

Improvement in strength properties of packaging paperboard using biopolymer chitosan following a green approach



Shubhang Bhardwaj



Nishi K. Bhardwaj



Yuvraj Singh Negi

Abstract

The share of paperboard in packaging industry has increased in recent years due to urbanization, higher living standards and changes in consumption and distributions patterns. Corrugated boxes are mostly used as tertiary packaging material, most of which are recycled for various purposes and have a major portion of recycled fibres. The recycling process reduces strength properties of old corrugated containers (OCC) pulp and to overcome a variety of approaches including blending with virgin softwood (SW) or other pulp and addition of dry strength additives and synthetic polymers are practiced. These synthetic polymers being carcinogenic may pose health hazards and are not environmentally safe. The technical advancements for using the biodegradable and health friendly biopolymers are required to make the packaging industry greener. In present study the increase in strength properties of recycled OCC (different types) and SW pulps was done with the use of biopolymer as strength additive. The burst factor and RCT value increased after addition of 5 kg/t chitosan in OCC-1, OCC-3, OCC-3 (refined) and SW pulp by 47.6% and 42.9%, 30.9% and 38.0%, 16.4% and 45.5%, and 19.5% and 11.8%, respectively. The tear factor, burst factor, CMT, and RCT value increased after addition of 1 kg/t of chitosan in OCC-2 pulp by 11.6%, 30.0%, 30.4%, and 39.7%, respectively. The tear factor, Taber stiffness, wax pick, tensile index, etc. were also improved.

Keywords: Chitosan, Old corrugated containers, Strength properties, Softwood and Papermaking

1. Introduction

Paperboard is one of the end products of packaging industry that has a significant niche in the commodities of routine requirement. Indian corrugated box industry is a rapidly growing industry due to increasing demand of corrugated packaging for finished goods transportation and handling. Paperboard packaging has been considered as a sustainable approach since it involves production from a renewable resource and its easy recyclability makes it the preferred choice of the consumers. As the world is facing the devastating effects caused by plastic over-use, paperboard packaging options mainly corrugated and folding cartons are now being considered as the most environmental friendly packaging option. Consumers have become more aware of the environmental and health benefits of green packaging practices. The market for green packaging is growing at a CAGR (compound annual growth rate) of 7% and expected to reach \$242.50 billion in 2021 [1]. Over 50% of all paperboard packaging is utilized in food product sectors including dairy products, beverages, frozen foods, candies, dry foods, etc. Its complete biodegradable

nature and sustainable production is therefore required for the environment as well as the industry. Approximately, 20% of total paper and paperboard production is used for packaging purpose [2].

The package material for packaging depends on the nature/sort of material/article to be stuffed within it, e.g. for food stuffs packaging, hindrance properties are highly essential and for solid materials, the strength properties must be taken into consideration. Most basic biodegradable packaging materials are cellulosic fibrous forms like paper, paperboard and layered boxes [3-5]. In paperboard, the cellulosic filaments are held together by hydrogen bonds and the presence of hydrogen bonds between the cellulose strands oversees the quality of paperboard [6-9]. To improve the physical properties, paper and paperboard making process must include the expansion of various quality added substances called strength additives.

A quality added substance for paper and paperboard at wet-end ought to be dissolvable in water, substantive to the cellulosic surface and it ought to contain functional group(s) for ionic

or covalent holding with the cellulosic filaments [7,9-10]. Paper and paperboard is assumed to be integral part of the packaging industry. Packaging is a mix of craftsmanship and science to secure and upgrade the timeframe of palatable or non eatable items which may include anything like solid or fluid or gas. A paper utilized for packaging reason must have high elasticity, burst quality, tear quality, firmness and hindrance for gases, moisture and so on.

The overall need of packaging material should be exhibiting high strength, durability along with biodegradability, antibacterial properties and safe to human and environment. The packaging material can be used as a primary package or secondary or tertiary. Corrugated boxes are mostly used as tertiary packaging material. They are mostly recycled for different purposes and contain a major portion of recycled fibers. The recycling process detrimentally affects the strength properties and other important properties of old corrugated containers (OCC) pulp. To impart strength to the recycled fibers, blending with virgin softwood (SW) or other pulp and addition of dry strength additives and synthetic polymers is

generally practiced. The major drawback of using synthetic polymers lies in the hazards they pose to the human health and the environment. Some of the commonly used synthetic polymers are known to cause mutations, cancers and severe health problems [11]. Since major portion of paperboard packaging is used for the food and beverages segment, it straight forward affects the health of consumers. The food wrapping paper should be antibacterial, to make a carton box or corrugated board (liner or medium) or carry bag, etc different properties are required but mostly all these packaging need a raw material of higher strength (tear, tensile, burst, stiffness, RCT, CMT etc.). For these reasons, the strength additive should be a green biopolymer with comparable potential to enhance the strength and other important properties of recycled fiber for packaging. The strength additives have been applied on low grammage wrapping paper as well as high grammage liner and medium.

Chitosan is a biodegradable polymer with high potential to be used as strength additive. It is the second most widespread natural polysaccharide derived from chitin that is extracted from the shells of sea animals like crustacean and insects [3]. Chitosan is constituted of anhydroglucose component bound together by oxygen linkages and exhibits the similar structure to cellulose that makes the leeway for its use as a papermaking additive for enhancing strength properties of paper/paperboard [3,11]. The presence of hydroxyl and amine groups in chitosan [3-4,9] $-(1,4)\text{-}2\text{-acetamido-}2\text{-deoxy-d-glucopyranose units and }-(1\text{-}4)\text{-}2\text{-amino-}2\text{-deoxy-d-glucopyranose units}$ all at once provides the chitosan a unique molecular structure that accelerates its applicability as a papermaking additive. Presence of

amine group makes it a polycation and due to electrostatic interactions, this polycation can be strongly adsorbed by anionic pulp. Chitosan improves the dry breaking length as well as the wet strength of paper by effectively facilitating fiber water interactions and crosslinking between the fibers of paper [3, 9].

The reported study was focussed on exploring the potential of chitosan on different types of OCC pulp and a softwood pulp in paperboard making in terms of strength properties like tensile index, burst factor, tear factor, CMT, TEA index, RCT and stretch.

2. Materials and Methods

2.1. Materials

The old corrugated containers pulps (OCC-1, OCC-2 and OCC-3) were prepared in lab by recycling. The softwood pulp was donated by a pulp and paper mill in North India. Different other additives like alkyl ketene dimer (AKD), cationic starch (CS), cationic polyamine fixing agent (CFA), retention aid were also donated by a pulp and paper mill. Chitosan flakes of 85% degree of deacetylation (DD) were procured from a chemical manufacturer in South India and the other chitosan, whose DD was not known, was donated by a supplier from IndiaMart online market place.

2.2. Characterization of pulp and wet-end chemicals

The methods and instruments used for characterization of pulp and wet-end chemicals are shown in table 1.

For the determination of moisture content and total ash of chitosan, the samples were kept at 105°C for 24 h in an oven and at 650°C for 3 h in a muffle furnace, respectively. For the determination of

moisture content and total ash of CS the standard method IS: 4706 (part II) was followed [11-12].

2.3. Preparation of wet-end chemicals

The cooking of CS 1% (w/v) was done at 90±2°C for 30 min. AKD emulsion was diluted to 1%, CFA solution of 0.1% concentration and retention aid solution of 0.1% (w/v) were prepared before addition to the wet-end paperboard making. Chitosan solution was prepared in 1% acetic acid at room temperature by stirring for 4 h using magnetic stirrer. In the sets in which all wet-end chemicals were added, the sequence was as follows: CFA-200 g/t, strength aid (CS or chitosan as per dose), AKD-1 kg/t and finally retention aid 200 g/t was added to the pulp slurry after consistency make up for handsheet making.

2.4. Handsheets preparation using different pulps with and without other chemicals and evaluation of different paperboard properties

Papermaking strength additives improve the strength properties by escalating hydrogen bonding between cellulosic fibers. For OCC-1 both CS (different dose 30 kg/t and 40 kg/t) and chitosan (different doses from 1 kg/t to 7.5 kg/t) were tried as strengthening agent at wet-end with other wet-end additives. In OCC-2 only chitosan (different doses from 0.3 kg/t to 1 kg/t) and other wet-end chemicals were not used because of high charge demand of chitosan (anionic). For OCC-3 (refined and unrefined) and SW pulp only one dose of 5 kg/t of chitosan was used a strength additive with no other wet-end additives. Sets (A to Q) of handsheets were prepared using different pulps and additives as shown in Table 2.

S.No	Test parameter	Instrument	Method
1	Charge demand of pulp and other chemicals	Mutek PCD 03 pH Particle Charge Detector	Instrumental
2	Schopper Riegler (°SR) of pulp	Schopper Riegler (°SR) Tester	TAPPI T 227 om-09
3	Fiber classification	Bauer McNett fiber classifier	TAPPI T 233
4	Viscosity of CS and chitosan	Brookfield viscometer	Instrumental

Table : 1 The methods and instruments used for characterization of pulp and wet-end chemicals

Pulp	Cationic starch (kg/t)	Chitosan (kg/t)	Wet-end chemicals	Set
OCC-1	0	0.0	No	A*
OCC-1	30	0.0	Yes	B*
OCC-1	40	0.0	Yes	C*
OCC-1	0	1.0	Yes	D*
OCC-1	0	2.5	Yes	E
OCC-1	0	5.0	Yes	F
OCC-1	0	7.5	Yes	G
OCC-2	0	0.0	No	H

Pulp	Cationic starch (kg/t)	Chitosan (kg/t)	Wet-end chemicals	Set
OCC-2	0	0.3	No	I
OCC-2	0	0.5	No	J
OCC-2	0	1.0	No	K
OCC-3	0	0.0	No	L
OCC-3	0	5.0	No	M
OCC-3 (refined)	0	0.0	No	N
OCC-3 (refined)	0	5.0	No	O
SW (refined)	0	0.0	No	P
SW (refined)	0	5.0	No	Q

* the test data of these sets is taken as control from our previous study [12].

Table : 2 The pulp, wet-end chemicals and strength additives during handsheets preparation

GSM of all sets was found in the range 204 ± 4 and 103 ± 3 for sets (A to K) and (L to Q), respectively. The bulk was in the range 1.57 ± 0.04 , 1.75 ± 0.04 , 2.32 ± 0.03 , 1.67 ± 0.01 and 1.29 ± 0.01 for sets (A to G), (H to K), (L to M), (N to O) and (P to Q), respectively. Sets of handsheets (A to Q) were prepared using different pulps and additives as per TAPPI T 272 sp-97 and pressed before drying as per TAPPI T 218 sp-02. The conditioning of handsheets prior to testing was done as per ISO: 187.

The thickness, burst factor, tensile index, stretch, TEA index, RCT and CMT (A Flute) were measured using L & W instruments as per standard methods. The average value of three repeat sets of experiments has been shown here.

3. Results

The study was done to analyze the effect of chitosan on different types of OCC pulps with and without other wet-end chemicals and on SW pulp without other wet-end chemicals. The end product paperboard made using chitosan as strength additive has higher strength properties than that made without strength additive.

3.1. Characterization of pulp and wet-end chemicals

Fiber classifier data showed that fibers retained over 28 mesh screen (+28 fraction) are long fibers and fines are those which pass through 200 mesh screen (-200 fraction).

Mesh Size	OCC-1	OCC-2	OCC-3	OCC-3 (refined)	SW (refined)
	(Weight %)				
+28	46.2	52.6	54.4	19.4	77.9
-28, +48	19.6	14.6	12.3	12.3	8.5
-48, +100	14.5	13.6	7.5	11.4	6.2
-100, +200	7.9	2.1	4.7	9.7	1.3
-200	11.8	17.1	21.1	47.2	6.1

Table : 3 Classification of pulp by Bauer McNett fiber classifier

The OCC pulps were having shown different types of fiber classification. The OCC-1 was having the least long fiber and fines whereas the OCC-3 pulp was having maximum long fibers and fines. The long fibers and the fines contents of OCC-2 pulp were lying between those of OCC-1 and OCC-3. The SW pulp has maximum number of long fibers. The refined OCC-3 has decrease in long fibers and increase in fines after refining.

Viscosity of chitosan and CS of 1% solution at 25°C was observed to be 185 cP and 25 cP, respectively. The pH, ash, moisture and charge demand ($\mu\text{eq/g}$) of chitosan were 4.20, 2.6%, 13% and 6030 (anionic), respectively whereas such properties for starch were found to be 6.2, 1.2%, 10% and 60.0 (anionic), respectively. Charge demand/ pH of CFA, CPAM and AKD was 3050 (anionic)/ 6.2, 1490 (anionic)/ 4.5 and 266 (anionic)/ 3.9, respectively [11-12].

3.2. Impact of addition of different wet-end chemicals, chitosan or starch to OCC-1 pulp on final pulp slurry and paperboard properties

The set A, consisting of only OCC-1 pulp, was having a cationic charge demand of $34.8 (\mu\text{eq/L})$ which was reduced after addition of different wet-end chemicals, starch and chitosan as $8.08 (\mu\text{eq/L})$, $6.0 (\mu\text{eq/L})$, $14.1 (\mu\text{eq/L})$ and $12.6 (\mu\text{eq/L})$ for set

B, set C, set E and set G, respectively. The chitosan used was the one whose DD was 85%. The reduction in charge demand of pulp is more on addition of higher dose of chitosan or starch with same amount of other wet-end chemicals because of anionic charge demand of both. The °SR of all sets was in the range of 22 ± 2 . The value of °SR was not considerably affected.

The tensile index, burst factor, tear factor, CMT, TEA index, RCT and stretch of set A were 20.1, 14.3, 75.1, 173, 378, 1.4 and 2.6, respectively as shown in fig 1, fig 2 and fig 3. These properties of paperboard after addition of 40 kg/t starch were increased to 29.9, 24.6, 92.8, 356, 920, 2.3 and 4.3, respectively. The tear factor, CMT, RCT attained by paperboard after adding 40 kg/t of starch were almost equal to that attained by adding 7.5 kg/t of chitosan during paperboard making at wet-end along with other chemicals whereas tensile index, burst factor, TEA index and RCT after adding 30 kg/t of starch were almost comparable to paperboard made using 5 kg/t of chitosan during paperboard making at wet-end with other chemicals. The RCT after addition of 30 kg/t and 40 kg/t of starch at wet-end was almost same.

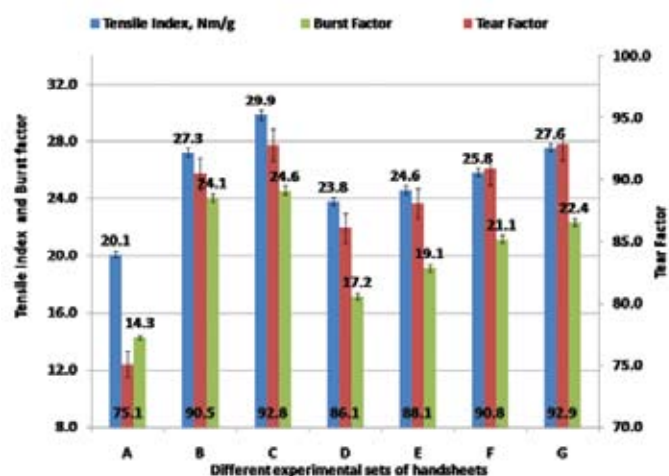


Fig : 1 Effect of wet-end chemicals, starch or chitosan to OCC-1 pulp on tensile index, burst factor and tear factor of paperboard

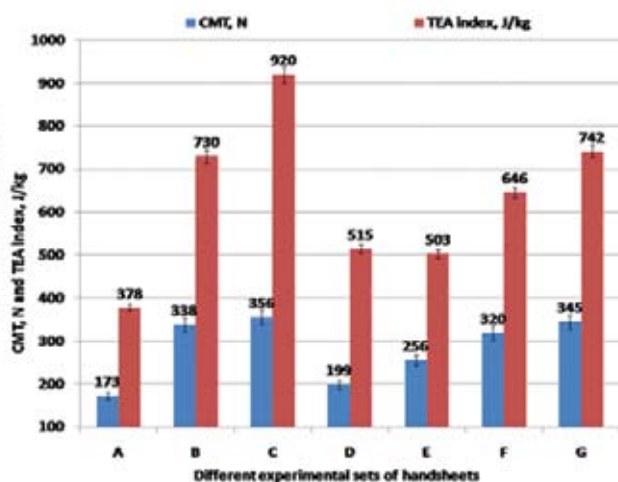


Fig : 2 Effect of addition of wet-end chemicals, starch or chitosan to OCC-1 pulp on CMT and TEA index of paperboard

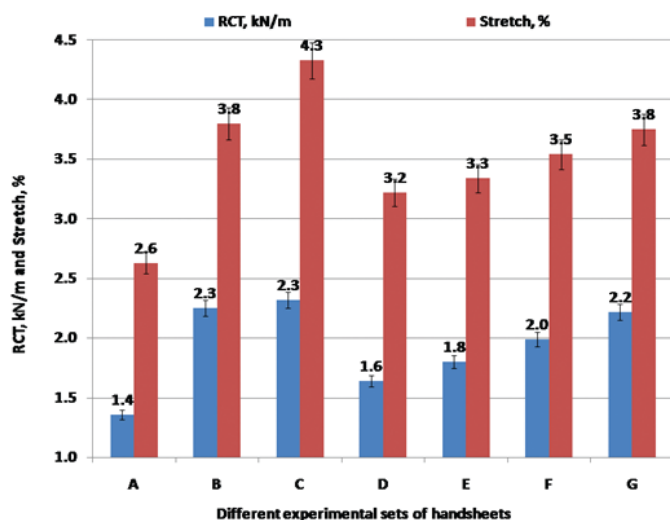


Fig : 3 Effect of addition of wet-end chemicals, starch or chitosan to OCC-1 pulp on RCT and stretch of paperboard

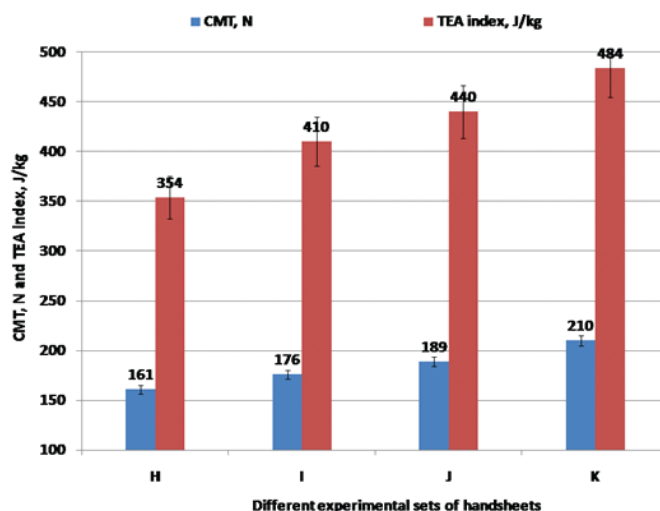


Fig : 5 Effect of addition of wet-end chemicals, starch or chitosan to OCC-2 pulp on CMT and TEA index of paperboard

3.3. Impact of addition of chitosan to OCC-2 pulp on final pulp slurry and paperboard properties

The only OCC-2 pulp has a cationic charge demand of 29.4 ($\mu\text{eq/L}$) which after addition of different chitosan doses like 0.3 kg/t, 0.5 kg/t and 1.0 kg/t reduced to 26.12 ($\mu\text{eq/L}$), 20.42 ($\mu\text{eq/L}$) and 15.40 ($\mu\text{eq/L}$), respectively. The chitosan used was the one whose DD was 85%. The other wet-end additives were not added to OCC-2 pulp because from the study of OCC-1 pulp it was confirmed that when chitosan dose was increased from 1 kg/t to 7.5 kg/t at fixed doses of other wet-end chemicals, still the charge demand of final slurry was reduced. The °SR of all sets made using OCC-2 was 20.

The burst factor, tear factor, CMT, and RCT of OCC-2 pulp was 11.5, 68.9, 161 and 1.21 which was further increased by 30.0%, 11.6%, 30.4% and 39.7%, respectively after adding 1 kg/t of chitosan only as wet-end additive. The increase in tensile index, TEA index and stretch was also significant after adding 1 kg/t of chitosan. The lower doses of chitosan like 0.3 kg/t and 0.5 kg/t to OCC-2 pulp also showed good impact on the strength of paperboard as shown in fig 4, fig 5 and fig 6.

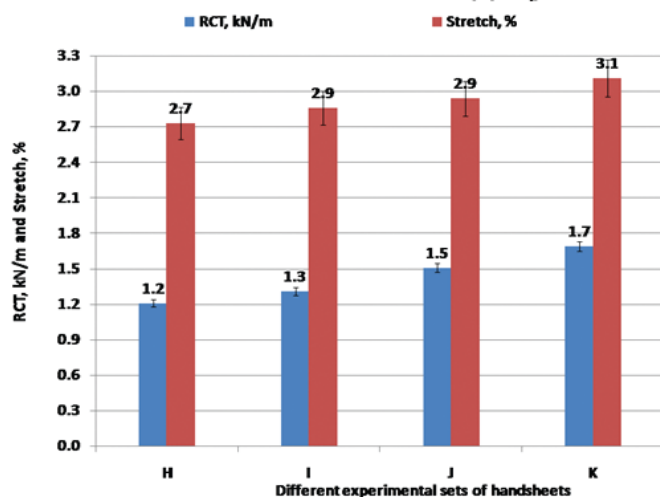


Fig : 6 Effect of addition of wet-end chemicals, starch or chitosan to OCC-2 pulp on RCT and Stretch of paperboard

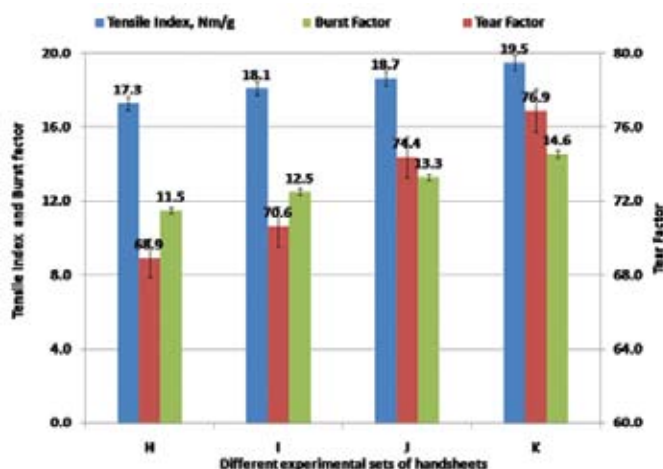


Fig : 4 Effect of wet-end chemicals, starch or chitosan to OCC-2 pulp on tensile index, burst factor and tear factor of paperboard

3.4. Impact of addition of chitosan to OCC-3 and OCC-3 (refined) pulp on final pulp slurry and paperboard properties

In the above study the OCC-1 and OCC-2 pulp after preparation showed the °SR in the range of 20 whereas the OCC-3 pulp °SR after preparation was only 10. Therefore, the effect of chitosan was studied on as such pulp (very low °SR). The OCC-3 pulp has a cationic charge demand of 20.1 ($\mu\text{eq/L}$) which reduced to 7.85 ($\mu\text{eq/L}$) after adding 5 kg/t chitosan dose. When 5 kg/t of chitosan was added to OCC-3 pulp the °SR was increased to 13 °SR from the initial 10 °SR value of as such pulp. In this study both chitosan were used, the one whose DD was 85% while for other the DD was unknown. The addition of unknown DD chitosan sample to OCC-3 pulp also increased the °SR when added at a dose level of 5 kg/t.

The tensile index, burst factor, tear factor, CMT, RCT and stretch of set L were 13.7, 12.3, 76.6, 54, 0.50 and 1.40, respectively while after addition of 5 kg/t dose (set M) of chitosan (known DD) all these properties were increased by 60.5%, 30.9%, 37.2%,

75.9%, 38.0% and 27.0%, respectively as shown in fig 7 and fig 8. The increment in properties was also observed after addition of unknown DD chitosan sample but the overall impact was less than that of known DD chitosan sample. This OCC-3 study showed that at such low °SR level the impact was positive on strength properties of paperboard. Therefore, the same OCC-3 pulp was refined for the further experiments.

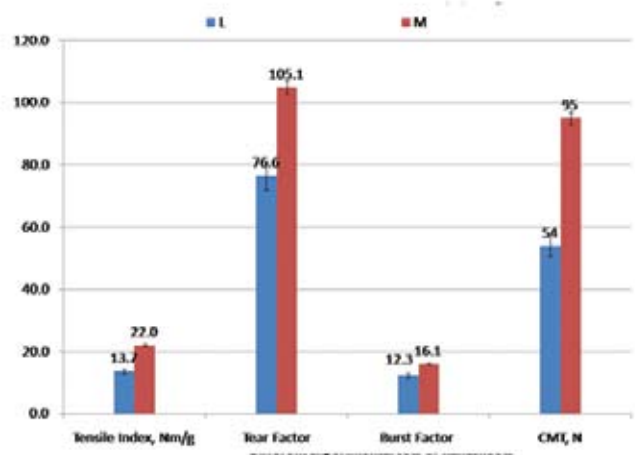


Fig : 7 Effect of addition of chitosan to OCC-3 pulp on tensile index, tear factor, burst factor and CMT of paperboard

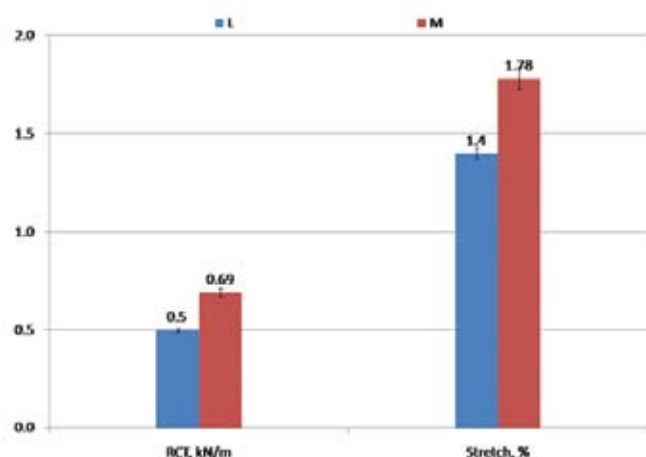


Fig : 8 Effect of addition of chitosan to OCC-2 pulp on RCT and stretch of paperboard

As the impact on OCC-1 and OCC-2 at a °SR level of 20 was already done therefore the OCC-3 pulp was refined to a level at which the °SR attained was 30. The refining of OCC-3 pulp increased the charge demand of pulp slurry to 24.4 (µeq/L). The refining increased the tensile index, burst factor, tear factor, CMT, RCT and stretch of the OCC-3 pulp (Set N). The addition of chitosan (known DD) at a dose 5 kg/t increased all the properties in comparison to the refined pulp as shown in fig 9 and fig 10. The tear factor of set O was only 96.6 which increased from 92.6 of set N but that of set M was 105.1. Similarly, the CMT of set M was 95 while that attained after adding 5 kg/t chitosan to refined OCC-3 pulp it was only 96. These results showed that CMT and tear factor were increased less in comparison to other strength properties after addition of chitosan to the refined OCC-3 pulp. This observation is not universal, for all pulps this cannot be concluded, to conclude a

further laboratory study need to be planned. The similar impact was shown by unknown DD chitosan sample on OCC-3 refined pulp but it was less than that of known DD chitosan sample.

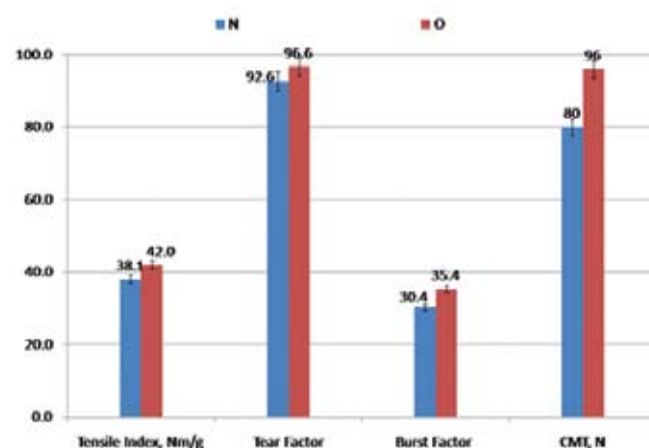


Fig : 9 Effect of addition of chitosan to OCC-3 (refined) pulp on tensile index, tear factor, burst factor and CMT of paperboard

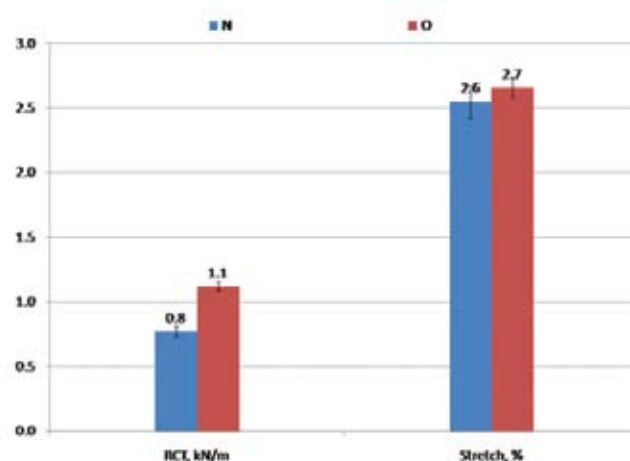


Fig : 10 Effect of addition of chitosan to OCC-3 (refined) pulp on RCT and stretch of paperboard

3.5 Impact of addition of chitosan to SW (refined) pulp on final pulp slurry and paperboard properties

The paperboard industry is mostly governed or managed by OCC pulps but when the paperboard maker need to further enhance the strength properties and the application of wet-end chemicals as well as the surface sizing is at their maximum level. The only scope the paperboard maker has is to blend some superior cellulosic fiber (SW or imported fibers/ pulp). Therefore, in the final part of this study the impact of chitosan to the SW pulp was explored. The °SR of refined SW pulp was 28 which was increased to 29 after 5 kg/t of chitosan (known DD) addition to the SW (refined). The refined SW pulp had a cationic charge demand of 35.1 (µeq/L) which was reduced to 18.9 (µeq/L) after 5 kg/t chitosan addition to the pulp.

The tensile index, burst factor, CMT, RCT and stretch of set P were 65.7, 52.2, 103, 1.1 and 3.5, respectively. When 5 kg/t of chitosan was added all these properties were increased as shown in fig 11 and fig 12.

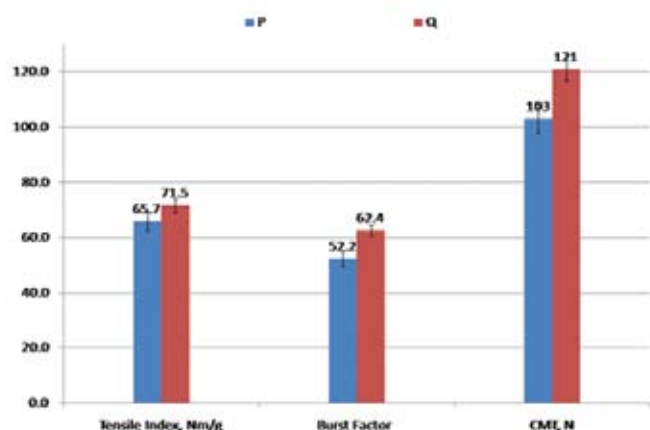


Fig : 11 Effect of addition of chitosan to SW pulp on tensile index, burst factor and CMT of paperboard

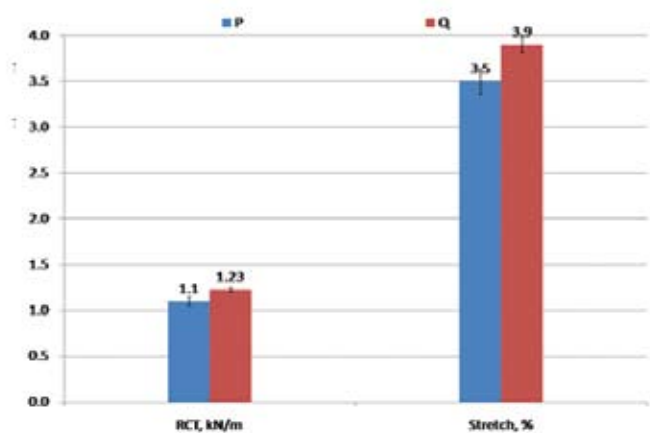


Fig : 12 Effect of addition of chitosan to SW pulp on RCT and stretch of paperboard

4. Discussion

Addition of wet end chemicals, chitosan and starch to OCC-1 pulp furnish decreased its charge demand in comparison to the control. The wet end chemicals, chitosan and starch were cationic in nature so, their application over the anionically charged fibers reduced the resultant charge of the pulp. On increasing the dose of chitosan to different pulp sets, the charge demand was reduced significantly that corresponded to the polycationic nature of the chitosan [13]. The strength properties like tensile index, burst factor, tear factor, RCT, CMT, stretch and TEA index were improved on addition of strength additives. This was attributed to the increase in non-covalent bonding of the strength additives with the cellulosic fibers. Due to chemical structure of these strength additives,

the hydrogen bonding also enhanced. In pulps, where different concentration of chitosan were added, the strength of pulps was improved significantly as in chitosan, sugar backbone is substituted with positively charged amino group at C2 position which provides high cationic character to it facilitating chitosan retention on cellulosic fiber due to increased ionic linkage and fiber adhesion. This film forming nature of chitosan provides the scope of welding between the fibers which enhances the fiber bonding strength that provides improved strength properties [11,14]. In present study, it was also observed that the role of fiber fraction is also important because it's the fiber fraction that also contributes to chitosan retention (more on fines because of more fibers to weld and complete film forming) as well as the inherent properties of long fiber. The strength of paper made of pulp having more long fiber will be more but when the short fibers will retain chitosan it will be increased more as seen in OCC-3 (unrefined). After adding 5 kg/t of chitosan to OCC-3 (unrefined) pulp strength properties were increased more than that attained by OCC-3 (refined) pulp after addition of same dose of chitosan.

5. Conclusion

The addition of wet-end additive, chitosan or starch to the OCC-1 pulp reduced the charge demand of the paperboard making system. Only addition of chitosan to OCC-2, OCC-3 (refined and unrefined) and SW (refined) also reduced the charge demand of the paperboard making system.

The °SR of the paperboard making system was not much affected by the addition of chitosan to OCC-2, OCC-3 (refined or unrefined) and SW. As the °SR was not much effected therefore the drainage time of pulp would also not be effected.

The strength properties like the tensile index, burst factor, tear factor, CMT, TEA index, RCT and stretch in all pulp cases were increased when chitosan at different doses was added (like in OCC-1 and OCC-2 pulp) or at a fixed dose (like in OCC-3 (refined and unrefined) and SW). The short fiber and the long fiber fraction also contribute in the strength properties of the paperboard and the chitosan retention which ultimately affects the hydrogen bonding, the welding and the film forming on the fibers provides positive impact on the strength properties.

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