Carbohydrate Polymers 87 (2012) 1269–1274

Contents lists available at SciVerse ScienceDirect

Carbohydrate Polymers

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

Strength-enhancing effect of cationic starch on mixed recycled and virgin pulps

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ARTICLE INFO

Article history:
Received 8 July 2011
Received in revised form 2 September 2011
Accepted 3 September 2011
Available online 10 September 2011

Keywords:
Cationic starch
Dry-strength additives
Neutral sulfite semi-chemical
Old corrugated container
Tensile index

ABSTRACT

The present work focuses on the effect of cationic starch (Cat St) on the properties of handsheets made from recycled old corrugated container (OCC) and virgin neutral sulfite semi-chemical (NSSC) pulp. This study also evaluates the usage of OCC fibers as a supplement raw material for papermaking. The handsheet samples contained a series of mixtures of OCC and virgin NSSC pulp (0:100, 20:80, 30:70 and 40:60 by weight) and Cat St (0, 0.5, 1.2, 2 and 3 wt.%). The results showed that addition of Cat St could enhance all the strength properties of handsheets remarkably. At certain Cat St dosage, with increasing OCC fibers, the strength properties were improved significantly, which could be related to the Cat St adsorption efficiency. OCC fibers have more specific surface area than NSSC fibers; therefore, they could adsorb much more amounts of Cat St.

1. Introduction

Iran has poor natural forest resources that cover only about 7% of the country's land area. Similar to many developing countries, deforestation and over-harvesting in Iran have created environmental issues. On the other hand, increasing demand for forest resources in various applications has led to the shortages of wood supply. Thus, there is a need to look for innovative ways of using non-traditional forest resources to substitute wood raw materials for pulp and paper industries. The increased demand for wood is likely to be met by one or more of potential supply sources such as: increased utilization of forest waste, increased utilization of waste paper, increased utilization of non-wood fibrous plants, more environmentally sound pulping process alternatives such as bio-pulping and/or, more efficient production of timber through forest/plantation improved management practices (Ashori, Jalaluddin, Wan, Zin, & Mohd Nor, 2006).

Among the possible alternatives, the development of pulp and paper using waste paper is currently at the center of attention (Afra, Resalati, Olson, & Pourtahmasi, 2009; Fatehi, Mcarthur, Xiao, & Yonghao, 2010; Park, Kim, Kim, & Lee, 2008; Wan, Yang, Ma, & Wang, 2011; Zanuttini, Mcdonough, Courchene, & Mocchiutti, 2007). Old corrugated container (OCC) is an excellent alternative waste material to substitute wood because it is plentiful, widespread, and easily available. In addition to its abundance and renewability, utilization of OCC has advantages for economy and environment (Ren, Peng, & Sun, 2009; Si-Yang, Ji-Cheng, Xin-Lu, Hui-Ren, & Yi-Mei, 2010).

The increasing use of recycled fibers and other inexpensive furnish components, such as fillers, has led to a decrease in the strength properties of the paper produced from theses materials. To overcome this problem, it is very common to add different dry-strength additives, such as cationic starch (Cat St), polyvinyl alcohol (PVA), modified polyacrylamide, polyamideamine epichlorohydrine resins (PAE) in combination with carboxymethyl cellulose (CMC) and chitosan (Ashori et al., 2006; Zakrajšek, 2008).

Dry-strength polymers can interact with fibers in a number of ways. In order of increasing bond energy, these interactions are generally termed as van der Waals forces, hydrogen bonding, ionic attractions and covalent bond formation. The majority of paper strength additives function on the basis of the first two energy levels, relying primarily on multiple hydrogen bond formation for their retention and effectiveness. Starch, for example, which has sufficient molecular size to span inter-fiber distances and also has a proclivity for forming hydrogen bonds with cellulosic materials, clearly functions by increasing the number of low energy bonds between fibers. Certain polymeric additives are capable of changing the energy of bonding in paper by supplementing low energy hydrogen bonds (4–6 kcal/mol) with higher energy ionic (10–30 kcal/mol) or covalent (50–100 kcal/mol) inter-fiber linkages (Ashori et al., 2006; Liu, Whiting, Pande, & Roy, 2001).

Native or unmodified starch has limited use in papermaking applications because it comes as viscous and unstable cooked pastes. Starch, therefore, is modified in the paper mill to impart desirable properties (Glittenberg & Becker, 1998). In order to gain good retention and improve the efficiency of starch utilization

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doi:10.1016/j.carbpol.2011.09.010

Table 1
Characteristics of used cationic starch.

Parameter	Cationic starch
pH	5.7
Viscosity, cP	75.7
Solubility, %	100
Ash content, %	1.84
Moisture content, %	9.3
Gel temperature, °C	62–67
Degree of substitution, %	3.5

Table 2
The mixing ratios of raw materials and their abbreviations used in this study.

Treatment		Mass ratios (wt.%)		
Group	Code	OCC	NSSC	Cat St
Z[a]	-	0	100	0
A	1	20	80	0
A	2	20	80	0.5
A	3	20	80	1.25
A	4	20	80	2
A	5	20	80	3
B	1	30	70	0
B	2	30	70	0.5
B	3	30	70	1.25
B	4	30	70	2
B	5	30	70	3
C	1	40	60	0
C	2	40	60	0.5
C	3	40	60	1.25
C	4	40	60	2
C	5	40	60	3

[a] Control sample.

in papermaking fibers, Cat St was developed. In papermaking, Cat St is the most commonly used additive (Pettersson, Höglund, & Waögberg, 2006; Zakrajšek, 2008). It provides many benefits, classified into 4 categories: improvement of mechanical strength; better retention of fines and fillers; faster drainage; and reduction of waste water pollution. The cationic charges on the starch promote strong adsorption onto fibers, fines and fillers through electrostatic attraction to the anionic groups on the surface of the fibers and particles (Fatehi et al., 2010; Pettersson et al., 2006).

A number of studies have reported the effects of dry-strength additives, especially Cat St, on the mechanical properties of virgin pulps. However, no published reports are available that evaluate effects of Cat St on the pulp and paper properties of mixed OCC and virgin neutral sulfite semi-chemical (NSSC). The main objective of this study is to use various mixtures of OCC and NSSC pulps, and consequently various contents of Cat St, and to test selected mechanical properties of the handsheets.

2. Materials and methods

2.1. Materials

The two types of fibrous raw materials used in this study were unbleached OCC and NSSC pulps. OCC fibers were collected from a local recycling company. The NSSC pulp was obtained from Mazandaran Wood & Paper Industries, Iran. The characteristics of NSSC pulp were as follows: freeness 510 mL CSF, consistency 11.1%, total alkaline 1.4, and pH 6.5. Morphological characteristics were measured for OCC and NSSC fibers as follows: fiber length 2.5 and 0.9 mm, fiber width 30 and 28 μm, and cell wall thickness 8 and 11 μm, respectively. Freeness of OCC pulp was measured to be 470 mL CSF. The disintegrated pulp was washed at both high and low pH in order to remove most of the remaining adsorbed metal ions and dissolved and colloidal materials.

Commercial Cat St used in this study was obtained from Iran Wood and Paper Company (Chouka), Iran (Table 1). It was prepared by suspending certain amount of Cat St powder in 100 mL of distilled water and heating the suspension to 80 °C on a water bath with continuous stirring, and then holding the same at this temperature for 25 min after the onset of gelatinization. The solutions were then diluted to 200 mL with distilled water, and refrigerated prior to use. All stock solutions used in the work were always freshly prepared to avoid any possible degradation.

2.2. Handsheet formation

The whole experimental plan is shown in Table 2, where the blending formulations are summarized. Unbleached OCC pulp was mixed with NSSC pulp in ratios of 0:100, 20:80, 30:70 and 40:60 by weight, and the mixes were used for the determination of properties. The weight of Cat St in the formulation was considered with respect to the total weight of handsheet and was deducted from NSSC pulp content. Control sample (Z) contained NSSC pulp without OCC fiber and Cat St. Various solutions of Cat St, in dosages of 0, 0.5, 1.2, 2, and 3% based on oven-dried weight of pulp, were added, to a 1% suspension of pulp. The suspension was then diluted with distilled water to 0.5% and mixed for 5 min. For evaluation of various mixtures, handsheets were made in a British handsheet former using the standard two-stage pressing and drying in rings according to TAPPI T 205 sp-02. For each treatment, 12 handsheets, each having an area of 200 cm^2 and basis weight of 127 g/m^2, were made by taking 2000 mL of pulp slurry at 0.12% consistency.

2.3. Testing methods

Ten handsheets (those closest to 2.45 g oven-dried weight) were chosen and tested for each treatment. The samples were conditioned at $50 \pm 2\%$ relative humidity and 23 ± 1 °C temperature according to TAPPI T 402 sp-98 for at least 4 h, before various tests for each pulp were carried out on them. Conditioned grammage was used for calculations of the strength indices. Handsheets were cut according to TAPPI Procedure T 220 sp-01 for tear, tensile, and burst measurements. The following Tappi test methods were used for sheets analysis: tensile index (T 494 om-01), breaking length (T 494 om-01), tensile energy absorption (TEA, T 494 om-01), stretch at break (T 494 om-01), tear index (T 414 om-04), burst index (T 403 om-02), corrugating medium test (CMT, T 809 om-99), and ring crush test (RCT, T 818 cm-97).

2.4. Statistical analysis

Data for each treatment were statistically studied by analysis of variance (ANOVA). When the ANOVA indicated a significant difference among factors and levels, a comparison of the means was done employing Duncan's multiple range test (DMRT) to identify the groups that were significantly different from other groups at 95% and 99% confidence levels.

3. Results and discussion

3.1. Tensile index and breaking length

Tensile strength is one of the basic strength properties tested on pulp and paper. According to the ANOVA tests, both variable factors exerted significant influence on the tensile index and breaking length as a single factor (Table 3). In addition, the interaction between the two variables was significant with the 95% and 99% confidence levels based on the ANOVA analysis. The results of DMRT

Table 3
Results of ANOVA test on the effect of variables on handsheets properties.

Source of variations	df	Tensile index			Breaking length			TEA			Stretch		
		SS	MS	F	SS	MS	F	SS	MS	F	SS	MS	F
A	3	0.0553	0.0184	3.09*	0.1356	0.0553	3.09*	48863.81	16287.93	288.22**	0.44	0.14	30.56**
B	4	232.3066	58.086	20.66**	0.8361	0.0184	54.23**	139680.02	34920.00	617.93**	2.38	0.59	125.47**
A × B	19	1.07643	64.07	54.98**	0.9817	0.07592	199.06**	17575.63	2196.95	38.88**	0.24	0.02	6.16**
Error	9	0.0417	25.2946		0.0417	0.0059		1695.34	56.51		0.14	0.004	
Total	35	257.6012			0.0970			222526.85			3.54		

Note: df, degree of freedom; MS, mean of squares; SS, sum of squares; *F*, *F* value; ns, not significant.
* Significant difference at the 5% level ($p \le 0.05$).
** Significant difference at the 1% level ($p \le 0.01$).

are shown by letters in Table 4. The lowest tensile index value was 12.2 N m/g for the control sample, while the highest value was found as 29.2 N m/g for the C_5. Similar trends were also observed for breaking length properties (Fig. 1a).

Tensile strength for treated samples is generally higher than those for untreated ones. As it can be seen from Fig. 1a, the C_5 pulp treated with 3% Cat St had the highest tensile index value among the pulps evaluated in this investigation. The tensile index of the pulp is independent of chemical properties and is very much dependent on the bonding ability of the fibers in the network (Ashori, 2006). As Pande, Rao, Kapoor, and Roy (1999) reported, a decrease in freeness contributes toward improving the fiber bonding within the fibrous network of the sheet and improving the tensile index. However, papers made from short fibered pulps are generally stiffer in extension than those made from long fibers.

Statistical analysis indicates that the differences in the mean values of breaking length within and among compared groups are significant at the 99% confidence level. The C_5 and C_4 pulps have 29 and 25 km breaking length, respectively, which are the highest values among the pulps examined in this study (Fig. 1a).

Breaking length is defined as the length of paper which would just break under its own weight. It is a measure of the resistance of paper to direct tension under specific conditions of rate of extension. It has been well known that inter-fiber bonding and fiber strength have the strongest influences on breaking length. Thin cell walls (collapsibility) and long fibers (number of bonds per fiber) improve breaking length (Wan et al., 2011). As mentioned in Section 2.1, OCC fiber is about 3 times longer than NSSC fiber. Its cell wall thickness is thinner than NSSC as well.

3.2. Stretch and TEA

Stretch (elongation) is defined as the ability of the fiber network to elongate under load without breaking and is a good measure of the toughness of paper. It is determined by fiber elongation, and the distortion of the fibrous network. It is measured as part of the tensile strength test, being defined as the strain of the paper at breaking point.

Use of DMRT indicates that the differences in the mean values of stretch within and among the compared groups are significant at the 95% confidence level. The same trend was observed with the TEA. The pulp type C, treated with 3 wt.% Cat St, has the highest stretch (1.7%) among the pulps studied in this work, although the variation in stretch is small in the range of 0.8–1.7% (Fig. 1b). It is apparent that a decrease in freeness causes better bonding of the fibrous network, which improves the stretch. Pande et al. (1999) reported that stretch is influenced by the morphological properties, and chemical properties do not significantly contribute toward the development of stretch.

Tensile energy absorption, commonly termed as TEA (and also known as "work to rupture"), is a very important measure of the strength of paper. Use of statistical analysis indicates that the differences in the mean values of TEA within and among compared groups are significant at the 99% confidence level. It is clear that TEA for core fibers depends on the strength of the individual fibers as well as the strength and stretch of the fibrous network. A decrease in freeness or an increase in fines content means a well-bonded network of fibers, which increases the TEA. For the OCC fibers, TEA depends more on the fiber bonding of the network than the fiber strength. Properties such as freeness, coarseness, and fines content contribute to the bonding potential of the fibrous network.

At a given Cat St charge, TEA and stretch for the mixed pulps with 40 wt.% content OCC (group C) were higher than those for the lower OCC (20 and 30 wt.%).

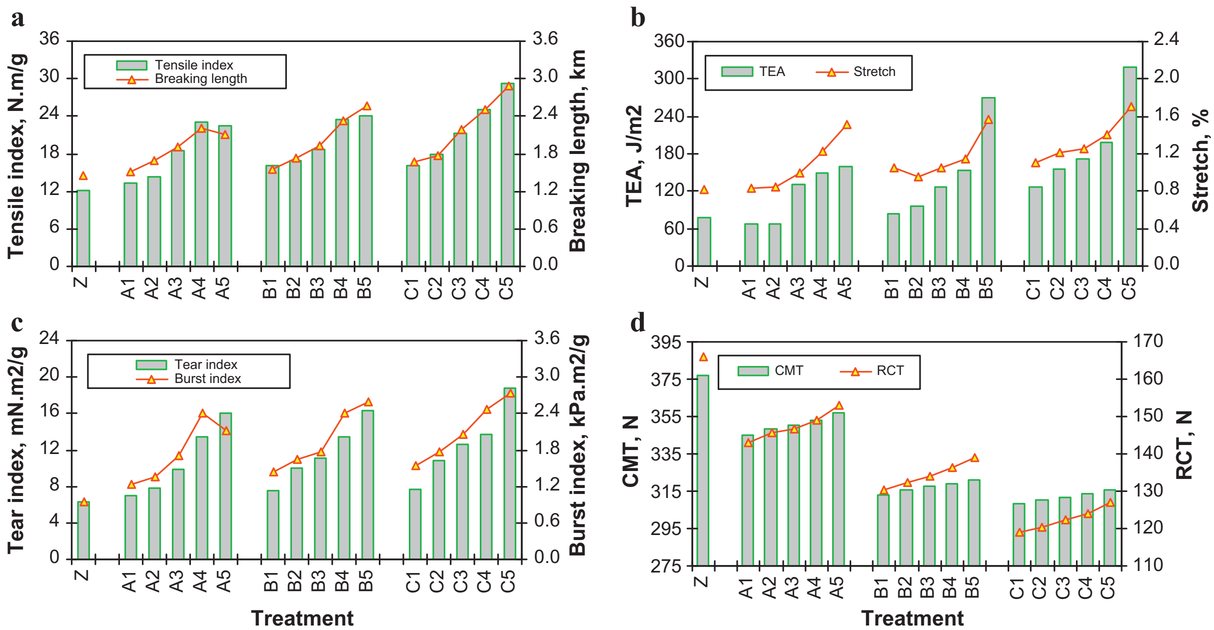

Fig. 1. Effects of various treatments on the properties of OCC/NSSC pulps.

3.3. *Tear and burst indices*

Analysis of variance showed that there was statistically significant (Table 5) difference between the control sample and untreated samples relating to content of OCC. Therefore, it can be concluded that the effect of OCC contents was a cause for the improvement of tear and burst indices. Use of DMRT indicates that the differences in the mean values of tear index within and among the compared groups are significant at the 99% confidence level (Table 4).

Table 4
DMRT of OCC pulp and Cat St on the mechanical properties.

Treatment	Property							
	Tensile index (N m/g)	Breaking length (km)	TEA (J/m^2)	Stretch (%)	Burst index (kPa m^2/g)	Tear index (mN m^2/g)	CTM (N)	RCT (N)
Z	12.2 (a)	1.46 (b)	78.5 (d)	0.82 (c)	0.94 (d)	6.2 (c)	377 (a)	166 (a)
A_1	13.3 (c)	1.51 (d)	66.5 (c)	0.83 (c)	1.23 (d)	7.1 (d)	345 (e)	143 (d)
A_2	14.3 (cb)	1.70 (c)	67.9 (c)	0.84 (b)	1.35 (d)	7.8 (d)	348 (d)	146 (c)
A_3	18.4 (ab)	1.90 (b)	130.2 (b)	1.00 (b)	1.71 (c)	9.9 (c)	350 (c)	147 (c)
A_4	23.1 (a)	2.20 (a)	150.2 (a)	1.23 (a)	2.40 (a)	13.4 (b)	353 (b)	149 (b)
A_5	22.4 (a)	2.11 (a)	160.5 (a)	1.51 (a)	2.12 (b)	16.0 (a)	357 (a)	153 (a)
B_1	16.2 (b)	1.55 (d)	84.1 (e)	1.05 (d)	1.44 (c)	7.5 (d)	313 (d)	130 (e)
B_2	17.0 (b)	1.73 (cd)	95.9 (e)	0.96 (c)	1.64 (b)	10.0 (c)	316 (c)	132 (d)
B_3	18.7 (b)	1.93 (c)	126.5 (c)	1.05 (b)	1.77 (b)	11.1 (c)	318 (b)	134 (c)
B_4	23.4 (a)	2.32 (b)	152.9 (b)	1.14 (a)	2.40 (a)	13.5 (b)	319 (b)	136 (b)
B_5	24.1 (a)	2.56 (a)	269.6 (a)	1.57 (d)	2.60 (a)	16.3 (a)	321 (a)	139 (a)
C_1	16.2 (c)	1.68 (c)	126.4 (c)	1.10 (c)	1.54 (c)	7.7 (d)	308 (e)	119 (c)
C_2	17.9 (c)	1.77 (c)	155.8 (d)	1.21 (d)	1.77 (c)	10.8 (b)	310 (d)	120 (c)
C_3	21.3 (cb)	2.19 (cb)	172.6 (b)	1.25 (d)	2.05 (b)	12.6 (c)	312 (c)	122 (b)
C_4	25.0 (ab)	2.50 (ab)	199.2 (b)	1.40 (d)	2.47 (a)	13.7 (b)	313 (b)	124 (b)
C_5	29.2 (a)	2.88 (a)	319.6 (a)	1.70 (a)	2.74 (a)	18.8 (a)	316 (a)	127 (a)

Note: The same letter denotes that the difference in the mean values of properties within and among the compared groups is not statistically significant.

Table 5
Results of ANOVA test on the effect of variables on handsheets properties.

Source of variations	df	Tear			Burst			CTM			RCT		
		SS	MS	*F*	SS	MS	*F*	SS	MS	*F*	SS	MS	*F*
A	3	3.0514	1.0171	142.30**	0.5001	0.1667333	72.34**	7377.8374	2459.2791	6799.85**	2948.5622	982.854	1780.07**
B	4	155.9712	3899.28	126.09**	2.6539	0.6634	146.21**	224.7042	56.176	137.95**	144.9985	36.2496	54.88*
A × B	19	265.8706	4321.0904	874.33*	4.6634	1.0973	543.69*	8954.0032	6541.0932	9004.32*	4678.3671	2958.0482	3045.85*
Error	9	0.05	0.0071		0.0161	0.0023		2.5316	0.3616		3.865	0.5521	
Total	35	3.1014			0.5163			7380.369			8796.84	4100.053	

Note: df, degree of freedom; MS, mean of squares; SS, sum of squares; *F*, *F* value; ns, not significant.
* Significant difference at the 5% level ($p \leq 0.05\%$).
** Significant difference at the 1% level ($p \leq 0.01\%$).

The length of fiber influences the tear strength. Longer fibers have higher tear strengths, since longer fibers naturally provide more points of bonding and are pulled a longer average distance from the network (Ashori, 2006). As shown in Fig. 1c, OCC pulps exhibit a distinct difference from control sample. The long (2.5 mm) fibered OCC pulp has very good tearing strength, whereas the NSSC pulp is relatively low in tear because of its short fiber (0.9 mm) length. The pulp type C_5, made using 3% Cat St, has the strongest tear index (18.7 mN m^2/g) among the pulps studied in this work. The burst values of the samples showed the similar tendency to the results of the tear index.

According to Van den Akker's strength theory, tearing work is composed of two different phenomena: the work of breaking the fibers and the frictional work pulling them out of the undamaged network (Park et al., 2008). Tearing strength is strongly influenced (positive correlation) by fiber strength, probably because the long fibers are very pliable and are entangled at many points, and so are able to distribute stress over a wide area. The rigidity of the fibers is determined by the positive relationship with the Runkel ratio ($2w/l$) or cell wall thickness. In OCC and NSSC fibers there is a large difference in tearing strength. The tearing strength of treated OCC/NSSC pulps is about 2–3 times higher than that of the untreated ones at a given breaking length. This is probably due to the morphological (principally fiber length) differences in the two fibrous materials. As mentioned earlier, the OCC fibers' length and Runkel ratio are 3 and 4 times more than NSSC fibers, respectively.

Use of DMRT indicates that the differences in the mean values of burst index within and among compared groups are significant at the 95% and 99% confidence levels. The pulp types C_5 and B_5 had 2.7 and 2.6 kPa m^2/g burst index, respectively, which was the highest among the pulps measured.

At a certain dosage of Cat St, with increasing OCC fiber the strength properties were improved remarkably. This could be explained by the Cat St adsorption efficiency, which depends on the specific surface of particles. Zakrajšek (2008) reported that in the case of pulp fibers, its values range from 0.2 to 5 m^2/g, depending on refining. His research has shown that 5–30 times greater amount of Cat St can be adsorbed onto fine stock and refined (beaten) fibers, which has much more specific surface than unbeaten fibers. During the refining process, fibers are fibrillated, and in some cases shortened as well. Consequently, the amount of fines, the total specific surface area and the adsorption of Cat St are all increased as well (Mendes, Costa, Silvy, & Belgacem, 2001; Mendes, Sansana, Silvy, Costa, & Belgacem, 2001). Unlike NSSC pulp, OCC pulp was refined and its freeness was less (470 CSF) than NSSC value (510 CSF).

3.4. CMT and RCT

Rigidity of the fluted structure is one of the essential characteristics of corrugated board and flat crush resistance is necessary to prevent crushing of the flute structure on the corrugators and other converting equipment. The CMT permits the evaluation of corrugating medium before it is fabricated into combined board.

The edgewise comparison strength of corrugated board is a principal element in determining the dynamic comparison strength of the container made from a board. Since fiberboard shipping containers are frequently subjected to loads which are resisted by compression strength, this property is an important measure of the performance characteristic of corrugated board, useful in measuring the quality of the finished product.

Statistical analysis showed that variable factors and their interactions had significant effect on the CMT and RCT (Tables 4 and 5). With the addition of OCC fibers in the furnish, both properties decreased significantly (Fig. 1d). From morphological point, this feature could be attributed to the fact that the recycled fibers are more damaged (due to the refining) than the virgin pulp, and their resistance to the CMT and RCT is already lower than the NSSC pulp. This behavior is probably also due to the thin cell wall of OCC fibers.

4. Conclusions

The main conclusions drawn from this study are as follows:

(a) The results clearly showed that the addition of Cat St to a sheet formed from OCC/NSSC fibers led to excellent improvement in strength properties compared to the control sample and untreated ones.
(b) From technical point, OCC fibers in comparison to virgin NSSC pulp, generally have improved conformability and inter-fiber bonding capability due to fiber morphology of these fibers.
(c) With the exception of CTM and RCT, the other properties were increased by addition of OCC fibers. However, Cat St could enhance all the studied properties. The negative influence of OCC on the CTM and RCT can be explained by the weaker (thinner) cell wall thickness of OCC compared to the NSSC fibers.
(d) Analysis of data revealed that the two variable factors (namely OCC and Cat St contents) and their interactions had significant effect on the strength properties. The use of DMRT in this work indicated that the differences between the mean values of the studied properties within and among each of the groups compared were highly significant.
(e) The pulp type C_5 had the most desirable properties in term of tensile, tear and burst indices and breaking length.
(f) Finally, it can be stated that OCC pulp has good potential as a supplement fibrous material, in combination with virgin pulp.

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