

# ORGANOSOLV PULPING OF COTTON LINTER.

## II. EFFECT OF DIOXANE AND ANTHRAQUINONE ON COTTON LINTER PROPERTIES

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*Received June 2, 2011*

Compared to soda pulping, soda-dioxane pulping of cotton linter stabilizes the long-chain cellulose macromolecules against alkaline degradation. The presence of dioxane also results in a more open and accessible fine structure, higher chemical reactivity (upon xanthation), and better viscose filterability.

Soda dioxane anthraquinone (AQ) pulping increases the stabilization of cellulose in cotton and gives pulp with higher accessibility, higher chemical reactivity and better viscose filterability, especially at a higher anthraquinone charge – of 0.1%. Acid prehydrolysis of cotton linter prior to soda dioxane AQ pulping favors the penetration of AQ and dioxane molecules among cellulose chains and breaks the hydrogen bonds, thus providing better accessibility of the hydroxyl groups to the reactant molecules, which leads to a pulp with higher chemical reactivity and better viscose filterability. It is evident that both the soda and the prehydrolyzed soda dioxane AQ pulping methods give cotton linter with better reactivity than that of commercial softwood viscose pulp.

**Keywords:** anthraquinone (AQ), cotton linters, chemical reactivity (xanthation), prehydrolysis soda pulping, prehydrolysis soda dioxane anthraquinone pulping

### INTRODUCTION

Chemical cotton is used in the manufacture of numerous products, from explosives to man-made fibers. Compared to wood cellulose and the cellulose obtained from other sources, chemical cotton is characterized by an exceptionally high degree of purity. For this reason, it has achieved outstanding success in the preparation of derivatives presenting good clarity and colorlessness, as well as good strength and fiber properties, which are essential for the final product quality. However, the ability of cotton cellulose to form viscose is lower than that of technical wood cellulose.<sup>1</sup> Also, the viscose obtained from cotton cellulose has a lower filterability than that prepared from technical wood pulp and pulps from Egyptian reed.<sup>2</sup> Cotton cellulose is characterized by a more compact and less accessible fine structure and, consequently, lower reactivity towards xanthation than bagasse cellulose.<sup>3</sup> Xanthation of bagasse viscose pulp results in higher solubility and lower  $\gamma$ -number for

the dissolved xanthate than in the case of cotton cellulose.<sup>4</sup> Other researchers<sup>5</sup> found out that the degree of crystallinity and the crystallite size were higher in organosolv pulps than in soda and sulphate pulps. At the same time, not only the chemical agents, but also the temperature and pressure in the cooking digester had major effects on the cellulose crystalline structure of the cotton linters. Compared to prehydrolysis soda pulping, prehydrolysis soda ethanol pulping of cotton linters stabilizes the long-chain cellulose macromolecules against alkaline degradation. Prehydrolysis soda ethanol anthraquinone pulping results in further stabilization of cellulose in cotton and gives a more open and accessible fine structure.<sup>6</sup> It has been found out that base-catalyzed dioxane pulping results in an increase in yield,  $\alpha$ -cellulose and DP, and in a decrease in hot-alkali solubility, induced by the formation of hydrogen bonds between the cellulose hydroxyls and the 1,4-dioxane oxygen atoms.<sup>7</sup> The

*Cellulose Chem. Technol.*, **46** (1-2), 137-145 (2012)

microcrystalline cellulose is treated in hot-compressed aprotic solvents, sulfonate and 1,4-dioxane. As a result, in both solvents, more than 90% cellulose is decomposed into a solvent-soluble portion, in which levoglucosane is the main component, with the highest yield of about 35% on the original cellulose basis. The decomposition rate to levoglucosane is, however, faster in sulfonate than in 1,4-dioxane, while levoglucosane is more stable in 1,4-dioxane.<sup>8</sup>

The aim of the present investigation was to obtain a highly reactive cotton linter cellulose with good viscose filterability. In the experiments, dioxane was used in pulping of cotton linter without prehydrolysis, as well as after a prehydrolysis step. The effects of dioxane on the pulp yield, chemical pulp properties and fine structure of cellulose, its chemical reactivity towards xanthation and its viscose filterability were investigated, including the effects of the anthraquinone added. Such a study would provide an insight into a matter that has not been demonstrated before.

## EXPERIMENTAL

### Raw material

The Egyptian cotton linters studied are characterized by longer fiber length and higher amounts of impurities, compared to other linter types. The linters were cut into approximately 10 mm long pieces, placed on a perforated plate with 2 mm in diameter holes, after which the material was mechanically purified by letting compressed air through it, which removed considerable amounts of inorganic matter, broken seeds and hull particles still remaining after its ginning.<sup>9</sup>

### Prehydrolysis, pulping and bleaching

The prehydrolysis, pulping and bleaching operations were carried out as previously described.<sup>10</sup>

### Chemical analysis

The ash and  $\alpha$ -cellulose contents were determined according to the American Tappi standard methods T 211 os-58 and T 203 os-61, respectively.<sup>11</sup>

### Physical properties

The average degree of polymerization (DP) and the degree of whiteness were determined according to earlier applied procedures,<sup>3,11</sup> while hot-alkali solubility was measured according to the Rapson method.<sup>12</sup>

### Fine structure

There are no absolute methods to measure the fine structure of pulp. However, some properties of

cellulose provide acceptable comparative indications on its fine structure. Good examples are the degree of cellulose swelling in water and in a sodium hydroxide solution. The fine structure is also indicated by the ratio between the crystalline and the amorphous cellulose fractions. In this work, the water retention value (WRV) was estimated according to Jayme's procedures.<sup>13</sup> The liquor retention value (LRV) and the sodium hydroxide retention value (NaOH-RV) were determined by allowing the pulp to swell in a sodium hydroxide solution of mercerizing strength at 20 °C, followed by centrifugation to remove the excess alkali. The centrifuged pulp was weighed, washed with distilled water to neutrality, dried to constant weight and weighed again. The washings were titrated against a standard acid. In addition, the degree of crystallinity was determined according to the method of Hessler and Power.<sup>14</sup>

### Reactivity towards xanthation

For estimating its reactivity, the pulp has to be xanthated under conditions that lead to a viscose with a still considerable content of undissolved cellulose, which is taken as indicating the reactivity of cellulose. In this work, the reactivity test was carried out with 0.5 g pulp, 50 mL of 8% sodium hydroxide and 1 mL of carbon disulphide. The dissolved cellulose was determined volumetrically and deduced from the original amount of cellulose, according to the method of Fock.<sup>15</sup>

### Viscose preparation and measurement of filterability

Filterability was determined by the modified method of Centola and Pancetolli.<sup>16</sup> Viscose solutions were obtained from 2 g pulp samples, by emulsion xanthation, using 100 mL of a 10% sodium hydroxide solution and 4 mL carbon disulphide. After xanthation, they were diluted with distilled water until weighing exactly 200 g, and then vigorously shaken.

Filterability was finally measured as the time of filtration, by forcing 25 mL of viscose to pass through a sintered glass funnel (G<sub>1</sub>) through suction. Another 25 mL of viscose was forced to pass through the same funnel and the time of filtration was recorded again. The ratio between the second and the first recorded times provides the filter clogging index (>1), indicating that the filter was partly clogged already during initial filtration.

## RESULTS AND DISCUSSION

### Soda dioxane pulping

In experiment 1 (Table 1), pulping was carried out by the action of sodium hydroxide while, in experiment 2, dioxane was added to the alkaline pulping liquor. It was found out that the presence of dioxane in water (0.25:1 v/v) in soda pulping of cotton linters significantly raised the pulp

yield,  $\alpha$ -cellulose and DP, while it decreased the hot-alkali solubility, which indicates the stabilization of cotton cellulose due to dioxane. The fine structure of cotton cellulose was also favorably affected by dioxane, whereby its affinity to water and alkali and its reactivity improved. It seems that dioxane penetrates in between the cellulose chains and forms hydrogen bonds with them, thus increasing the intermolecular distances. This results in a higher degree of swelling in water and aqueous alkali, higher chemical reactivity and better viscose filterability.

### Soda dioxane anthraquinone pulping

The presence of different amounts of anthraquinone (AQ 0.025, 0.05 and 0.1% based on pulp), together with dioxane, in the soda pulping of cotton linters exerts a more stabilizing effect on the cellulose macromolecules than when using dioxane alone, as indicated by a higher yield and a higher  $\alpha$ -cellulose content, especially at the highest AQ charge: 0.1%. It also results in a

more open and accessible fine structure, as indicated by the increased levels of WRV, NaOH-RV, and by the higher chemical reactivity and better viscose filterability, particularly at high AQ charges (Table 1).

### Effect of prehydrolysis

The effect of prehydrolysis is illustrated by experiments 1, 6 and 11, as shown in Table 2. It is evident that prehydrolysis decreased pulp yield, increased considerably hot-alkali solubility, decreased DP and, in the case of sulfuric-acid prehydrolysis, decreased the degree of whiteness. However, prehydrolysis resulted in higher WRV, LRV and slightly higher NaOH-RV, better xanthation reactivity and a considerably higher viscose filterability. Compared to sulfuric acid, the hydrochloric acid was more effective in depolymerizing cellulose, resulting in lower reactivity to xanthation and lower viscose filterability.

Table 1  
Effects of AQ and dioxane in pulping of cotton linters\*

Experiment no.	1	2	3	4	5
Concentration of NaOH solution, %	1	1	1	1	1
Concentration of dioxane solution, %	-	0.25:1	0.25:1	0.25:1	0.25:1
Concentration of AQ/100 g linters	-	-	0.025	0.05	0.1
Maximum temperature, °C	100	100	100	100	100
Time at maximum temperature, h	2	2	2	2	2
Analysis of pulp					
Yield, %	92.0	93.4	94.6	97.6	98.9
$\alpha$ -cellulose, %	98.2	99.4	98.7	99.2	99.9
Ash, %	0.02	0.02	0.03	0.02	0.01
Hot-alkali solubility, %	2.9	2.0	2.5	2.6	2.1
DP <sup>a</sup>	1590	1802	1705	1579	1523
Degree of whiteness, %	89	90	90	91	91
WRV <sup>b</sup> , %	30.4	38.9	34.6	38.4	39.4
LRV <sup>c</sup> , %	189.0	205.0	205.7	206.0	207.5
NaOH-RV <sup>d</sup> , %	35.9	44.8	37.1	41.0	43.7
Crystallinity, %	96	95	96	95	95
Reactivity (as % insoluble cellulose), %	70.9	37.0	43.5	35.6	32.5
Filterability	3.7:1	2.9:1	2.7:1	1.8:1	1.6:1

\*Liquor ratio in all experiments – 20:1; <sup>a</sup>DP – degree of polymerization; <sup>b</sup>WRV – water retention value; <sup>c</sup>LRV – liquor retention value; <sup>d</sup>NaOH-RV – sodium hydroxide retention value

Table 2  
Effect of acid prehydrolysis on pulp properties

Experiment no.	1	6	11
Acid used in prehydrolysis	-	H <sub>2</sub> SO <sub>4</sub>	HCl
Concentration of acid solution, %	-	0.1	0.1
Maximum temperature, °C	-	100	100

Time at maximum temperature, h	-	4	1
Concentration of NaOH solution, %	1	1	1
Maximum temperature, °C	100	100	100
Time at maximum temperature, h	2	2	1
<b>Analysis of pulp</b>			
Yield, %	92.0	83.9	90.6
$\alpha$ -cellulose, %	98.2	99.1	98.9
Ash, %	0.02	0.04	0.03
Hot-alkali solubility, %	2.9	3.1	7.3
DP	1590	745	660
Degree of whiteness, %	89	85	89
WRV, %	30.4	33.5	31.8
LRV, %	189.0	199.6	190.2
NaOH-RV, %	35.9	36.7	36.1
Crystallinity, %	96	94	96
Reactivity (as % insoluble cellulose ), %	70.9	63.3	68.0
Filterability	3.7:1	1.5:1	2.8:1

### Prehydrolysis soda dioxane pulping

Pulping of cotton linters in the presence of dioxane (dioxane:water – 0.25:1 v/v) initiated by sulfuric acid prehydrolysis had a significant effect on the pulp properties, increasing both the yield and DP, which indicates that dioxane works in two different ways. First, it stabilizes the long cellulose chains and, secondly, it solubilizes the short cellulose chains produced due to acid hydrolysis, as shown by the increased levels of  $\alpha$ -cellulose, DP and hot-alkali solubility. The most

important effect of this pulping liquor was manifested in the fine structure of the pulp, as pulp affinity to water, as well as in the alkali increase, while its crystallinity decreased, and its chemical reactivity towards xanthation and viscose filterability were both improved (experiments 6 and 7, Table 3). The same general effects were observed in the case of a hydrochloric acid prehydrolysis soda dioxane pulping experiment (experiments 11 and 12, Table 4).

Table 3  
Effect of sulfuric acid prehydrolysis on soda AQ dioxane pulp properties

Experiment no.	6	7	8	9	10
Concentration of acid solution, %	0.1	0.1	0.1	0.1	0.1
Maximum temperature, °C	100	100	100	100	100
Time at maximum temperature, h	4	4	4	4	4
Concentration of NaOH solution, %	1	1	1	1	1
Concentration of dioxane solution, %	-	0.25:1	0.25:1	0.25:1	0.25:1
Concentration of AQ/100 g linters	-	-	0.025	0.05	0.1
Maximum temperature, °C	100	100	100	100	100
Time at maximum temperature, h	2	2	2	2	2
<b>Analysis of pulp</b>					
Yield, %	83.9	92.4	92.5	93	93.0
$\alpha$ -cellulose, %	99.1	98.6	99.2	99.1	98.8
Ash, %	0.04	0.03	0.03	0.02	0.03
Hot-alkali solubility, %	3.1	3.6	4.1	3.9	4.6
DP	745	981	980	901	830
Degree of whiteness, %	85	86	91	90	90
WRV, %	33.5	32.4	32.5	35.7	34.4
LRV, %	199.6	206.5	203.7	215.5	208.0
NaOH-RV, %	36.7	38.8	43.0	43.5	42.0
Crystallinity, %	94	93	93	93	92
Reactivity (as % insoluble cellulose ), %	63.3	50.4	49.1	35.0	30.1
Filterability	1.5:1	1.3:1	1.3:1	1.1:1	1.1:1

### Prehydrolysis soda AQ dioxane pulping

The addition of AQ in different concentrations, together with dioxane, in the soda pulping of prehydrolyzed cotton linters stabilized the long chain macromolecules and thus produced pulps with an open and accessible fine structure. This is attributed to acid prehydrolysis, which favors the penetration of the AQ and dioxane molecules in between the cellulose chains. This helps the further separation of the long cellulose chains by breaking the hydrogen bonds, which provides a better accessibility of hydroxyl groups and produces a more open structure, as evidenced by

the lower crystallinity, higher WRV, LRV, NaOH-RV and better reactivity to xanthation, as well as by the improved viscose filterability. Compared to sulfuric acid, the hydrochloric acid had a more intense hydrolyzing effect on the cellulose chains, as shown by the higher values of hot-alkali solubility, lower DP and lower viscose filterability. The best results were obtained at the highest AQ charge (0.1%), applied after sulfuric acid prehydrolysis (Table 3 and Figs. 1-9), and at the lowest AQ charge (0.025%), following hydrochloric acid prehydrolysis (Table 4 and Figs. 10-18).

Table 4  
Effect of hydrochloric acid prehydrolysis on soda AQ dioxane pulp properties

Experiment no.	11	12	13	14	15
Concentration of acid solution, %	0.1	0.1	0.1	0.1	0.1
Maximum temperature, °C	100	100	100	100	100
Time at maximum temperature, h	1	1	1	1	1
Concentration of NaOH solution, %	1	1	1	1	1
Concentration of dioxane solution, %	-	0.25:1	0.25:1	0.25:1	0.25:1
Concentration of AQ/100 g linters	-	-	0.025	0.05	0.1
Maximum temperature, °C	100	100	100	100	100
Time at maximum temperature, h	1	1	1	1	1
Analysis of pulp					
Yield, %	90.6	92.1	93.9	92.5	92.5
$\alpha$ -cellulose, %	98.9	99.3	99.5	98.8	98.9
Ash, %	0.03	0.02	0.03	0.02	0.04
Hot-alkali solubility, %	7.3	5.6	5.0	5.2	4.3
DP	660	720	750	830	825
Degree of whiteness, %	89	87	90	90	90
WRV, %	31.8	33.6	34.4	36.0	31.1
LRV, %	190.2	232.0	241.4	215.7	216.6
NaOH-RV, %	36.1	47.2	45.9	43.1	42.4
Crystallinity, %	96	95	94	94	95
Reactivity (as % insoluble cellulose), %	68.0	35.8	31.0	40.3	45.0
Filterability	2.8:1	1.4:1	1.3:1	2.0:1	2.4:1

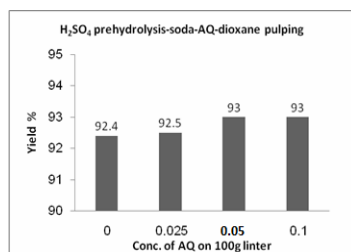


Figure 1: Change in yield, %, with increase in AQ concentration

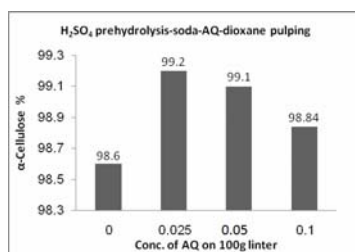


Figure 2: Change in  $\alpha$ -cellulose, %, with increase in AQ concentration

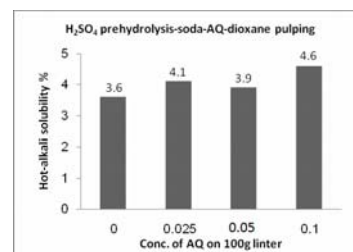


Figure 3: Change in hot-alkali solubility, %, with increase in AQ concentration

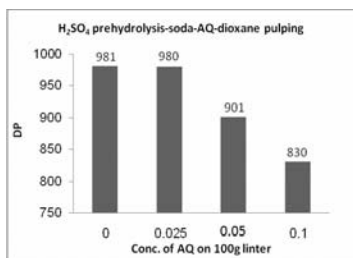


Figure 4: Change in DP with increase in AQ concentration

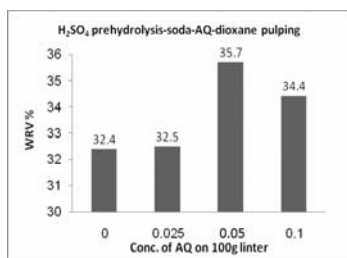


Figure 5: Change in WRV, %, with increase in AQ concentration

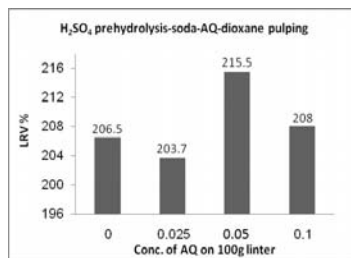


Figure 6: Change in LRV, %, with increase in AQ concentration

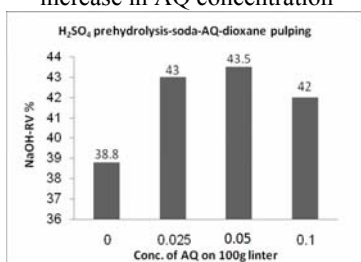


Figure 7: Change in NaOH-RV, %, with increase in AQ concentration

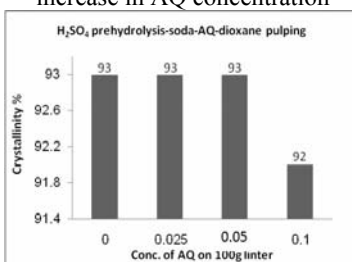


Figure 8: Change in crystallinity, %, with increase in AQ concentration

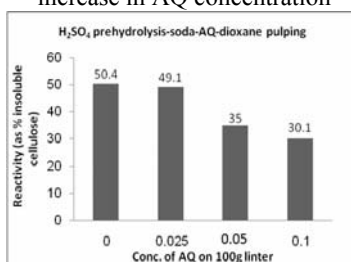


Figure 9: Change in reactivity (as % insoluble cellulose) with increase in AQ concentration

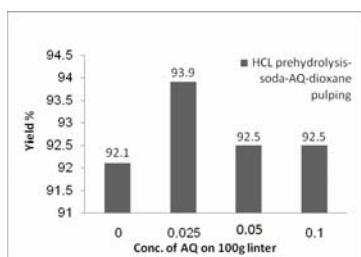


Figure 10: Change in yield, %, with increase in AQ concentration

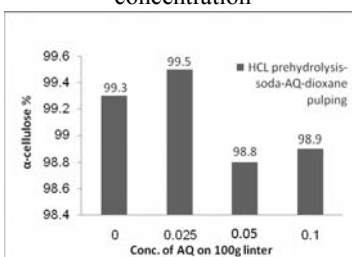


Figure 11: Change in alpha-cellulose, %, with increase in AQ concentration

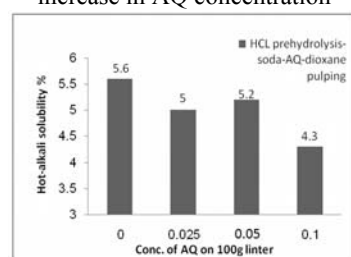


Figure 12: Change in hot-alkali solubility, %, with increase in AQ concentration

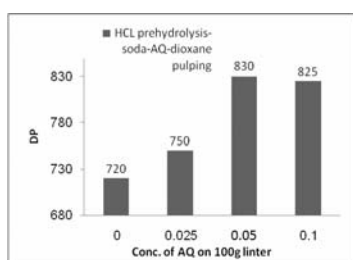


Figure 13: Change in DP with increase in AQ concentration

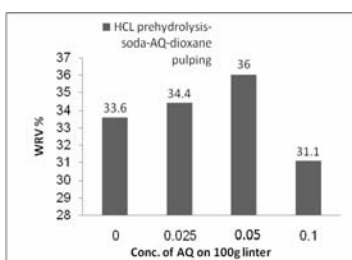


Figure 14: Change in WRV, %, with increase in AQ concentration

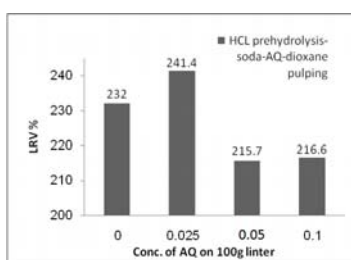


Figure 15: Change in LRV, %, with increase in AQ concentration

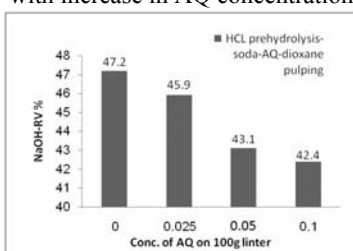


Figure 16: Change in NaOH-RV, %, with increase in AQ concentration

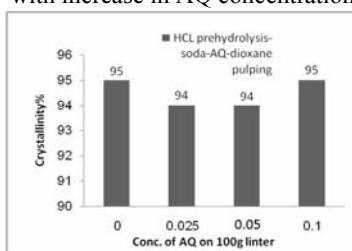


Figure 17: Change in crystallinity, %, with increase in AQ concentration

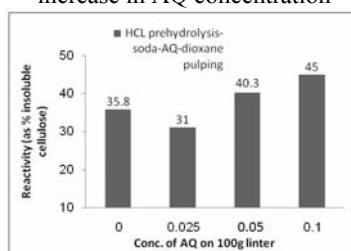


Figure 18: Change in reactivity (as % insoluble cellulose) with increase in AQ concentration

### Comparison of the most reactive cotton linter pulps and commercial softwood pulp

Table 5 and Figures 19-25 list the most reactive pulps obtained by soda AQ dioxane and acid prehydrolysis soda AQ dioxane pulping methods, and a commercial softwood viscose pulp. It is evident that both plain soda and prehydrolyzed soda cotton linter pulps had a much higher  $\alpha$ -cellulose content, higher DP, lower hot-alkali solubility and a much higher crystallinity than the commercial softwood viscose pulp. Their less accessible fine structure

was indicated by lower WRV, LRV and NaOH-RV values. They had a higher reactivity than the commercial pulp, as their viscose filterability was of the same order.

A comparison of the sulfuric acid and hydrochloric acid prehydrolyzed pulps showed that hydrochloric acid prehydrolysis resulted in higher yield and  $\alpha$ -cellulose content, higher hot-alkali solubility, lower DP and higher NaOH-RV, while chemical reactivity and viscose filterability were the same for both acids.

Table 5  
Comparison of the most reactive prehydrolyzed soda AQ dioxane pulp and softwood viscose pulp

Experiment no.	5	10	13	Softwood viscose pulp
Acid used in prehydrolysis	-	H <sub>2</sub> SO <sub>4</sub>	HCl	-
Concentration of acid solution, %	-	0.1	0.1	-
Maximum temperature, °C	-	100	100	-
Time at maximum temperature, h	-	4	1	-
Concentration of NaOH solution, %	1	1	1	-
Concentration of dioxane solution, %	0.25:1	0.25:1	0.25:1	-
Concentration of AQ/100 g linters	0.1	0.1	0.025	-
Maximum temperature, °C	100	100	100	-
Time at maximum temperature, h	2	2	1	-
<b>Analysis of pulp</b>				
Yield, %	98.9	93.0	93.9	-
$\alpha$ -cellulose, %	99.9	99.4	99.5	92.0
Ash, %	0.01	0.03	0.03	0.02
Hot-alkali solubility, %	2.1	4.5	5.0	9.0
DP	1523	930	750	612
Degree of whiteness, %	91	90	90	90
WRV, %	39.4	34.4	34.4	57.9
LRV, %	207.5	208.0	241.4	254.4
NaOH-RV, %	43.7	42.0	45.9	51.9
Crystallinity, %	95	92	94	87
Reactivity (as % insoluble cellulose), %	32.5	30.1	31.0	40.2
Filterability	1.6:1	1.1:1	1.3:1	1.1:1

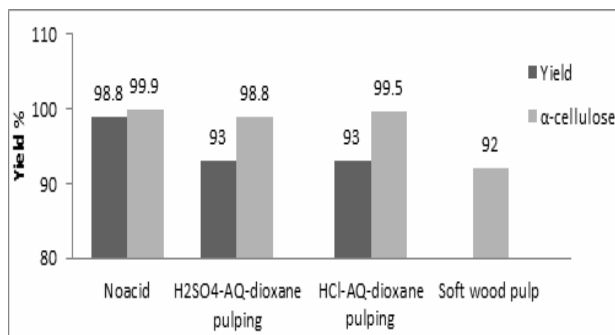


Figure 19: Effect of AQ-dioxane on  $\alpha$ -cellulose, %, and pulp yield, %

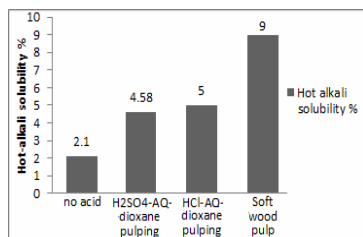


Figure 20: Effect of AQ and dioxane on hot-alkali solubility, %

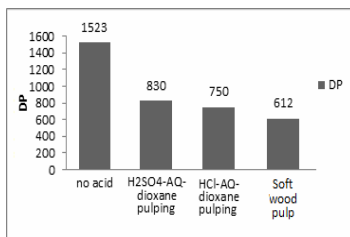


Figure 21: Effect of AQ and dioxane on DP

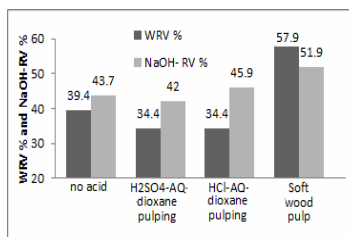


Figure 22: Effect of AQ and dioxane on WRV, %, and NaOH-RV, %

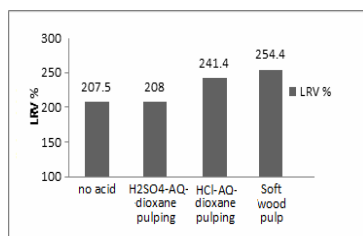


Figure 23: Effect of AQ and dioxane on LRV, %

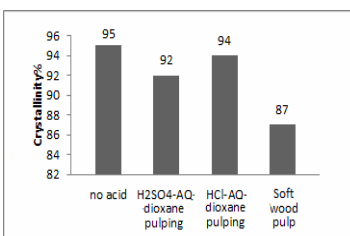


Figure 24: Effect of AQ and dioxane on crystallinity, %

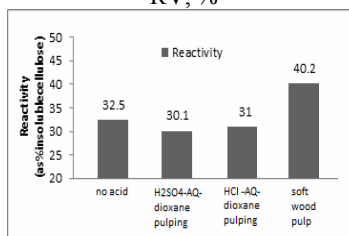


Figure 25: Effect of AQ and dioxane on reactivity (as % insoluble cellulose)

## CONCLUSIONS

Cotton linter was subjected to soda pulping and soda dioxane pulping. It was found out that the presence of dioxane in water in a 0.25:1 (v/v) ratio in soda pulping has a stabilizing effect on the long chain cellulose macromolecules and improves the accessibility of the pulp fine structure and, hence, its reactivity. This is due to the fact that the dioxane molecules penetrated well in between the cellulose chains and thus formed hydrogen bonds with them, subsequently increasing the intermolecular distances between the chains. The extent of hydrogen bonding increased with the addition of different AQ concentrations, together with dioxane, in the pulping liquor, which resulted in a more open and accessible fine structure. Acid prehydrolysis prior to soda AQ dioxane pulping resulted in a better stabilization on the long chain macromolecules and gave higher chemical reactivity and better viscose filterability, especially at higher AQ concentrations. However, the best results were obtained with 0.025% AQ, following HCl prehydrolysis, and with a 0.1% AQ charge, after H<sub>2</sub>SO<sub>4</sub> prehydrolysis. This is attributed to acid prehydrolysis, which favors the penetration of the AQ and dioxane molecules in between cellulose chains, which are more effectively separated, due to breaking of the hydrogen bonds. This provides a better accessibility for the xanthogenation procedures. The comparison of the most reactive pulps obtained by soda AQ dioxane and acid

prehydrolysis soda AQ dioxane pulping methods and the commercial softwood viscose pulp reveals that both the soda and the prehydrolyzed soda cotton linter pulps had a much higher  $\alpha$ -cellulose content, higher DP, lower hot-alkali solubility, a much higher crystallinity and a higher chemical reactivity than the commercial softwood viscose pulp.

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