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## The effect of green biobased binder on structural, mechanical, liquid absorption and wetting properties of coated papers

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### ABSTRACT

Synthetic styrene-butadiene (SB) and styrene-acrylic (SA) latex binders used in paper coating formulations are common and based on unsustainable petroleum sources. Today's papermaking industry turns towards sustainable substitutes that do not compromise quality, and reduce carbon emission, toxic substance release and waste disposal concerns related to fossil fuel sources. In this study, colloidal starch-based latex nanoparticles that do not require cooking were used for pigment coating and coated on the paper surfaces. The effects of these new biobased binders on the structural and mechanical strength properties, liquid absorption, wetting and surface topography of the paper were investigated and compared with the properties of coated paper that SB latex was used as a coating binder. The results demonstrated that the biobased latex decreases the grammage of paper while maintaining an identical thickness relative to the SB latex samples. The porosity, permeability and roughness of biobased latex are found higher than the SB latex. The biobased latex successfully replaced SB latex at 1:1 ratio and enabled an equivalent of bursting, tear, tensile strength and structural properties.

### 1. Introduction

Structural, mechanical, optical and barrier properties of paper-based products are directly related to fiber-fiber interactions, single fiber properties and the chemical and mechanical structure of the coating layer applied on paper surfaces (Kulachenko, 2012). The binder in the coating layer is critical for film formation and imparts certain properties such as flexibility, deformability and elasticity to the paper products (Tayeb, 2017). The type of the binder controls the fractional void volume called porosity and the liquid resistance of the coated layer, while the strength of binder withstands tensions applied during printing and post-printing applications such as creasing, folding, glueing and binding (Coffin, 2017).

Non-renewable petroleum-based synthetic binders (i.e., SB and SA latexes binders or polyvinyl acetate) have been widely used as coating binders in paper and packaging applications to improve mechanical properties (Zhu, 2018). Non-degradable polymers decrease the

biodegradability of papers and make recyclability harder than their sustainable counterparts (Lavoine, 2014). The price of these petroleum-based materials escalates depending on increases in the oil price that are subject to both political and economic effects, forcing paper manufacturers —*who are the single largest users of the SB latex with ~35% of global consumption*— confront increased production costs (MarketersMedia, 2021). Paper companies are looking for sustainable solutions that have a good environmental footprint (Flory, 2013).

Starch is a sustainable binder material and one of the most abundant, renewable carbohydrate-based biopolymers in nature (Sheikhi, 2018). Salam reported that starch is often used as a co-binder with synthetic materials and can improve the biodegradability and recyclability of paper products (Salam, 2013). Conventionally, modified starches are purchased as granular powder and dispersed in water for cooking or they are chemically/enzymatically modified into a colloidal dispersion prior to its addition in the production line (Klass, 2011). Cooking high molecular weight starch slurries can be challenging due to the improper

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**Table 1**

The results of replacing SB latex at 1:1 ratio by the biolatex.

Properties	Reference
73% decrease in carbon footprint	Lee (2010)
Improved brightness	Lee (2010)
Improved gloss	Lee (2010)
Improved opacity	Lee (2010)
1–3% improved additive usage	Shin (2013)
Decreased the use of water	Shin (2013)
Decreased the use of energy	Shin (2013)
Decrease in CMC, polyvinyl alcohol or rheology modifier usage	Shin (2013)
Improved calendering performance	Shin (2013)
Improved foldability	Klass (2007)
Improved stiffness	Oberndorfer (2011)
Decreased binder migration	Figliolino (2009)
Improved solid level adjustment	Shin (2013)
Improved rheological properties	Chen (2016a)
Improved curtain coating stability	Chen (2016b)
Pick strength	Chen (2015)
Cost saving	Lee (2015)

**Table 2**

Coating formulation ingredients (Dry parts).

Formulation	SB latex	Biobased latex
Pigment	GCC	60
	Clay	40
Binder	Starch	2
	SB latex	8
	Biolatex	0
Additive	Dispersing agent	0.2

granule rupture and the low solids required to manage the viscosity of the resulting starch. In addition, the amylose molecules of starch rearrange and cause retrogradation/setback problems at the cooling stage.

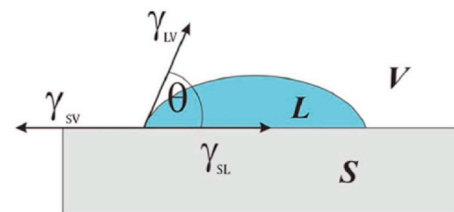
Pal reported that the efforts have been focused on increasing the use of renewable, biodegradable, sustainable, and recyclable materials in paper and packaging industries as well as reducing water and energy consumption in production and creating product innovations that are multi-dimensional in nature (Pal, 2017). Replacing synthetic polymers with more natural materials such as biopolymers in paper coating processes is suggested to solve these problems (Khwaldia, 2010). Han presented a number of biodegradable and renewable biopolymers such as microfibrillated cellulose, whey proteins, chitosan, alginates and starch have been evaluated as a paper coating material (Han, 2010). Zhu showed that the biopolymers improve the strength properties of papers (Zhu, 2018).

Wildi, Egdorn and Bloembergen patented that a multifunctional starch-based nanoparticle biolatex™ (biolatex) has been offered as an alternative to the synthetic binders for papermaking, adhesives, molecular biology and chemistry, food and medical applications (Zhang, 2018). The biolatex is also reported to be used as thickener, rheology modifier, adhesive, wet-end additive or surface sizing agent (Altay, 2017). Wildi explained in the patent that a reactive extrusion process utilizes crosslinking agents and suitable processing aids to prepare the biolatex as a ready-to-use material; thus, eliminates starch cooking procedure and decrease carbon emissions (Wildi, 2017). The high molecular weight and high density of hydroxyl groups of the biolatex nanoparticles enable high binding strength, compatible to SB and SA latex. Table 1 presents up to date the research findings that are obtained by the replacement of SB latex binder at 1:1 ratio with the biolatex. The research revealed that the biolatex improves optical properties of paper, reduces material and resource usage and has better calendering, folding and stiffness performance; however, the biolatex effect on structural and mechanical properties of papers is still an unknown. In this study, the effect of replacing SB latex by biolatex on structural and mechanical

**Table 3**

Experiments according to TAPPI procedures.

Properties	Reference
Grammage	T-410
Thickness	T-411
Contact angle	T-458
Moisture content	T-550
Burst strength	T-403
Tear strength	T-414
Tensile strength	T-494



**Fig. 1.** Schematics of the equilibrium of forces determining the contact angle of a liquid on a surface (Pekarovicova, 2005).

strength properties of papers is investigated. The effect of biolatex on surface energy and surface topography is also studied to investigate the effect on liquid absorption ability and wetting properties.

## 2. Materials and methods

**Coating formulation:** Table 2 represents the coating formulations that contain ground calcium carbonate (GCC) and clay pigments. SB latex binder in the control group was replaced at 1:1 ratio by a colloidal biolatex nanoparticle binder (*EcoSynthetix, Burlington, Canada*). The nanoparticles were formed from a high amylopectin-based starch (>95% amylopectin, <5% amylose) (Wildi, 2017). The coating formulation was applied onto 95 gsm wood-free base paper surface at 12 gsm coat weight using a bent blade coater operated at 3,000 ft/min (915 m/min). The paper samples provided by EcoSynthetix were coated and calendered by an undisclosed papermaking mill.

**Structural and mechanical properties:** The samples were conditioned prior to testing and tested at %50 ( $\pm 2$ ) relative humidity and 23 °C ( $\pm 1$ ) temperature for 24 h according to TAPPI T-402 standard (TAPPI: Technical Association of the Pulp and Paper Industry). The data evaluated using two-sample independent *t*-test using JMP 14 statistical software to obtain *p*-values. The confidence level was set to  $\alpha=0.05$  (95%). The hypothesis was to investigate whether any of the differences between the means of structural and mechanical properties of the samples were significant. The tests performed according to TAPPI procedure listed in Table 3 and the average of 10 replicates per test was reported. The burst strength measured using a Mullen tester (*BF Perkins: Chicopee, MA*), tensile strength measured using an Instron tester (1 kN load cell, 25.4 mm/min load rate. *Model 43K1: Instron, England*) and tear strength measured using an Elmendorf tester (*1,600 gf pendulum capacity, 9.8 m/s<sup>2</sup> gravitational rate. TMI: New Castle, DE*). Load cell in the Instron was 1 kN and the speed of jaw separation rate was 25.4 mm per minute. Tear Tester was 1,600 gf, and the rate would be gravity.

Surface roughness was measured using 3D profilometry, Contour GT-KO vertical scanning white light interferometer microscope (*Bruker Corporation, Billerica, MA*) with a 5x lens at 100  $\mu$ m back scan, 100  $\mu$ m length and 5% threshold.

The permeability coefficient was calculated by Darcy's equation (Pal, 2006). PPS porosity, compressibility and roughness were measured by the Parker Print Surf method (*TMI Testing Machines: New Castle, DE, US*). A scanning electron microscopy (SEM) was used for the microstructure imaging of sample surfaces.

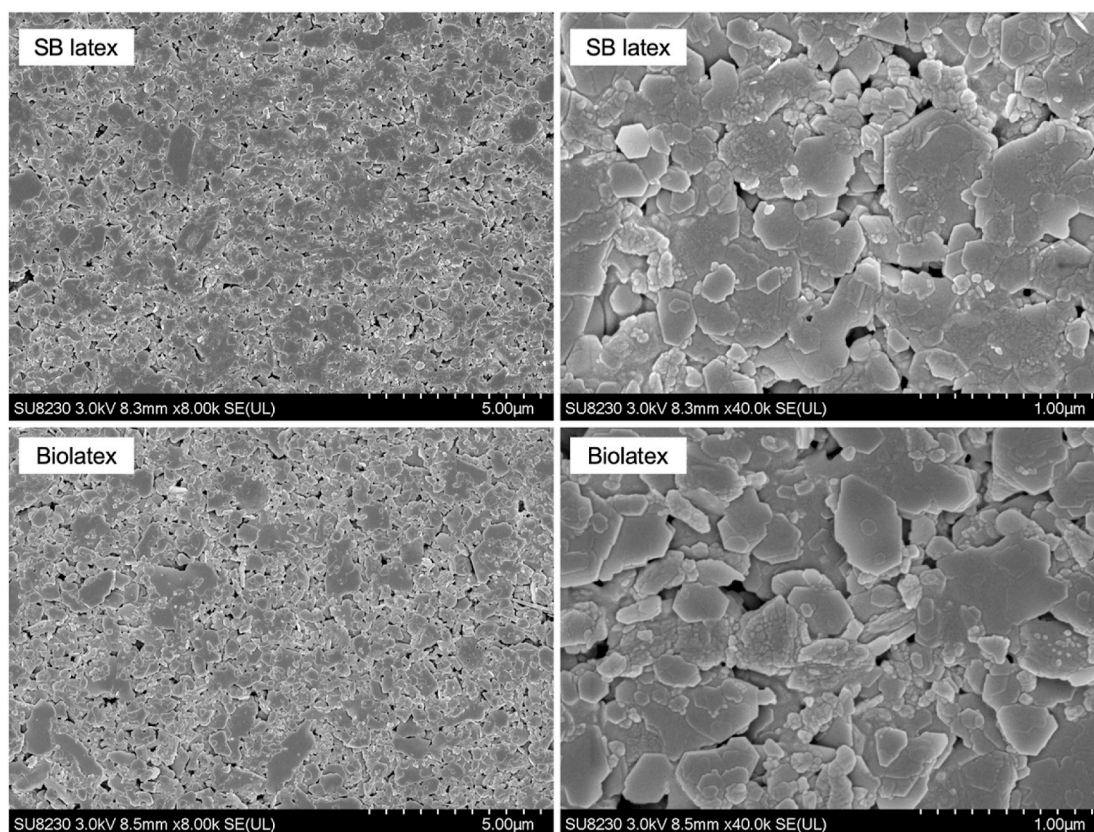


Fig. 2. SEM images of the microstructure of the SB latex and biolatex coating surface: magnified at 8Kx (left) and 40Kx (right).

**Liquid contact angle and time dependent drop volume change analysis:** Contact angle between liquids and polymer film surface measure the wettability of the paper surface (Aydemir, 2019). When the drop of liquid is placed on a surface, an equilibrium is established as presented in Fig. 1. The angle defined by the Young equation in Eq. (1):

$$\gamma_{SV} = \gamma_{LV} \cos \theta + \gamma_{SL} \quad (1)$$

where  $\gamma_{SV}$  is the surface energy of solid/vapor interface,  $\gamma_{LV}$  is the surface tension of liquid/vapor interface and  $\gamma_{SL}$  is the surface energy of solid/liquid interface.

An FTA200 (First Ten Angstrom; Portsmouth, VA) video system was used to measure surface tension and contact angle analysis. The system was calibrated prior to running the experiments. Test liquids were deionized water (DI) (ultra-filtered, density 1 g/cc; Fisher Scientific, Fair Lawn, NJ) and diiodomethane (MI) (reagent plus 99%, density 3.325 g/cc; Sigma Aldrich, St. Louis, MO). A 10-mL syringe (BD Luer Lok, Becton Dickinson, Franklin Lakes, NJ) with a 0.9 and 0.5 needle tips (JG20-1.0., Jensen Global, Santa Barbara, CA) was used to dispense DI and MI under ambient conditions, respectively. Five drops were dispensed, and the average surface tension was reported. For the contact angle, the paper samples were cut into 0.5 x 6 in. pieces and mounted on device holder with a double-sided tape. Each test liquid was deposited onto the substrate under ambient conditions. The evolution of contact angle changing with time was recorded and plotted as a curve of contact angle vs. time. The average contact angle at equilibrium was used for the surface energy estimations by Owens-Wendt (OW) (Owens, 1969) and Altay-Ma-Fleming (AMF) methods (Altay, 2020).

### 3. Result and discussion

#### 3.1. Structural and wettability properties

Grammage is one of the fundamental properties that describe the weight per unit area of papers. The weight was measured to be  $121 \pm 1$  gsm for the SB latex, and  $118 \pm 1$  gsm for the biobased latex samples. The difference between the samples was found significant ( $p:0.0002^*$ ). Bureau (1995) and Näätsaari (2006) studied that variation in grammage is usually caused by the difference in coat weight due to different rheological properties of the SB latex and biobased latex coating formulations. Lower grammage is reported to be advantageous for reduction in operational, production and transportation cost of final products (Venditti, 2012). Grammage also influences paper thickness (Scott, 1989). SB and biobased latex samples were measured to be  $110 \pm 2$   $\mu\text{m}$  and  $109 \pm 3$   $\mu\text{m}$ , respectively. The biolatex samples were thinner than the SB latex samples; however, they were virtually identical within statistical confidence limits ( $p:0.2232$ ).

SEM images were recorded from the samples at 8Kx and 40Kx magnifications present the microstructure of the coating surface in Fig. 2. The images depict the particle shapes of rhombohedral crystals of GCC and rhombo-hexagonal platelet clay that tend to be in irregular size fragments. The pigments fill the voids between the fibers and improve the smoothness of the sheet by reducing microdeviations within the base sheet surface. Smoothness, surface energy and porosity are important paper properties impacted by the type of binder and they effect ink penetration and print quality. The deviation in distribution of thickness and grammage could cause irregularities in surface roughness and smoothness, which is generally perceived as a deficient paper surface quality (Ek, 2009). The surface structure is highly determinative in the process of settlement and penetration of printing ink on the paper surface (Aydemir, 2014). Rougher surfaces affect transfer and spreading of inks and coatings onto the paper, causing, in turn, bad printing (Alam,

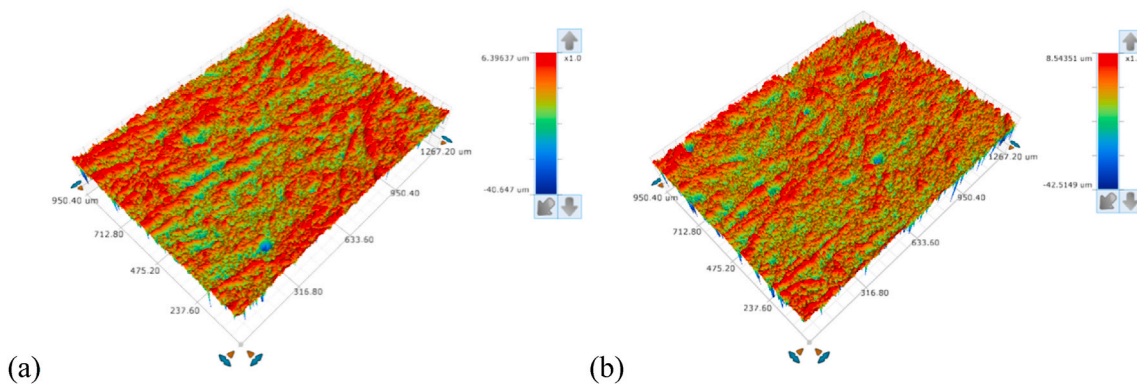


Fig. 3. 3D surface roughness interferometer image of paper samples: (a) SB latex binder, (b) biobased latex binder.

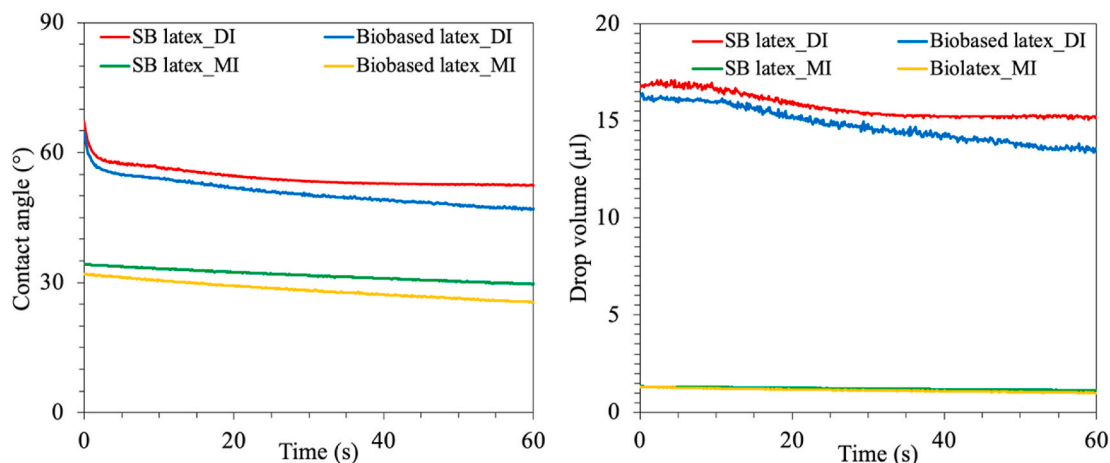


Fig. 4. Liquid contact angle and time-dependent volume change on paper surfaces.

**Table 4**  
Contact angles and surface energy values of liquids on paper samples prepared with SB latex and Biobased latex binders.

Liquid	Surface tension (mN/m)	Contact angle (°)	
		SB latex	Biobased latex
DI	72.14 ± 0.59	52.76 ± 0.16	47.19 ± 0.17
MI	48.36 ± 0.38	30.24 ± 1.00	26.28 ± 1.34

2012). Using the white light interferometer, the surface roughnesses of the samples was found to be  $1.44 \pm 0.05 \mu\text{m}$  for the SB latex and  $1.45 \pm 0.04 \mu\text{m}$  for the biolatex, that was statistically identical ( $p=0.4679$ ) (Fig. 3).

Paper is a porous medium contains voids, pores and open spaces. The resistance of paper surface to wetting by liquids have repercussions with respect to the strength of the paper and have an effect on the interactions between ink and paper (Pal, 2006). To study the liquid wetting and absorption behavior of the samples, contact angle of the drop was measured initially when the drop is placed on paper surface and 60 s later according to T-458 standard (Fig. 4). The DI and MI drops

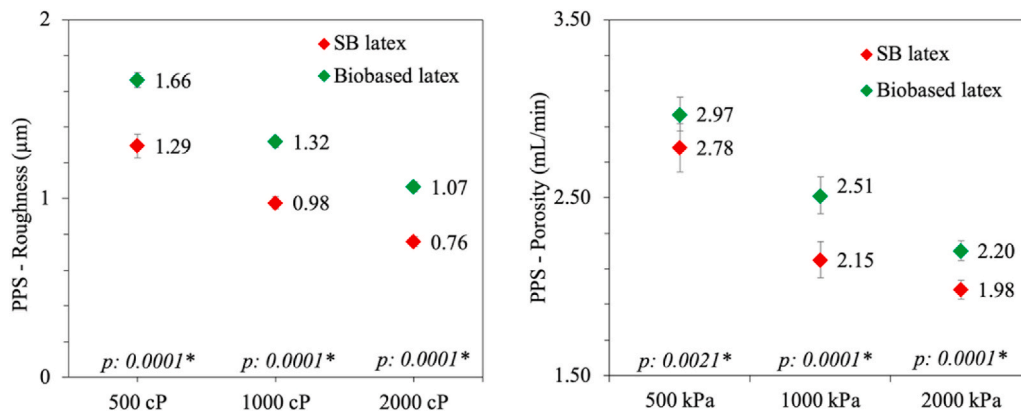


Fig. 5. Comparison of PPS roughness and porosity of paper samples at different clamping pressures.

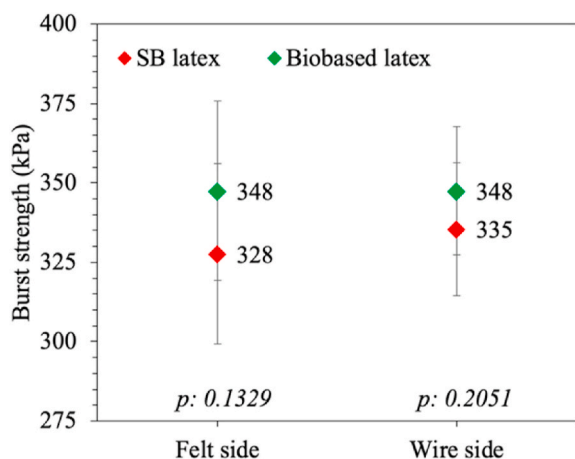


Fig. 6. Bursting strength of the paper samples.

presented smaller contact angles on the biolatex samples, indicating the biolatex binder improves the wettability of papers (Table 4, Fig. 4).

The total surface energies of the SB latex and biobased latex samples were estimated as 51 mN/m and 55 mN/m with the OW method and 58 mN/m and 61 mN/m with AMF method, respectively. In terms of liquid drop volume change, the drop volume on the SB latex samples decreased over time and started equilibrating at the 35 s, while equilibration started at 55 s for the biolatex samples, indicating that liquids spread and penetrate more into the biolatex samples than the SB latex samples. This can be interpreted as follows: a given volume of ink may penetrate more into the paper samples containing biobased binder than the paper samples containing SB latex binder, resulting in lower ink film thickness, lower print gloss and possibly lower color strength in printing. The higher PPS porosity value (Fig. 5) and different permeability coefficient value of the paper samples containing biolatex ( $1.34 \pm 0.06 \times 10^{-5} \mu\text{m}^2$ ) showed that the use of biolatex results in papers with a more porous structure relative to the paper containing SB latex binder ( $1.16 \pm 0.06 \times 10^{-5} \mu\text{m}^2$ ) ( $p=0.0001^*$ ). Thereby, the former would allow more ink penetration into the substrate (Gane, 2018).

PPS roughness tested at different clamping pressures in Fig. 6 shows the ability of paper to reduce its thickness when exposed to a compressing force, giving a measure of the paper sample compressibility. Higher smoothness and compressibility are conducive to high print quality images by enabling more complete contact between paper and printing plate or blanket (Wu, 2008). Compressibility is a function of paper density, degree of refining, calendering and supercalendering during papermaking. Increased moisture content also allows higher degree of compressibility. It is assumed that paper porosity (PPS, really a measure of permeability) decreases in a linear fashion when exposed to increasing compression (Hsu, 1961). The higher the compressibility of the paper samples containing biolatex is in line with its higher moisture content of 5%. The higher degree of correlation found in the biolatex paper samples, that is an indicator of better compressibility, and in turn, enables better print performance (Wu, 2008). The moisture content of the paper samples containing SB latex binder was 4.9%. Increased moisture is reported to have a positive impact on creasing applications (Coffin, 2017).

### 3.2. Mechanical properties

Burst strength shows the maximum hydrostatic pressure needed to burst the paper samples. The paper samples containing biolatex binder showed negligibly higher strength relative to the paper samples containing SB latex, since they were statistically identical within confidence levels because of the large variations of each (Fig. 6).

Tear strength shows how much force (tearing resistance) is needed to

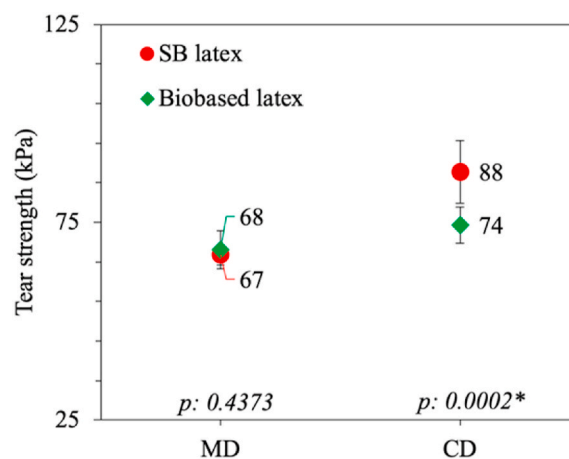


Fig. 7. Comparison of tear strength of paper samples.

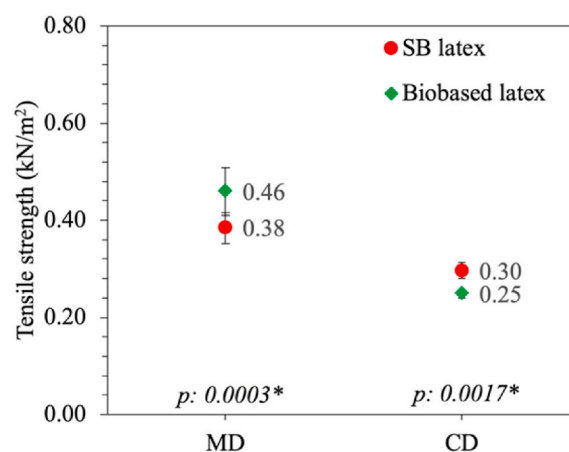


Fig. 8. Tensile strength of the paper samples.

tear the paper sample through a fixed distance after the tear has been initiated. The test is critical for converting applications as well as wall papers, kids' books or maps, where tear is usually started in actual use. Tearing resistance is affected by the paper direction, usually stronger in the CD (cross) direction than in the MD (machine) direction. The paper samples were tested and found statistically identical in terms of their tear strength in the MD direction ( $p:0.4373$ ) (Fig. 7). In the CD direction, the paper samples containing SB latex showed significantly higher tearing strength than the paper samples containing biolatex binder, 88 kPa and 74 kPa, respectively ( $p:0.0002^*$ ). Tear index was calculated to 0.62 for the SB latex-containing sample and 0.60 for the biolatex binder-containing samples.

Tensile strength is the paper's resistance to breaking under tension and measured by clamping a strip of sample with a specified width and length and applying force at a specified load rate. It is defined as the highest force that the sample can withstand before it ruptures. Fig. 8 shows the effect of the use of biolatex binder on the tensile strength. Within statistical confidence levels, a significant difference was found in the MD direction with biolatex containing samples, while higher strength in the CD direction was found with SB latex containing samples. Tensile strength is always higher in the MD direction than the CD due to the majority of paper fibers' strong bonding of their axes aligned toward the direction of forward movement on the papermaking machine (Scott, 1989). The reason for the higher tensile strength in MD direction for the biobased latex sample would be attributed to the high density of hydroxyl groups in the binder contributing the higher tensile strength (Zhu, 2018). Tensile energy absorption (TEA) shows an empirical

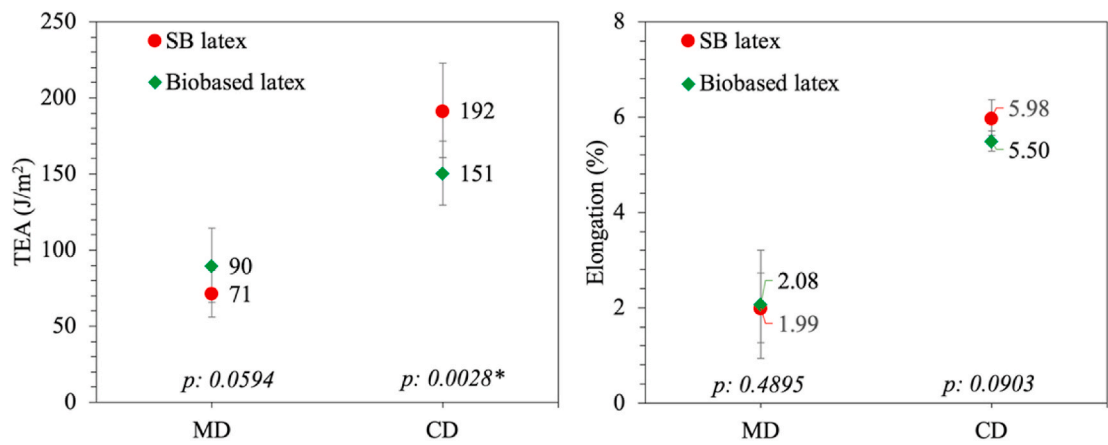


Fig. 9. Tensile energy absorption and elongation of the paper samples.

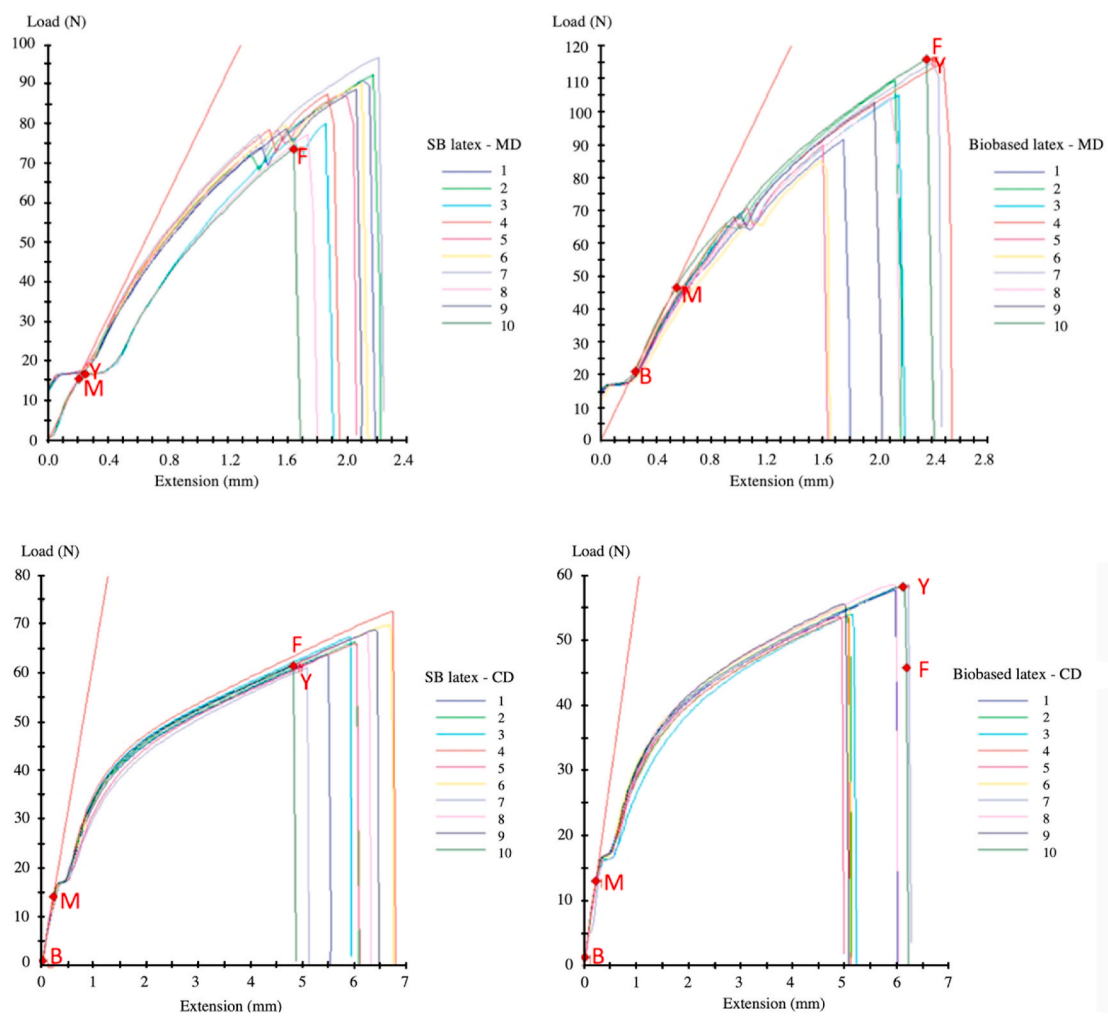


Fig. 10. Force-extension curves of paper samples on machine and cross directions.

relation between the surface strength of coated paper and the fracture energy in the plane of the coating layer (Morsy, 2004). It is critically important that the paper withstands, without cracking, the loads and deformations applied during printing, calendering and various converting processes. Fig. 9 shows that the paper samples containing biolatex binder has an equivalent TEA strength in MD direction and lower TEA strength in CD directions compared to the paper samples containing SB latex binder. Elongation properties of the paper samples presented in

Fig. 9 shows the samples have equivalent stretching properties, which is an indicator of paper ability to withstand web tensions that paper rolls experience during high-speed printing (Grenfell, 1964). Sababi indicated that general mechanical properties of the paper products such as mechanical strength can be evaluated with conventional mechanical tests like indentation, dynamic mechanical analysis, and tensile testing (Sababi, 2012). In Fig. 10, the tensile force-extension curves of the paper samples for ten different measurements represents the specific load in

the paper plane (Shiviyari, 2016). The area between B-M shows the slope of the linear part of the curve anywhere below the Elastic limit is equivalent to Young's Modulus of Elasticity. Y and F show the ultimate tensile strength and fracture points. Elastic moduli in the MD direction were measured to be 5.2 MPa for the biolatex containing samples and 4.8 MPa for the SB latex containing samples. The moduli in the CD direction were measured to be 3.5 MPa for the biolatex and 3.9 MPa for the SB latex samples. These results indicate that the biolatex binder improves elasticity in the MD direction, which is critical for creasing, folding and binding that requires applications in MD direction that have a larger window of acceptance (Coffin, 2017).

#### 4. Conclusion

Paper industry is the largest market that uses SB latex binder. In this research, replacing SB latex by an all-green starch-based biolatex nanoparticle binder was studied to understand the effect on structural and mechanical properties, liquid absorption, wettability and surface property of paper.

- Reduction in grammage is a highly-desired important property in the paper industry to decrease transportation costs for the final products and carbon footprint generation. Using biolatex nanoparticles was reduced the grammage of paper.
- The thickness and surface topography of the biobased latex samples were found to be equivalent to the SB latex samples.
- Using biolatex nanoparticles improved the liquid absorption and wettability properties.
- Notably, the strong binding strength across the interface of structured coated paper that contains the biolatex binder was enabled equivalent bursting, tear and tensile strength properties relative to the SB latex samples at 1:1 replacement ratio.
- The moisture content, PPS porosity, roughness and compressibility of the paper samples containing biolatex binder was higher than the paper samples containing SB latex.

This research concluded that the bulk mechanical properties of the samples were similar and SB latex binder can be replaced with a starch-based biolatex binder at 1:1 ratio without compromising mechanical and structural properties. In future studies, investigating the effect of replacing traditional starch binder in coating formulation on paper properties is recommended to provide additional reduction in heat-trapping greenhouse gasses, energy consumption and cost by eliminating starch cooking procedure.

#### Availability of data and materials

The datasets analyzed during the current study are available from the corresponding author on reasonable request.

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#### Authors' contributions

BNA designed the research, analyzed the data and wrote the manuscript. TC and AF contributed in part writing the manuscript. CK, PDF, CA and KA reviewed and edited the manuscript. All authors have approved the manuscript.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

the work reported in this paper.

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