*CELLULOSE CHEMISTRY AND TECHNOLOGY*

# CROSS-LINKING OF COTTON FABRIC USING MALEIC ANHYDRIDE AND SODIUM HYPOPHOSPHITE

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Performance properties of cotton fibers are normally improved by using different cross-linking agents. In the present study, cotton fibers were cross-linked using the combination of maleic anhydride (MA) and sodium hypophosphite ($NaH_2PO_2$). Cellulose fibers were first esterified using MA, and then the cellulose maleate was cross-linked using $NaH_2PO_2$. The effect of cross-linking conditions on wrinkle resistance and whiteness was studied. The data indicated that the H–P–H bonds of $NaH_2PO_2$ react with the >C=C< bonds of two MA molecules already esterified with cotton to form a cross-linkage between two cellulose chains. The cotton fabrics treated with the MA/$NaH_2PO_2$ system demonstrated a high level of wrinkle resistance with significantly lower tensile strength loss, as compared to formaldehyde based commercial cross-linking agents. The whiteness index, bending length and wrinkle recovery ratings were also reported. The cross-linking of cotton (using the MA/$NaH_2PO_2$ system) was demonstrated by FTIR and solid-state CP/MAS $^{13}$C NMR spectroscopic analyses.

***Keywords***: cotton, cross-linking, esterification, maleic anhydride, sodium hypophosphite, wrinkle resistance

## INTRODUCTION

Cross-linking is an important technique to improve the performance properties of cotton based textile materials. The wrinkle resistance of cotton fabrics is usually improved by introducing covalent cross-links between cellulose chains inside cotton fibers.[1,2] The most commonly used cross-linking agents are the traditional formaldehyde based dimethyloldihydroxyethyleneurea (DMDHEU) and its derivatives. However, their hazardous impacts on human health and the environment have limited their use in industry. Extensive efforts have been made for the development of formaldehyde-free cross-linking agents to replace the traditional dimethyloldihydroxyethyleneurea (DMDHEU).[3-6]

Polycarboxylic acids (PCAs), which are non-formaldehyde reactants, have been identified as a potential replacement for conventional formaldehyde containing crease recovery agents.[7,8] Among the PCAs, 1,2,3,4-butanetetracarboxylic acid (BTCA) is used as a cross-linking agent along with inorganic phosphoric alkali salts, e.g., $NaH_2PO_2$.[9-13] Such ester cross-linking of cotton cellulose improves the wrinkle resistance of cotton fabrics.[14-16]

Acids with three or more carboxylic groups are able to cross-link with cotton cellulose, but an increase in molecular size reduces the mobility of an acid to access the hydroxyl groups of cellulose.[17-20] Additionally, PCAs have high cost and cause a drastic loss in tensile strength coupled with yellowing of cured cotton fabrics.[21-23] All the drawbacks of PCAs mentioned above can be minimized using dicarboxylic acid compounds. Here, it is also worth mentioning that the anhydrides of dicarboxylic acids (e.g., MA) are also important due to their high reactivity towards esterification of polysaccharides and their copolymerization reactions.[24]

Herein, we aimed to esterify cotton fabric with MA in the first step, and then to treat the cotton fabric maleate with $NaH_2PO_2$ in order to obtain cross-linked cotton fabric. Further, we aimed to study the properties of the cross-linked cotton fabric, i.e., wrinkle resistance improvement, bending length, whiteness index and wrinkle resistance grading. The results were compared with the values obtained for unmodified cotton fabric and DMDHEU cross-linked cotton fabrics.

## EXPERIMENTAL

### Materials

100% pure bleached cotton fabric of 130 GSM was purchased from a local market. Analytical grade solvents were used for all analyses and reactions. MA (Sigma-Aldrich), sodium hypophosphite (UNI-CHEM), acetone and magnesium chloride (Merck) and commercial grade DMDHEU (Clariant) were used as received.

### Cotton fabric treatment

The scoured and bleached cotton fabrics were cut into specimens of equal size (24 × 12 inch). The cotton fabric specimens were first impregnated in a solution containing MA (6%, w/v) in acetone. The specimens were then passed through a laboratory padder to remove excessive solvent (with two dips and two nips) at an air pressure of 1 bar to reach a wet pick up of approx. 80% on the weight of fabric. The padded specimens were dried at 100 °C for 1 min on a laboratory stenter. After cooling to room temperature, the fabric specimens were treated with three different concentrations of $NaH_2PO_2$ (2, 4 and 8%, w/v) used as a catalyst. The specimens were again subjected to a laboratory padder, the wet pick up being approximately 80%. The treated specimens were dried at 100 °C for 1 min on the laboratory stenter. Finally, the dried specimens were cured at 150, 160 and 170 °C, respectively, for 2 min on the laboratory stenter. The treated fabric properties were measured before and after one home laundering and drying cycle.

### Control sample

For comparison purposes, DMDHEU was applied as a 6% aq. solution containing magnesium chloride as a catalyst instead of $NaH_2PO_2$. The specimens were then passed through a laboratory padder with two dips and two nips to reach an average wet pick up of 80%. The specimens were dried at 100 °C for 1 min and cured at 150 °C for 2 min on the laboratory stenter. The treated fabric properties were measured before and after one home laundering and drying cycle.

### Evaluation of fabric crease recovery angle (CRA)

The CRA was measured according to American Association of Textile Chemists and Colorists (AATCC) Testing Method 66-2008 by a Crease Recovery Tester (WPCTT, Pakistan) available at National Textile University (NTU), Faisalabad, Pakistan. Three samples of each specimen were tested and average values are shown in Table 2.

### Evaluation of fabric stiffness

Fabric stiffness was measured according to American Society for Testing and Materials (ASTM) D1388-08 using a Cantilever Fabric Stiffness Tester available at NTU, Faisalabad, Pakistan. Flexural rigidity or bending stiffness was calculated separately in warp and weft directions by the following equation:

$$G = 1/8 \times W \times C^3 \qquad (1)$$

### Evaluation of fabric whiteness

The whiteness index of untreated and cross-linked fabrics was examined according to AATCC Test Method 110-2011 by a CIE Color-Eye 7000A spectrophotometer (Gretagmacbeth, United Kingdom).

### Evaluation of fabric smoothness

Fabric appearance was checked after induced wrinkling as per AATCC Test Method 128-2009.

### Spectroscopic analysis

Fourier transform infrared (FTIR) spectra (NS 20, range 450-4000 $cm^{-1}$) of untreated and cross-linked cotton fabrics were recorded on an FTIR spectrometer IR Prestige-21 (Shimadzu, Japan). Potassium bromide (KBr) pellet technique was used to collect all infrared spectra. Base line corrections were not employed on spectra. The cotton samples were prepared for recording the FTIR spectra as reported.[25] Cotton fabrics first underwent a home laundering cycle to remove impurities, i.e., unreacted acid and catalyst. Further, the cotton fabrics were impregnated with 0.1M NaOH for 10 min to convert free COOH of MA functions on cotton fabric samples into a salt form. This way, ester carbonyl appears as a separate and easily recognizable signal. These samples were dried at 90 °C and ground to powder in a grinding mill. This fine powder was ground with KBr pellets. The pellets with glassy appearance were dried at 50 °C under vacuum before recording the spectra. The spectra were normalized against the COC band of cellulose (i.e., unmodified cotton fabric sample).

The solid-state CP/MAS $^{13}C$ NMR (100 MHz, NS 5000, acquisition time 0.032 s, delay time 2 s, proton 90°, pulse time 4.85 $\mu$s) spectrum of cross-linked cotton was acquired at ambient temperature using a Bruker DRX-400 machine.

## RESULTS AND DISCUSSION

### Synthesis of maleic anhydride treated cotton fibers and cross-linked samples 1-3

Maleic anhydride (MA) is a bi-functional carboxylic acid anhydride, which forms only one ester linkage with cellulose and is not capable of cross-linking cellulose chains alone. So, by immersing cotton samples and drying at 100 °C, it was esterified *in situ* into cotton maleate due to elevated curing temperatures. However, the anhydride group is very sensitive to hydrolysis and acid may be formed on addition of water in the preparation of the padding bath, therefore, acetone was used instead of water to avoid the hydrolysis of anhydride to acid in the present work. In the next step, $NaH_2PO_2$ was used as a catalyst to cross-link cellulose chains. $NaH_2PO_2$ reacts with >C═C< of the MA already esterified with cellulose, thus making it possible to form cross-linking on cotton cellulose while curing at 150, 160, and 170 °C. In this way, phosphorus bonds in the cross-linked cotton cellulose.[26,27] Reactions and conditions involved are outlined in Fig. 1.

## FTIR Spectroscopic analysis of cotton fibers, MA linked cotton fibers and cross-linked samples

The FTIR spectrum of untreated cotton fabric (Fig. 2a) showed characteristic hydroxyl group (OH) stretching at 3277-3600 $cm^{-1}$. The peak due to the $CH_2$ stretching was observed at 2899 $cm^{-1}$. The $CH_2$ absorption was recorded at 1435 $cm^{-1}$ and $COC_{(AGU)}$ absorption was recorded at 1016 $cm^{-1}$. The FTIR spectrum of cotton fabric treated only with MA (Fig. 2b) displayed a distinct ester carbonyl ($C{=}O_{Ester}$) band at 1722 $cm^{-1}$. Hydroxyl group (OH) absorptions were observed in the region 3230-3439 $cm^{-1}$, which is indicative of incomplete substitution of the hydroxyl groups with anhydride moieties. The FTIR spectra of samples 1, 2 and 3 showed distinct ester ($C{=}O_{Ester}$) peaks at wavenumbers 1712, 1714 and 1718 $cm^{-1}$, respectively, when prepared with different concentrations (2, 4 and 8%) of $NaH_2PO_2$. Overlaid FTIR spectra of samples 1 and 2 are shown in Fig. 2c and 2d, respectively, and the data are summarized in Table 1.

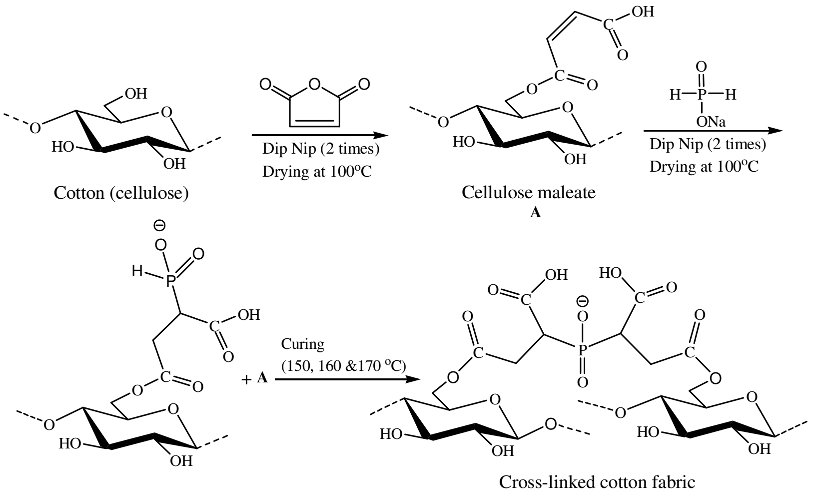

Figure 1: Fabrication of cotton maleate and its cross-linking using $NaH_2PO_2$

Table 1

Characteristic FTIR peaks of different functional groups in the spectra of untreated and cross-linked samples

| Sample name | -OH ($cm^{-1}$) | $-CH_2$ ($cm^{-1}$) | $-CH_2$ ($cm^{-1}$) | $COC_{(AGU)}$ ($cm^{-1}$) | $C{=}O_{(Ester)}$ ($cm^{-1}$) |
|---|---|---|---|---|---|
| Untreated fabric | 3277 | 2899 | 1435 | 1016 | - |
| MA treated fabric | 3230-3439 | 2899 | 1429 | 1039 | 1722 |
| Sample 1 | 3209-3498 | 2897 | 1431 | 1001-1037 | 1712 |
| Sample 2 | 3223-3280 | 2899 | 1427 | 1047 | 1714 |
| Sample 3 | 3238 | 2897 | 1431 | 1001 | 1718 |

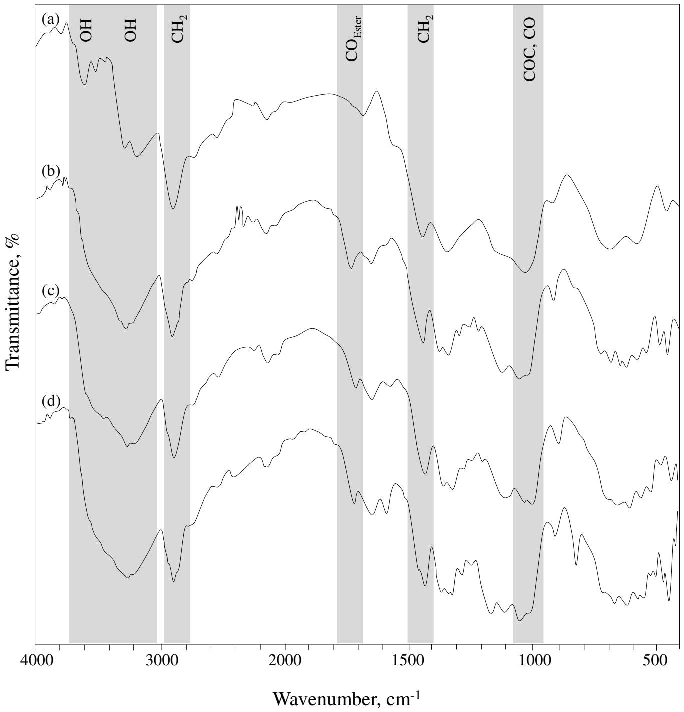

Figure 2: FTIR (KBr) spectra for: a) unmodified cotton, b) MA treated cotton, c) sample 1, and d) sample 2

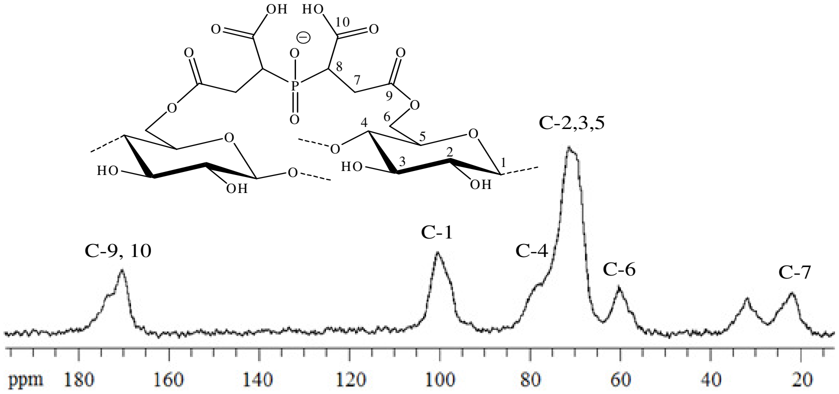

Figure 3: Solid-state CP/MAS $^{13}$C NMR spectrum of cross-linked cotton (sample 3) with 8% $NaH_2PO_2$

## Solid-State CP/MAS $^{13}$C NMR Spectroscopic analysis

The solid-state CP/MAS $^{13}$C NMR spectrum of cross-linked cotton (sample 3) is depicted in Fig. 3. The spectrum shows successful cross-linking as ester signals are observed at about 172 ppm and a carbonyl group of the carboxylic acid is observed at 174 ppm. Both the carbonyls and other signals appear as broad signals, as it is the usual pattern of solid state NMR spectra. The $CH_2$ is observed at about 22, while CH at 32 ppm, indicating that one of the double bonds of MA moieties is cross-linked with phosphorus coming from $NaH_2PO_2$ during the cross-linking reaction. The cellulose backbone signals have also been assigned in the spectrum (see Fig. 3) in a self-explanatory way.

## Crease recovery angle

The crease recovery angles (CRA) of cross-linked cotton samples using MA and $NaH_2PO_2$ were measured. The changes in warp and weft-way CRA of samples 1, 2 and 3 due to changes in curing temperature and concentration of $NaH_2PO_2$ are shown in Fig. 4. It was observed that with the same curing temperature, CRA increased in both directions when the concentration level of $NaH_2PO_2$ was increased, which shows an improvement in the crease recovery behavior of 100% cotton. The increasing trend in CRA was also observed with increasing curing temperature while keeping the $NaH_2PO_2$ concentration constant. It is obvious from Fig. 4 that with a higher level of $NaH_2PO_2$ concentration and higher curing temperature, all the samples show better performance in CRA in both warp and weft directions. Among all samples, the maximum value (98.6°) of CRA was observed in the warp-way specimen of sample 3 cured at 170 °C, compared to the control value (73.6°) and DMDHEU treated control (84.6°). All the results of CRA for the control, cross-linked cotton samples and DMDHEU treated control are gathered in Table 2. In general, warp-way recovery was found to be a little higher than weft-way for all samples. The difference was noted when gradually increasing the curing temperature, while keeping the concentration of $NaH_2PO_2$ constant.

## Bending length

The bending length of samples 1-3 was measured in both warp and weft directions. For each group of specimens, the average value of bending length was calculated. For analyzing the results, all warp-way and weft-way readings were taken thrice and their average is presented in the present work.

The variations in the warp and weft-way bending length of samples 1-3 (Fig. 5) due to the varying concentration of $NaH_2PO_2$ and increasing curing temperature were recorded. It was shown that at the same curing temperature, bending length increased in both directions, when the concentration level of $NaH_2PO_2$ was increased, which shows an improvement in the bending length of cotton fiber. A similar trend was found in bending lengths when cross-linked samples were compared at same concentration of $NaH_2PO_2$, but with increasing curing temperature. It was observed that with a higher level of $NaH_2PO_2$ concentration and higher curing temperature, all cross-linked samples showed improved bending length values in both warp and weft directions. Among all samples, the maximum value (2.33 cm) of bending length was observed in the warp-way specimen of sample 3 when cured at 170 °C, compared to the control value (2.12 cm) and DMDHEU treated control (2.30 cm). A similar comparison was found for the weft-way direction (Table 3) as well.

## Fabric stiffness

Fabric stiffness was also measured using Eq. 1. The warp-way stiffness value is slightly higher than the weft-way value. However, fabrics with low stiffness can bend more easily around small radii than those with high stiffness.

## Whiteness index

All samples were subjected to whiteness measurement to examine a possible yellowing effect of the cross-linking treatment on white cotton fabric. The average value of whiteness for each sample is presented in Table 4. The differences in the whiteness index of cross-linked samples 1-3 due to the changes in the concentration of $NaH_2PO_2$ and curing temperature are shown in Fig. 6.

Table 2
CRA of untreated and cross-linked cotton fabric samples

| Sample name | Concentration of $NaH_2PO_2$ (%) | Curing temperature (°C) | Curing time (min) | CRA (degrees) | |
|---|---|---|---|---|---|
| | | | | Warp-way | Weft-way |
| Control | - | - | - | 73.6 | 73 |
| Sample 1 | 2 | 150 | 2 | 78.3 | 77.3 |
| | | 160 | 2 | 88.3 | 87 |
| | | 170 | 2 | 95.3 | 94.3 |
| Sample 2 | 4 | 150 | 2 | 79.3 | 79 |
| | | 160 | 2 | 90 | 89 |
| | | 170 | 2 | 96.6 | 95.3 |
| Sample 3 | 8 | 150 | 2 | 82 | 80 |
| | | 160 | 2 | 92 | 90.6 |
| | | 170 | 2 | 98.6 | 96.6 |
| DMDHEU treated control | - | 150 | 2 | 84.6 | 81 |

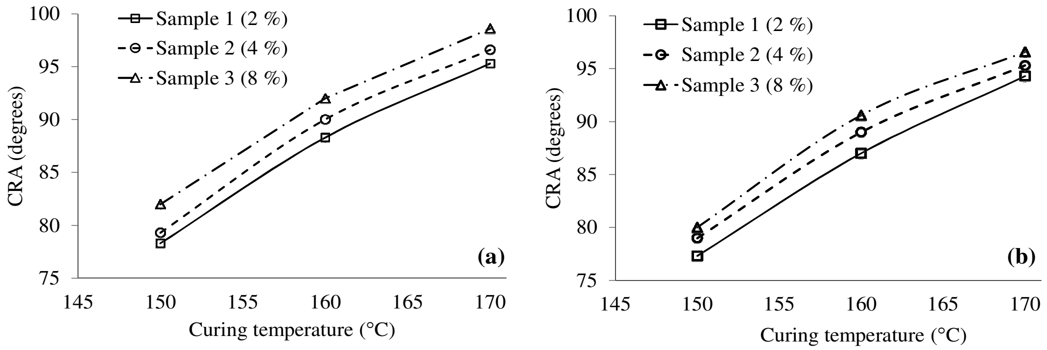

Figure 4: Warp-way (a) and weft-way (b) CRA of samples 1-3 cross-linked with 2, 4 and 8% $NaH_2PO_2$, respectively

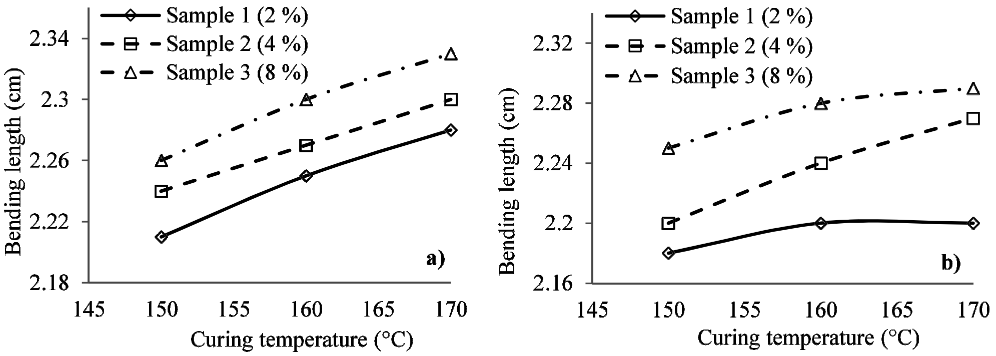

Figure 5: Warp-way (a) and weft-way (b) bending length of samples 1-3 cross-linked with 2, 4 and 8% $NaH_2PO_2$, respectively

Table 3
Bending length of untreated and cross-linked cotton fabric samples

| Sample name | Concentration of $NaH_2PO_2$ (%) | Curing temperature (°C) | Curing time (min) | Bending length (cm) | |
|---|---|---|---|---|---|
| | | | | Warp-way | Weft-way |
| Simple cotton fabric | - | - | - | 2.12 | 2.1 |
| Sample 1 | 2 | 150 | 2 | 2.21 | 2.18 |
| | | 160 | 2 | 2.25 | 2.2 |
| | | 170 | 2 | 2.28 | 2.2 |
| Sample 2 | 4 | 150 | 2 | 2.24 | 2.2 |
| | | 160 | 2 | 2.27 | 2.24 |
| | | 170 | 2 | 2.3 | 2.27 |
| Sample 3 | 8 | 150 | 2 | 2.26 | 2.25 |
| | | 160 | 2 | 2.3 | 2.28 |
| | | 170 | 2 | 2.33 | 2.29 |
| DMDHEU treated control | - | 150 | 2 | 2.3 | 2.29 |

At lower concentration of $NaH_2PO_2$ and at lower curing temperature, cross-linked samples show lower whiteness than untreated fabric, but at higher concentration levels of $NaH_2PO_2$ and at higher curing temperatures, whiteness increased. It was noted that at the same curing temperature, the whiteness increased with an increase in the concentration level of $NaH_2PO_2$. Similar results were also observed when the samples were compared at the same concentration of $NaH_2PO_2$, but with a gradual increase in curing temperature.

The whiteness indices for samples 1-3 and 100% cotton fabric were found comparable at different concentration levels of $NaH_2PO_2$ and curing temperature. Among all the samples, the maximum value (57.72) of the whiteness index was observed for sample 3 cured at 170 °C, compared to the control value (58.71). The results for the whiteness index of samples 1-3, cross-linked with $NaH_2PO_2$ (8%) and cured at 170 °C, are compared with those for the control samples prepared with the standard cross-linking agent DMDHEU (see Table 4). The whiteness index for the DMDHEU treated sample is 59.75, while for the samples treated with MA and cross-linked with $NaH_2PO_2$, its value is 57.72. This shows that cross-linking of cotton cellulose fibers by the esterification reaction of MA has imparted somewhat lower, but comparable whiteness compared to the DMDHEU process.

## Smoothness

The recovery rating of untreated and cross-linked cotton samples is based on the reference standards. For each sample, there were 3 specimens and 3 measurements were made. For reporting the results, the measurements were averaged to the nearest of a rating.

The effects of changing the concentration of $NaH_2PO_2$ on smoothness are shown in Table 5. The results indicate that at constant curing temperature, smoothness was improved with increasing the concentration of $NaH_2PO_2$. The highest improvement was observed for sample 3 when cured at 170 °C. The appearance of samples 1, 2 and 3, cross-linked with $NaH_2PO_2$ (8%) and cured at 170 °C was compared with that of the control samples prepared with the standard cross-linking agent DMDHEU. All the samples showed improved smoothness, however sample 3 showed the highest improvement at 170 °C curing temperature. DMDHEU treated samples showed no change in fabric smoothness, indicating that MA with $NaH_2PO_2$ as a catalyst is more effective for esterifying cotton cellulose than DMDHEU.

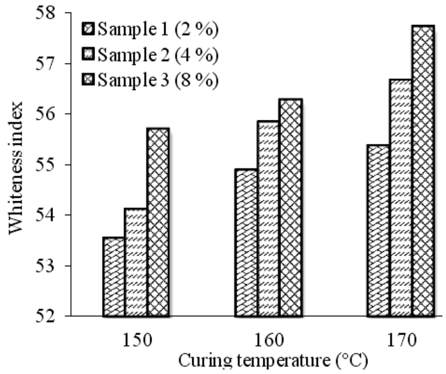

Figure 6: Comparison of whiteness indices for samples 1-3 cross-linked with 2, 4 and 8% $NaH_2PO_2$, respectively

Table 4
Whiteness indices of untreated and cross-linked cotton fabric samples

| Sample name | Concentration of $NaH_2PO_2$ (%) | Curing temperature (°C) | Curing time (min) | Whiteness index |
|---|---|---|---|---|
| Simple cotton fabric | - | - | - | 58.71 |
| Sample 1 | 2 | 150 | 2 | 53.55 |
| | | 160 | 2 | 54.89 |
| | | 170 | 2 | 55.36 |
| Sample 2 | 4 | 150 | 2 | 54.11 |
| | | 160 | 2 | 55.84 |
| | | 170 | 2 | 56.66 |
| Sample 3 | 8 | 150 | 2 | 55.71 |
| | | 160 | 2 | 56.28 |
| | | 170 | 2 | 57.72 |
| DMDHEU treated control | - | 150 | 2 | 59.75 |

Table 5
Smoothness of untreated and cross-linked cotton fabric samples

| Sample name | Concentration of $NaH_2PO_2$ (%) | Curing time (min) | Wrinkle recovery ratings |
|---|---|---|---|
| Simple cotton fabric | - | - | WR-1 |
| Sample 1 | 2 | 2 | WR 1-2 |
| Sample 2 | 4 | 2 | WR 1-2 |
| Sample 3 | 8 | 2 | WR-2 |
| DMDHEU treated control | - | 2 | WR-1 |

## CONCLUSION

The presented new cross-linking strategy (combination of MA and $NaH_2PO_2$) has yielded cross-linked cotton with excellent fiber properties. The method has resulted in cotton fabric with improved wrinkle resistance, stiffness, etc. Using a concentration of $NaH_2PO_2$ of 8% and curing temperature of 170 °C has resulted in better performance of the samples in wrinkle resistance in both warp and weft directions, due to significant cross-linking. In comparison with a commercially available cross-linking agent, i.e., DMDHEU, the combination of MA and $NaH_2PO_2$ showed better results. Therefore, the method has great potential to be exploited at industrial level.

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