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PRODUCTION OF PACKAGING MATERIAL USING RECYCLED FIBERS AND UNBLEACHED SUBABUL SAPWOOD PULP THROUGH NANOTECHNOLOGY




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Abstract:

In the packaging industry, there are significant incentives and pressures to reduce the use of plastics and the allied products. As a direct result of this, there is a growing demand for packaging products which are renewable, recyclable, and biodegradable. These advantages arise from the fact that the raw materials utilised are plant-based and/or recycled fibers. In the present work, Subabul sapwood pulp and recycled "Amazon packing" soaked pulp was utilized to make nanocellulose films. First, the Subabul sapwood chips (2cm x 2cm x 2.5 mm) were subjected to Kraft cooking at 165^{0}C for 3 h. The unbleached pulp obtained from the cooking and the recycled amazon packing soaked pulp was taken in 50:50 ratio and soaked for the 48 h. Pulp refining was carried out in two different ways: one is Super Masscolloider (SMC) and the other one is Lab Valley beater (LVB). Initially, the refining of this pulp slurry was carried out in SMC using 1 mm clearance. Later the gap between the two grinders of SMC was reduced to 0.4, 0.1, and 0 clearances, so the combination of micro and nanocellulose fibers can be obtained. The LVB refining is carried out at 0 clearance only. These fibers can be used to make paper plates with reduced porosity by using Hand Sheet Former equipped with vacuum filtration. During the recycling and refining processes, a significant number of big pores (collapsed lumen) and the majority of the very small pores in the cell wall get closed, resulting in the less coarse and fiber surface area. The porosity and the burst strength measurements were carried out on these paper sheets. It was taking more time for air to pass through the paper sheets compared to the commercial paper plates without coating. In the Burst test also, more stress was reported for SMC based paper sheet without coating. This added advantage is due to high packing density of nanofibers which will enable liquid food consumption easier without plastic wrap and these are biodegradable in nature.

Keywords: Nanocellulose, Recycled fiber, Kraft cooking, Super Masscolloider, Porosity, Burst test.

1. Introduction:

Food packaging to store food or paper plates to consume food is an emerging field in developing countries like India. Packaging is characterised as a protective layer that is located outside the food, and its primary function is to safeguard the food from any potential physical, chemical, or biological risks [1]. Paper plate is characterized as thick plate that should have a zero permeability for liquid food. Research has been done on a wide variety of polymers, many of which are already commercially available and contribute significantly to the production of packaging films or paper plates. The packaging business makes extensive use of a variety of polymers, including cellulose and its derivatives, which are among the most common and prevalent types. Polymers can be classified into two primary categories: natural and synthetic. Natural polymers are a type of biopolymer that are derived from natural sources, such as extracting them from plants, obtaining them from food wastes (such as the peel of bananas or oranges), or obtaining them from microbes, among other natural sources [1].

Utilizing natural polymers or green polymers is one way to cut down on the amount of toxic non-biodegradable substances that are created each day by a variety of different industries. Cellulose, one of the many types of polymers, is one of the most abundant polymers on earth. It may be easily extracted from the cell walls of plants, where it is found naturally. In recent years, there has been a rise in interest in nanocellulose due to the fact that it is a renewable and it has a high surface area. Wood-based nanoparticles can be used in a wide variety of products and applications today as a direct result of on-going advancements in nanotechnology [2-4]. Cellulose Nanofibers (CNF) are being heralded as a potentially game-changing material of the future. CNFs are more environmentally friendly alternative to synthetic petroleum-based materials for use in applications such as food packaging [5], coatings [6], binders [6], and high strength sheets due to their high surface area [5].

Recycling of recovered paper has become a fast expanding area of the pulp and paper industry due to the combined effects of economics, policies aimed at conserving resources, and legislation mandating the use of recycled content. In recent years, there has been a substantial development in the use of recycled fiber (RCF), which stands for recycled content fiber, in printing paper grades, particularly magazine papers. The swelling and bonding abilities are partially regenerated by the various RCF therapies, but this is not sufficient [5]. In order to regenerate the fibers, a more extensive treatment such as refining is required. The formation of fibrils during refining contributes to an enhanced propensity to bond. The Lab Valley Beater, PFI Mill, Super Masscolloider, Micro fluidiser, and High Pressure Homogenizer are all examples of several mechanical treatments that can be utilized for the refining process. Several researchers have provided evidence that supports the validity of studies that involve the extraction of CNF by the use of mechanical grinding [3]. Nanocellulose has a tremendous potential for usage in applications relating to environmentally friendly packaging as a result of its high specific surface area as well as its strong mechanical qualities (sapwood fibers) [7] and biodegradable nature [8]. Unbleached sapwood sheets (nanocellulose sheets) are recommended to use in packaging applications [9]. There are a few different forms that nanocellulose can take, including cellulose nanocrystals, nanofibrils, micro fibrillated cellulose, and bacterial nanocellulose [10].

In this study, Kraft cooking process is used to convert sapwood part of Subabul into the cellulose fibers. Later, the mixture of "Cellulose fibers" and "Amazon packaging" soaked pulp was refined using SMC and LVB. The micro and nanocellulose fiber based sheets were prepared using SMC and LVB refined pulp by using Hand Sheet Former equipped with vacuum filtration. The porosity and burst tests were carried out on the SMC and LVB refined sheets. The sheets produced from this process can be used in packaging applications without any plastic wrap.

2. MATERIALS AND METHODOLOGY

2.1 Kraft cooking

Subabul sapwood, used as raw material in this study, was acquired from Hyderabad, India. Sapwood was separated from hard wood with the help of a wood turning machine and was cut down to small pieces (2 cm x 2 cm x 2.5 mm) using a chisel which was later dried at 103.5^{0}C in hot air oven. The sapwood chips were subjected to pre-treatment at 120^{0}C for an hour in the autoclave (Metalab MSI-41) to soften lignin and remove organic extractives. The Kraft process (delignification) was carried out in a Rotary Pulp Digester (UEC-2015) at 165^{0}C for 3 hours with 1:4 ratio of wood to liquor (Na_2S and NaOH expressed as Na_2O having 28% sulphidity, 20% Na_2O (active alkali)). 180-degree oscillation of digester is critical for maximum delignification. This digester is equipped with flat head and teflon gasket to avoid leakage while rotation. The drain valve at bottom is used for liquid discharge. The unbleached sapwood pulp was separated from black liquor after 3 hours and was dried at 103.5^{O}C in a vacuum oven for 6 hours after washing it thoroughly with deionized water.

2.2 Pulp refining

In Lab Valley Beater

LVB contains 2 main parts made of acid proof steel: stationary blade (beater plate) and rotating blades (beater roll) to obtain fine pulp (short fibers). The gap between these blades is reduced by adding weight of 5.5 kg. 25 g of sapwood pulp and 25 g of Amazon packaging soaked pulp was taken in 50:50 ratios. The pulp mixture of "Subabul" sapwood and "Amazon Packaging" is soaked in 2 litres of water overnight. 3 litres of water was added to the 2 litres of soaked pulp and fed from the top into the beater chest. The initial clearance value was 10 mm which was then reduced to less than 20 microns (which was considered as zero clearance). Wet pulp undergoes shearing action due to which the lumen gets collapsed, cell wall disintegrated and a slight change in colour was observed due to the removal of a small quantity of lignin (dark brown slurry). Refining of mixed unbleached pulp slurry was carried out for 60 min at 600 rpm. Finally, at zero clearance a decrease in size of cellulose fibers (short fibers) was observed which are in micron size.

In Super Masscolloider

SMC is an ultrafine grinder which consists of 2 grinders, one at the top and one at the bottom. The upper grinder is fixed in position whereas the bottom one is variable and rotating. These two grinders are made up of composites (SiC, Al_2O_3). The grooves on these grinders ensure nanofibers instead of nanoparticles. 25 g of sapwood pulp and 25g of Amazon packaging soaked pulp was taken in 50:50 ratios. This pulp mixture was soaked in 2 liters of water overnight. 3

Fig. 1. Vortex formation during refining of nanocellulose at Zero clearance (left). Stationary upper grinder with grinding zone of 17 mm width (gap between solid and dotted circles) (right)

liters of water was added to the 2 liters of slurry at 1 and 0.4 clearances, and 1 more litre at 0.1clearance, and 1 more litre at 0.01 clearance. The refining of unbleached pulp slurry was performed starting with 1 mm clearance at 1000 rpm which was then gradually decreased to 0.4, 0.1, 0.01 clearance (fibers refined for 15 min) and then refining of cellulose fibers at 0 clearance level for 60 min. As the clearance between the two grinders reduced, the diameter of fibers reduced and viscosity of slurry increased. The vortex formation (Fig. 1 - left) at zero clearance is minimized by placing one baffle. The clearance value had to be reduced while the SMC is running with wet slurry. Water was added to make sure of the smooth flow of slurry and that the reduction of size of fibers was only due to interaction between the grinder and fibers and not the interaction amongst the fibers themselves. Micro and nanocellulose fibers (long fibers) obtained from the Super Masscolloider are collected and stored in plastic bottles at 5^0C.

2.3 Preparation of micro/nanocellulose sheets

The preparation of micro/nanocellulose sheets using the Hand Sheet former equipped with Whatman filter paper (10 µm pores) along with vacuum capable of retaining fibers. The inner mesh diameter is 165 mm and the outer one is 175 mm. 1000 ml of LVB/SMC refined pulp slurry was poured into the equipment's feed inlet. Water from the sheet is removed by opening the pressure releasing valve and the vacuum is applied to remove the excess water present in the sheet. Sheet densification and unbound moisture removal happened through heavy roller, followed by drying in a hot air oven (Osworld OOG – 90) at 60^0C for 2-3 h.

2.4 Porosity measurement of the nanocellulose sheets

Porosity can be measured in terms of the time required for a specific volume of air to travel through a test specimen. The longer time means lower porosity. A gravity-loaded cylinder that employs a liquid seal to enclose an air volume within a chamber serves as the source of the pressurised air that is produced. This compressed amount of air is then delivered to the clamping gasket ring, which holds the test specimen in place. The downstream clamping plate has holes in it that allow air to travel through the specimen (the sheet) to escape into the atmosphere. As shown in Fig. 2, SMC_0_clearance sheet, LVB_0_clearance sheet, thin paper plate (aluminum foil is partially removed), and green paper plate (plastic wrap is partially removed) are used for porosity measurements.

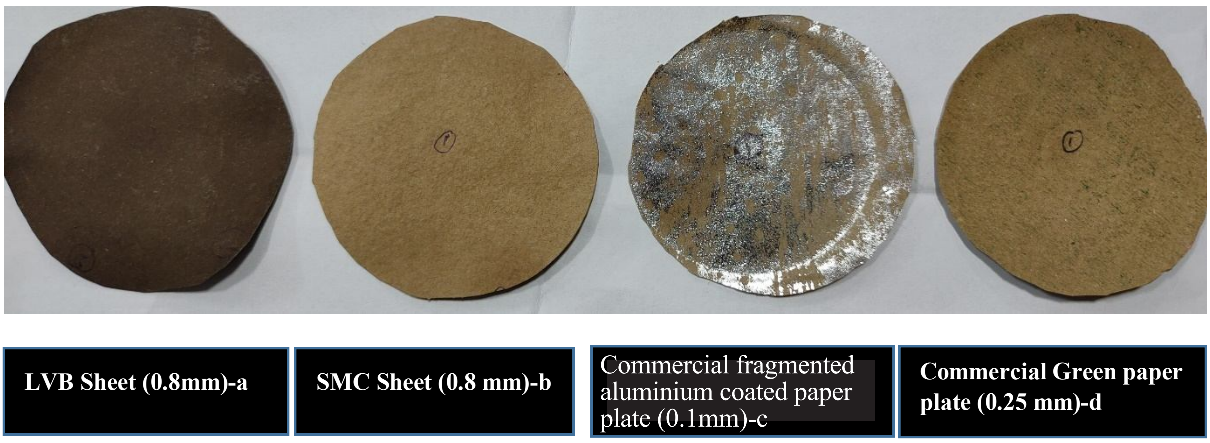

Fig. 2. Four different sheets used for porosity test

2.5 Burst strength of the nanocellulose sheets

Burst strength is the maximum pressure a material can withstand before it bursts or fails destructively under pressure. In this test, 3 mm probe with a contact area of 7.07 mm^2 is used with a test speed of 0.5 mm/sec, target distance of 5 mm, and specimen (sheet) size of 30 x 30 mm^2. The specimens were conditioned at 23^0C, and 55% relative humidity for 24 h prior to testing. Burst test is used to measure the pressure at which substances deform plastically. It helps in determining the weakest point in a product line and the failure mode of a material which is very important for pressurized components used in exposed conditions.

3. Results and Discussion

3.1 Scanning electron microscopy of LVB and SMC sheets

Micro and nanocellulose fibers obtained after LVB and SMC refining were examined using scanning electron microscopy (SEM). The samples were gold coated before examination. As shown in Fig. 3a and 3b, the nanocellulose fibers are in the range of 60-260 nm and micro cellulose fibers are in the range of 1-5 μm respectively.

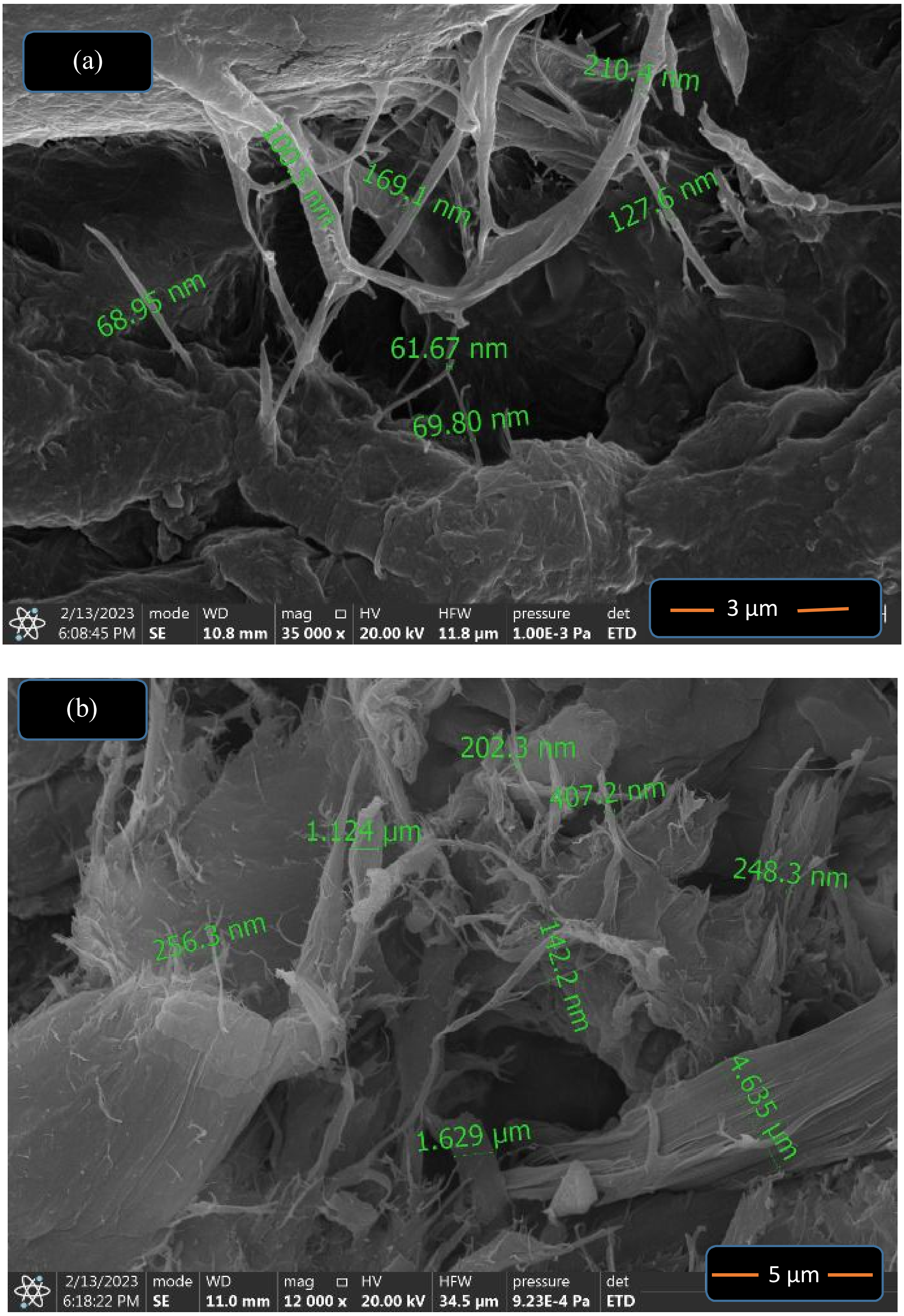

Fig. 3. SEM of sheets made from SMC (a) and LVB (b) refined fibers

3.2 Porosity test of the sheets

The Gurley and the Sheffield are the two primary types of devices that are used in porosity measurements. The Gurley instrument is utilized for testing porous materials and measures the amount of time needed for a certain volume of air to pass through a single sheet of material. If the reading is high, it means the paper is dense and has fewer pores. It is well known that the air permeability of sheets has a one-to-one correlation with the porosity of those sheets; specifically, the higher the porosity, the higher the permeability.

Micro and nanocellulose sheets obtained after LVB and SMC refining, commercial aluminum coated and green paper plates were tested for air permeability with different volumes of air (100, 150, 200, 250, and 300 ml). SMC and LVB sheets are 0.8 mm thick, aluminum coated paper plate is 0.1 mm thick, and green paper plate is 0.25 mm thick. SMC sheets took more time compared to LVB sheets due to the more number of nano-fibers and higher packing density. Time taken to passage of air is increasing by increasing volume of air which indicates homogeneous flow of air through the specimen. Fragmented aluminum coated paper plate took less time compared to LVB and SMC sheets due to less thickness and also due to the micron size fibers (low packing density). Green paper plate took more time compared to all sheets due to fragmented plastic wrap. As shown in Table 1, the difference in time taken to pass through the sheets is 15-20 % between SMC sheet (without coating) and green paper plate (fragmented plastic wrap) though 220% variation exists in thickness. Normally this green paper plate is used with plastic wrap of less than 40 μm causing tonnes of plastic waste. It is known that air permeability decreases by the coated layer in case of commercial paper plates, hence tests were carried out with fragmented aluminum or plastic wrap based sheets. From these results, it is clear that presence of nanocellulose fibers in SMC sheets are contributing towards less porosity which can replace the plastic wrap based paper plates.

3.3 Burst strength of sheets

Maximum pressure applied on a specimen with a 3 mm diameter probe (contact area of 7.07 mm^2) is continuously increasing with depth while loading and decreasing with unloading. Burst test is useful for packaging paper, newsprint, bag paper and printing papers to understand the level of packing density. Increasing the number of longer fibers (shorter diameter) with high surface area by increasing refining time leads to high packing density and high burst strength. Burst test is carried out on the LVB and SMC refined sheets. SMC sheets have little higher burst strength compared to LVB sheets as shown in Table 2. In case of LVB sheets, packing density is obtained using more number of short length fibers. So it is important to have mix of short length and short diameter fibers. SEM fractography after burst test is being carried out for LVB and SMC sheets after gold sputtering (Fig. 4). Noted the brittle fracture (Fig. 4c) and ductile fracture (Fig. 4a) in LVB sheets and SMC sheets respectively. Ductile fracture in SMC sheets is due to the long and low diameter (nm) fibers/fribrils as shown in Fig. 4b. It is important to note that LVB have shorter length and large diameter fibers (fibrils of nm size shown in Fig. 4d are small in length) whereas SMC have shorter diameter and high length fibers, which play main role in interconnecting fibers in fabricating sheet [4].

Table. 1. Time taken to pass the volume of air through a refined sheets and commercial paper plates

Refining (LVB/SMC)	Time (seconds)				
	100 (ml)	150 (ml)	200 (ml)	250 (ml)	300 (ml)
LVB_0_60 (0.8 mm thick)	10.30±1.4	18.43±2.7	26.33±4.1	26.33±4.1	43.42±6.7
SMC_0_60 (0.8 mm thick)	14.72±2.9	25.90±4.8	37.76±7.3	50.42±10	62.79±12.2
Fragmented aluminum coated paper plate (0.1 mm thick)	7.075±0.5	12.35±0.8	17.44±1.2	22.97±1.9	28.69±2.5
Green paper plate with fragmented plastic wrap (0.25 mm thick)	16.46±0.5	29.93±2.0	42.57±3.9	55.86±4.4	72.05±4.8

Table 2. Burst strength of SMC and LVB refined sheets

Refining method	Burst Strength (N/mm^2)
SMC_0_60 min	22.35 ± 0.88
LVB_0_60 min	19.98± 1.54

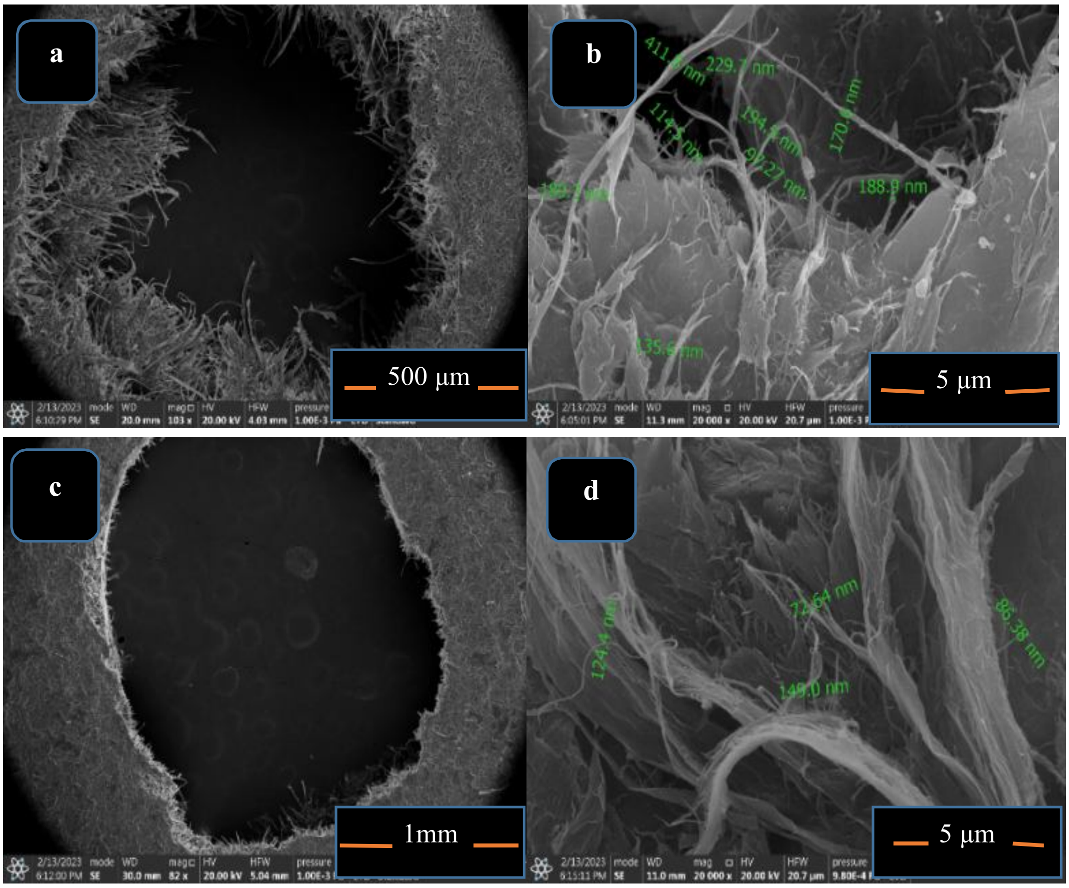

Fig. 4. SEM images of Burst tested samples of SMC (a, b) and LVB (c, d) sheets

Recycling of packaging grade paper (unbleached), recycling of office paper (bleached) and producing new pulp using these recycled pulp either 100 % or by mixing partially with fresh pulp is already in commercial practice. But converting the low grade packaging material into high grade nanocellulose is new and can be categorized as green manufacturing of nanocellulose (avoids cooking and partially bleaching chemicals). Lignin present in cell walls is partially removed while doing refining especially at zero clearance of SMC refining. In this particular investigation, the raw material consisted of recycled Amazonian packaging as well as fresh Subabul sapwood fibers are considered. We are able to save resources and cut down on our overall energy usage if we use recycled fibers. A growing number of e-commerce businesses are rapidly embracing environmentally responsible business practices such as green manufacturing of nanocellulose using their disposed packing material.

4. Conclusions

From this study, it is concluded that refining on LVB and SMC greatly affects the morphology of cellulose fibers. LVB resulted short length and high diameter fibers by decreasing the gap (zero clearance) between beater roll and beater plate. SMC resulted the long length and small diameter fibers by decreasing the clearance (zero) between two grinders. Most of the nanocellulose is obtained at the zero clearance which should be performed as wet grinding only. The micro and nanocellulose fibre based sheets (LVB and SMC) were fabricated using hand sheet former equipped with vacuum filtration. During the recycling process, a significant number of big pores (collapsed lumen) and the majority of the very small pores in the fiber wall closed (recycled fibers), resulting in the less coarse and porous fiber surface. Burst test and porosity test were carried out on the LVB and SMC sheets. From SEM, it is observed that the micro cellulose is in the range of 1-5 microns, and nanocellulose is in the range of 60-260 nm. SMC refined sheets have high burst strength (10 %) and low porosity (40 %) compared to LVB refined sheets due to the more number of nano fibrils which also led to ductile fracture of the sheet. SMC sheets without coating (0.8 mm thick) and Green paper plate with fragmented plastic wrap (0.25mm thick) showing only 15 % difference in porosity value even though 220% difference exists in thickness. The high burst strength and low porosity of SMC_0_60 min sheet is very useful in green packaging (food packaging), paper plates (without plastic wrap) and also in dust capturing applications due to its high surface area, high packing density of the sheet and biodegradability.

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