactive dyes were prepared as follows:

Dye	30 g
Kerosene	500 g
Soda ash	25 g
Resist salt (medoxy T)	7 g
Urea	100 g
Sequestering agent (aquafil T)	5 g
Thickening agent	Lg
Water	Χg
Total	1000 g

2.4.2. Preparation of pigment printing paste

Dye	30 g
Binder	100 g
Diammonium phosphate	10 g
Kerosene	Lg
Emulsifier	9 g
Thickening agent	Χg
Total	1000 g

2.5. Printing procedure

Printing was carried out in El-Nasr Company for Spinning, Weaving and Dying, Mehalla El-Kubra, Egypt, using a printing machine (Zimmer Flat Sampling Table). Samples printed with the prepared printing pastes containing reactive dyes were first dried then steamed at 120 °C for 5 min at an atmospheric pressure for the fixation of dye on the fabric. These samples were subsequently washed with cold water followed by hot water. Samples printed with the prepared pigment printing paste were dried at 80 °C then cured at a temperature of 145 °C for 5 min.

2.6. Testing and analysis

2.6.1. Color fastness to washing

The color fastness to washing was determined according to the AATCC test method (AATCC AATCC Technical Manual, 1972).

2.6.2. Color fastness to rubbing

Rubbing tests (dry and wet) were assessed according to standard methods (AATCC AATCC Technical Manual, 1989).

2.6.3. Color strength

Color strength expressed as (K/S) was calculated according to standard method (Judd & Wyszecki, 1975).

2.6.4. Stiffness

Stiffness of printed and unprinted samples was determined according to ASTM test method D 1388-96 (ASTM Method, 1388).

3. Results and discussion

To start with, HPC was prepared from flax processing wastes. The resultant HPC was used as a starting material for the preparation of poly (AA)-HPC composite. HPC and poly (AA)-HPC composite were used as thickening agents in pigment printing paste, and poly (AA)-HPC composite was used as thickening agent in printing paste containing reactive dyes.

Trials have been made to partially replace kerosene oil with HPC, poly (AA)-HPC composite and their co-mixture (aqueous thickening agents). Partial replacement was carried out under a variety of conditions. Variables studied include type and concentration of the aqueous thickening agent and type of pigment dyes applied.

On the other hand, poly (AA)-HPC composite (neutralized with sodium hydroxide) was tried to replace sodium alginate in reactive printing pastes containing different reactive dyes. Given below are the results obtained along with their appropriate discussion.

3.1. Reactive printing

3.1.1. Comparison between poly (AA)-HPC composite and sodium alginate

To confirm the suitability of poly (AA)-HPC composite as a thickener for printing cotton fabrics with reactive dye, comparison was made between fabric samples printed using the composite and traditionally printed samples using sodium alginate which is universally accepted thickener for printing using reactive dyes.

Hence, printing pastes of both sodium alginate and the composite containing Drimarine Blue P dye were prepared and used independently for printing cotton fabrics under similar conditions. The printed samples were assessed for color strength (K/S) and overall fastness properties and the results obtained are summarized in Table 1

It is seen (Table 1) that increasing the amount of poly (AA)-HPC composite incorporated in the printing paste formulation (10–

Table 1Effect of concentration of poly (AA)-HPC composite on the color strength, stiffness and fastness properties of the reactive printing

[Composite]	Color strength (K/S)	Washing fastness	Rubbing fastness		Stiffness
(g/kg paste)			Dry	Wet	(g/cm ²)
Normal	24.27	4-5	5	4	100.13
10	22.24	4-5	5	4-5	106
15	23.45	5	5	4	116.3
20	23.85	5	4	3-4	106.6
25	24.22	4-5	4-5	3-4	111.4
30	24.47	4	4–5	3	119.5

Stiffness of untreated fabric: 94.8 g/cm².

Normal: 12 g alginate/kg paste.

Dye, Drimarine Blue P.

30) g/kg printing paste is accompanied by significant enhancement in the values of K/S (color strength). Equal K/S values were obtained on using 15 g composite/kg reactive printing paste against 12 g sodium alginate/kg reactive printing paste. The rubbing and washing fastness of the composite and alginate printed samples were comparable. On the other hand, the composite printed samples exhibit slightly higher stiffness values than that of the alginate printed samples.

The good results of using poly (AA)-HPC composite as a thickener for printing cotton fabrics with reactive dyes suggest that polymerization of HPC with acrylic acid converts HPC into stable thickener with high ability to transfer the dye from the printing paste to the fabric without reacting with it. The polyacrylic acid whether grafted or homopolymerized associated with the HPC molecules seems to create negative charges on the HPC surface which repel the similar-charge dye ions thereby preventing reaction of the reactive dyes with the hydroxyl groups of HPC.

3.1.2. Type of reactive dye used

Table 2 shows the effect of type of dye used on the color strength, overall fastness properties and stiffness of cotton fabric samples printed using both poly (AA)-HPC composite and sodium alginate as thickeners. It is seen that the values of *K*/*S* obtained with printing paste thickened by sodium alginate is some what slightly higher than those of pastes thickened by poly (AA)-HPC composite. This is observed irrespective of the dye used. It is also seen that printing with sodium alginate and poly (AA)-HPC composite using Drimarine Blue P, Ismative Red SHB and Indofix Golden Yellow 3RN dyes brings about the same fastness properties (rubbing and washing).

Table 2The ability of poly (AA)-HPC composite pastes to print different reactive dyes

Type of dye	Color strength (K/S)	Washing fastness	Rubbing fastness		Stiffness
			Dry	Wet	(g/cm ²)
N1	27.5	5	5	4	98.3
A1	26.2	4-5	4	3-4	105.5
N2	26.4	4-5	5	4-5	99.5
A2	25.95	4-5	4-5	4	106.6
N3	24.27	4-5	5	4	100.13
A3	23.45	5	4	3-4	116.3

Stiffness of untreated fabric: 94.8 g/cm2.

N1: Indofix Golden Yellow 3RN with Alginate.

N2: Ismative Red SHB with Alginate.

N3: Drimarine Blue P with Alginate.

A1: Indofix Golden Yellow 3RN with poly (AA)-HPC composite.

A2: Ismative Red SHB with poly (AA)-HPC composite.

A3: Drimarine Blue P with poly (AA)-HPC composite.

15 g poly (AA)-HPC composite/kg paste.

12 g Alginate/kg paste.

3.2. Pigment printing

To confirm the suitability of the prepared HPC and poly (AA)–HPC composite as aqueous thickening agents to partially replace the traditional kerosene oil emulsion for printing cotton fabrics with pigment colors, a comparison was made between prints obtained using these partially replaced printing pastes with prints obtained via full kerosene oil emulsion.

3.2.1. Comparison between full kerosene oil emulsion and partially substituted printing pastes

Tables 3 and 4 show the effect of the amount of HPC and poly (AA)–HPC composite, respectively, used for partial replacement of kerosene emulsion on color strength, fastness properties and stiffness of pigment prints. It is clear from the data (Table 3) that (a) regardless of the amount of HPC incorporated in the printing paste formulation, the color strength (K/S) is lower than that obtained using full emulsion, (b) the fastness properties are slightly lower than those obtained using the traditional full emulsion and (c) the stiffness properties are significantly higher compared with the traditional method.

It is evident (Table 4) that partial substitution of kerosene oil emulsion with poly (AA)–HPC composite gives acceptable prints compared with HPC but still the color strength and fastness properties are slightly lower than prints obtained using full emulsion.

Table 5 shows the effect of incorporation of mixture of the prepared HPC and poly (AA)–HPC composite as a partial substitution for kerosene emulsion in the printing paste formulation. The data (Table 5) indicates that 50% partial substitution of kerosene emulsion using mixture containing equal amounts (10 g/kg printing

Table 3Effect of concentration of HPC on the color strength, stiffness and fastness properties of the pigment printing

[HPC]	Color strength	Washing fastness	Rubbin	Stiffness	
(K	(K/S)		Dry	Wet	(g/cm ²)
F	14.7	4–5	4-5	4	146.3
Α	10.87	3-4	3-4	3-4	190
В	8.3	3-4	3-4	3	215.8
C	7.2	3-4	4	3-4	231.4
D	9.5	3-4	4	3	238

Stiffness of untreated fabric: 106.5 g/cm².

F: Full emulsion (800 g Kerosene/kg paste).

A: (10 g HPC + 600 g kerosene)/kg paste.

B: (20 g HPC + 400 g kerosene)/kg paste.

C: (30 g HPC + 200 g kerosene)/kg paste.

D: 40 g HPC/kg paste. Dye: Miner Daico Blue SRF.

Table 4Effect of poly (AA)-HPC composite concentration on the color strength, stiffness and fastness properties of the pigment printing

[Composite]	Color strength	Washing fastness	Rubbing fastness		Stiffness (g/cm ²)
(K/S)		Dry	Wet		
F	14.7	4-5	4-5	4	146.3
Α	11.84	4	4	3-4	158.3
В	11.79	3-4	3-4	3	185
C	12.5	4	4	3-4	196
D	10.64	3-4	3-4	3–4	212

Stiffness of untreated fabric: 106.5 g/cm².

F: Full emulsion (800 g Kerosene/kg paste).

A: [10 g poly (AA)-HPC composite + 600 g kerosene]/kg paste.

B: [20 g poly (AA)-HPC composite + 400 g kerosene]/kg paste.

C: [30 g poly (AA)-HPC composite + 200 g kerosene]/kg paste.

D: 40 g poly (AA)-HPC composite/kg paste. Dye: Miner Daico Blue SRF.

Table 5Effect of concentration of mixture of HPC and poly (AA)-HPC composite on the color strength, stiffness and fastness properties of the pigment printing

[Composite]	Color strength	Washing fastness	Rubbin	g fastness	Stiffness
	(K/S)	(K/S)	Dry	Wet	(g/cm ²)
F	14.7	4–5	4-5	4	146.3
Α	15.2	4	3-4	3-4	165.7
В	14.5	4	4	4	159.2
C	13.7	3-4	4	3-4	178.6
D	12.7	3-4	4-5	4	212.5

Stiffness of untreated: 106.5 g/cm².

F: Full emulsion (800 g Kerosene/kg paste).

A: [5 g poly (AA)-HPC composite + 5 g HPC + 600 g kerosene]/kg paste.

B: [10 g poly (AA)-HPC composite + 10 g HPC + 400 g kerosene]/kg paste.

C: [15 g poly (AA)-HPC composite + 15 g HPC + 200 g kerosene]/kg paste.

D: [20 g poly (AA)-HPC composite + 20 g HPC]/kg paste.

Dye: Miner Daico Blue SRF.

Table 6
The ability of HPC and Poly (AA)-HPC composite pastes to print different pigment colors

31	Color strength	Washing fastness	Rubbing fastness		Stiffness
	(K/S)		Dry	Wet	(g/cm ²)
N1 A1	14.99 13.63	4-5 4	4–5 4	3–4 3	148.6 165.2
N2	14.7	4-5	4-5	4	146.3
A2	14.5	4	4	3-4	159.2
N3	16.27	4–5	4–5	3	142
A3	15.85	4	4	3	152.4

Stiffness of untreated: 106.5 g/cm².

N1: Pigma Color G Yellow KRE with kerosene.

N2: Printofix Red HB with kerosene.

N3: Miner Daico Blue SRF with kerosene.

A1: Pigma Color G Yellow KRE with mixture of poly (AA)-HPC composite and HPC.

A2: Printofix Red HB with mixture of poly (AA)-HPC composite and HPC.

A3: Miner Daico Blue SRF with mixture of poly (AA)-HPC composite and HPC.

N: Full emulsion (800 g kerosene/kg paste).

A: [10 g poly (AA)-HPC composite + 10 g HPC + 400 g kerosene]/kg paste.

paste) of HPC and poly (AA)–HPC composite in the pigment printing paste formulation leads to pigment prints with color strength, fastness properties and stiffness comparable with the traditionally full emulsion printed samples.

Taking in mind the environmental impact it could be concluded from the above mentioned results that utilization of mixture from the prepared aqueous thickening agent as a 50% substitution for kerosene emulsion reflects the aim from the study.

3.2.2. Type of pigment color

Table 6 confirms the success of using mixture of the prepared aqueous thickening agent for 50% partial substitution of kerosene oil emulsion to be applied in pigment printing of cellulosic based textiles using different pigment colors.

References

AATCC Technical Manual, Method 36 (1993). 68, 23.

AATCC Technical Manual, Method 8 (1993). 68, 23.

ASTM Method 1388–96. Standard test method for stiffness of fabric, annual book of ASTM standards (2002). 7(01) 320.

Baranov, A. V., Dymnikova, N. S., & Il'in, A. V. (2002). Alkali cellulose as a regulator of the rheological behavior of solutions of dyes. Fiber Chemistry, 34(1), 38–40.

Barba, C., Montane, D., Rinaudo, M., & Farriol, X. (2002). Synthesis and characterization of carboxymethyl celluloses (CMC) from non wood fibers I. Accessibility of cellulose fibers and CMC synthesis. Cellulose, 9, 319–326.

El-Molla, M. M., & Schneider, R. (2006). Development of eco-friendly binders for pigment printing of all types of textile fabrics. *Dyes and Pigments*, 71, 130–137.

El-Rafie, M. H., Abdel-Halim, E. S., & Emam, H. E. (2007). Utilization of industrial flax fiber waste (machine tow) for preparation of hydroxypropyl Cellulose (HPC). Egyptian Journal of Textiles. Polymer Science and Technology, 11(2), 197–216.

El-Rafie, M. H., Abdel-Halim, E. S., & Emam, H. E. (2008). Preparation and characterization of water soluble poly(acrylic acid)-hydroxypropyl cellulose composite. In 5th international conference of textile research division. Cairo, Egypt: NRC.

El-Sherbiny, S. (2004). Effect of polymeric thickeners on pigment coatings: Adsorption, rheological behaviour and surface structures. *Journal of Materials Science*. 39, 4487–4493.

Fijan, R., Sostar-Turk, S., & Lapasin, R. (2007). Rheological study of interaction between non-ionic surfactants and polysaccharide thickeners used in textile printing. *Carbohydrate Polymers*, 68, 708–717.

Judd, B. D., & Wyszecki, G. (1975). Color in business Science and Industry (3rd ed.). Oblosek, M., Sostar, S., Schneider, R., & Lapasin, R. (2000). Interactions between polysaccharides and surfactants and the influence on the Rheological Properties. In Proceedings of the 13th international congress on rheology (Vol. 4) (pp. 154–156). United Kingdom: Cambridge.

Oblosek, M., Sostar-Turk, S., & Lapasin, R. (2003). Rheological studies of concentrated guar gum. *Rheologica Acta*, 42, 491–499.

Saffour, Z., Viallier, P., & Dupuis, D. (2006). Rheology of gel-like materials in textile printing. *Rheologica Acta*, 45, 479–485.

Samah, A. (2004). Printing properties of carbamoylethyl guar and its mixtures with alginate. Egyptian Journal of Textiles. Polymer Science and Technology, 8(1), 105–120.

Sostar, S. T., & Sehneider, R. (2000). Printing properties of high substituted guar gum and its mixture. *Dyes and Pigments*, 47, 269–281.

Zahran, M. K., Mohmoud, R. I., & El-Rafie, M. H. (2007). Utilization of partially methylolated polysaccharide guar gum in pigment printing. RJTA, 11(2), 41–48.