ISSN: 0379-5462
IPPTA: Quarterly Journal of Indian Pulp and Paper Technical Association
Vol. 38, E2, 2026, p. 158-162

IPPTA

Modified cellulose-oxidized starch formulation for surface sizing: An innovative approach for high performance cultural paper

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Abstract: *The present study introduces a novel and sustainable approach for the surface sizing of writing and printing grade paper through the development of a blended oxidized starch and modified cellulose formulation as an alternative to conventional oxidized starch based surface sizing systems. In this work, cellulose was modified through different chemical and mechanical treatments by which the hydroxyl groups of cellulose were partially transformed into functional groups such as aldehyde and carboxylic acid groups. The modified cellulose was subsequently blended with oxidized starch and applied on paper surfaces through a size press.*

The blended oxidized starch and modified cellulose formulation exhibited outstanding improvements in the strength, surface, and barrier properties of paper compared to both unsized paper and paper surface sized with conventional oxidized starch alone. The treated paper showed an overall 36–67% enhancement in strength properties. The resistance to air permeance increased approximately by 500%, indicating the formation of a denser and less porous paper structure. Surface morphological analysis using scanning electron microscopy (SEM) further confirmed the development of a smoother, more compact, and uniformly coated surface in papers treated with the oxidized starch and modified cellulose blend, making the formulation highly suitable for high-performance writing and printing grade papers with improved smoothness and surface texture.

The findings clearly establish the advantage of oxidized starch and modified cellulose surface sizing system as a cost-effective, environmentally sustainable, and high-performance alternative to conventional oxidized starch based surface sizing formulations. The superior performance achieved at low coating dosage highlights its strong potential for industrial application in the manufacturing of advanced writing and printing papers with improved strength, surface, and barrier properties.

Keywords: *Oxidized starch; Modified cellulose; Surface sizing; Writing and printing grade paper; Paper properties.*

1. Introduction

Surface sizing is a very important step in the paper making process to improve the surface properties and performance of paper in general (1). In this process, a sizing solution is applied on the surface of the dried paper sheet with the help of a size press. The applied material enters the pores present between fibers and forms a thin layer on the paper surface. The surface sizing process improves different paper properties like strength, stiffness, printability, and resistance to water penetration to a considerable extent. The effectiveness of surface sizing mainly depends on factors like viscosity of the solution, pickup of the sizing material, machine speed and the type of additives used (2,3).

Starch is the most commonly used material for surface sizing due to its easy availability, low cost, biodegradability and good compatibility with cellulose fibers (4). However, in paper industries, oxidized starch is generally preferred over native starch because of its higher solubility and better performance in application (5). The modified functional groups in the starch molecules improve film formation and better bonding with the cellulosic fibers (6). Therefore, oxidized starch is more effective in enhancing paper strength, surface smoothness, ink holdout and print quality.

In addition to oxidized starch, many other natural polymers are explored for paper surface applications currently due to increased environmental and sustainability concerns (7). Natural polymers such as cellulose derivatives, chitosan, guar gum, sodium alginate and gelatin have shown good potential of improving the fiber-fiber bonding and various properties of paper. These bio-based materials are biodegradable in nature and can contribute in reducing the use of synthetic chemicals in papermaking process (8). Hence, the use of oxidized starch with additives based on natural polymers can be considered as a promising approach for the development of environmentally friendly and high-performance paper products (7).

Cellulose is the main structural component of plant fibers. It is one of the most commonly found natural polymers in nature (9). Cellulose plays an important role in paper industry as the strength and quality of paper depends mainly on bonding ability of cellulosic fibers. The numerous hydroxy groups in cellulose molecules lead to the formation of hydrogen bonding between fibers, which add to the mechanical strength of paper (10). Its biodegradability and good film-forming properties have attracted much attention of paper makers for developing sustainable paper additives for wet-end and surface sizing application (9).

In recent years, researchers have focused on modifying cellulose to improve its performance for different industrial applications, including papermaking (10). Modification of cellulose can be carried out through physical, chemical, or mechanical treatments in order to alter its structure and surface characteristics (10, 11). Modified cellulose materials generally show better dispersibility, higher surface area, improved bonding ability, and enhanced interaction with fibers as compared to native cellulose. Because of these advantages, modified cellulose has been explored for improving paper strength, barrier properties, surface smoothness, and printability (10).

The novelty of the present work lies in the development of a bio-based surface sizing formulation comprising oxidized starch and chemically modified cellulose produced through sequential oxidation and microfluidization treatments. Unlike conventional surface sizing systems that rely solely on oxidized starch, the proposed formulation utilizes modified cellulose containing aldehyde and carboxyl functional groups, which enhances inter-fiber bonding and film formation on the paper surface. The study demonstrates that the oxidized starch–modified cellulose blend can achieve significantly higher strength and barrier properties at a lower coating dosage than conventional oxidized starch sizing. This synergistic effect between oxidized starch and modified cellulose has not been extensively reported for writing and printing grade papers and provides a sustainable approach for producing high-performance cultural papers.

2. Materials and Methods

2.1 Materials

In the present study, surface sizing was performed on handsheets prepared from a pulp blend consisting of 80% agro-based pulp and 20% hardwood pulp. The blended pulp and imported softwood sheets was obtained from an integrated paper mill located in northern India. Laboratory-prepared modified cellulose (MC), derived from bleached softwood pulp, was used as a surface-sizing additive.. The softwood pulp underwent sequential chemical oxidation and mechanical processing to obtain the modified cellulose employed in the surface-sizing experiments.

2.2 Preparation of Modified Cellulose (MC)

The bleached softwood pulp used for the preparation of modified cellulose exhibited a freeness of approximately 840 ± 15 CSF indicating that pulp was completely unbeaten. The preparation procedure is presented in Figure 1.

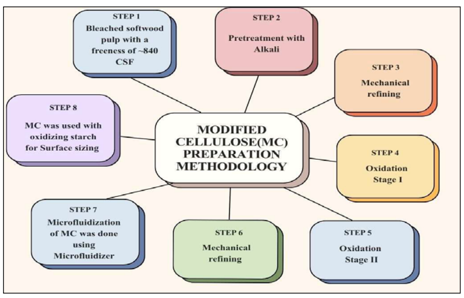

Figure 1: The preparation procedure of modified cellulose for surface sizing application

Pulp oxidation was conducted in two sequential stages (Stage I and Stage II). During this process, hydroxyl groups were oxidized first to aldehyde groups and subsequently to carboxyl groups. To enhance its suitability for surface-sizing applications, the modified cellulose was further processed by microfluidization. This treatment resulted in the formation of a highly viscous, gel-like cellulose dispersion.

2.3 Preparation of oxidized starch and blend of oxidized starch with modified cellulose

The oxidized starch (OS) used in the present study was first tested for moisture content by oven drying at 105 ± 2°C. An 8% (w/v) OS solution was then prepared by dissolving 40 g (oven-dry basis) of OS in 500 mL of distilled water. The solution was cooked in a water bath at 95 ± 5°C for 30 ± 3 min under continuous stirring. The prepared OS solution was subsequently blended with modified cellulose (MC) using a microfluidizer operated at a pressure of 30,000 psi for two cycles. The total solids content of the OS solution, MC suspension, and the blended OS/MC solution was determined by oven drying at 105 ± 2°C for 24 h. The viscosity of the samples was measured using an Anton Paar MCR 102 rotational rheometer. The charge demand of all samples was determined using an AFG Analytic GmbH CAS Charge Analyzing System.

2.4 Handsheet Preparation, Surface sizing and Paper Properties

The handsheets were prepared from a blended pulp consisting of 80% agro-based pulp and 20% hardwood pulp. Handsheets with a basis weight of 100 g/m² were produced using a laboratory handsheet former in accordance with TAPPI T 205 sp-02. Prior to handsheet preparation, the pulp was refined using a PFI mill to achieve a Canadian Standard Freeness (CSF) of 400 mL. The surface-sizing solutions were maintained at 60 ± 2°C before application. Surface sizing was carried out using an Automatic Film Applicator (ZAA 2300) equipped with a 36 µm profile rod. The increase in the grammage of the sheets after surface sizing, compared with the unsized sheets, was considered the coat weight imparted by the surface-sizing treatment. The handsheets, both with and without surface sizing, were conditioned at 27 ± 1°C and 65 ± 2% relative humidity for 24 h prior to testing.

Various physical and mechanical properties of the handsheets were then evaluated as per standard test methods: Grammage IS - 1060 (Part 5/Sec 5), Burst IS - 1060 (Part 6/Sec 2), Tensile strength, tensile energy absorption (TEA), elongation at break, and tensile stiffness - IS 1060 (Part 5/Sec 6) and Gurley porosity - IS 1060 (Part 5/Sec 11). The morphological characteristics of the handsheets, both with and without surface sizing, were examined using a scanning electron microscope (SEM). The instrument used for the analysis was a TESCAN MIRA 3 LMH (USA).

3. Results and discussion

This study was conducted using unsurface-sized handsheets with a basis weight of 100 ± 2 g/m², prepared from a blended agro residue and hardwood pulp. The objective was to evaluate the surface sizing potential of blended solutions of oxidized starch and modified cellulose and their effects on various paper properties. The effectiveness of blend was compared with starch surface sized sheet as starch is currently the most widely used surface sizing agent in the global paper industry, particularly in India.

3.1 Characteristics of oxidized starch, modified cellulose and blended solution of oxidized starch and modified cellulose

The characterization results for oxidized starch, modified cellulose, and their blends for different parameters are presented in Table 1. As shown in Table1, the viscosity of modified cellulose solution at a concentration below 0.5% was comparable to that of 8% oxidized starch solution. Streaming potential measurements indicated that both oxidized starch and modified cellulose possess an anionic charge. Consequently, the charge demand of oxidized starch, modified cellulose, and their blends was found to be cationic.

Table 1. Properties of oxidized starch, modified cellulose and blended solution of oxidized starch and modified cellulose

Parameters	Oxidized starch	Solution of		
		Modified cellulose	Oxidized starch	Blend of oxidized starch and modified cellulose`
Moisture Content, %	10.2±0.55	-	-	-
Ash content, %	0.50±0.05	-	-	-
Total solids, %	89.2±0.45	0.35±0.01	8.3±0.2	0.50±0.02
Viscosity, Cp @ 25°C and Shear rate of 100 s^{-1}	-	37.6±2	36.9±2	45±4
Streaming Potential, mV	-	-90±5	-116±9	-120±8
Charge demand, µeq/l	-	25±3	352±10	400±11

3.2 Effect of surface sizing on various properties of paper

The effects of surface sizing on various strength properties like tensile strength, burst factor, elongation, TEA index and porosity were investigated. The effect of surface sizing on the tensile strength of the handsheets is presented in Figure 2. The unsized handsheets exhibited a tensile strength of 3.45 kN/m. Surface sizing with oxidized starch at a coating dose of 2.5 g/m² increased the tensile strength by 16.2% compared with the unsized handsheets. These findings are consistent with previous studies that reported improvements in tensile strength following surface sizing with oxidized starch (3, 12, 13). Surface sizing with a blended solution of oxidized starch and modified cellulose at a coating dose of 1.5 g/m² improved the tensile strength of the unsized paper by 17.5% as compared to the unsized handsheets. These results indicates that surface sizing with oxidized starch alone at a dose of 2.5 g/m² and surface sizing with a blended solution of oxidized starch/ modified cellulose at a lower dose of 1.5 g/m² showed almost similar improvement in tensile strength.

As the surface sizing dose of the oxidized starch/ modified cellulose blend was increased to 1.8 g/m², the tensile strength improved by 41.1% and 21.4% compared with the unsized handsheets and the oxidized starch sized handsheets, respectively as shown in Figure 2. This substantial enhancement may be attributed to the presence of modified cellulose, which likely increased the available surface area and site for bonding, while its functional groups promoted the formation of additional hydrogen bonding between fibers.

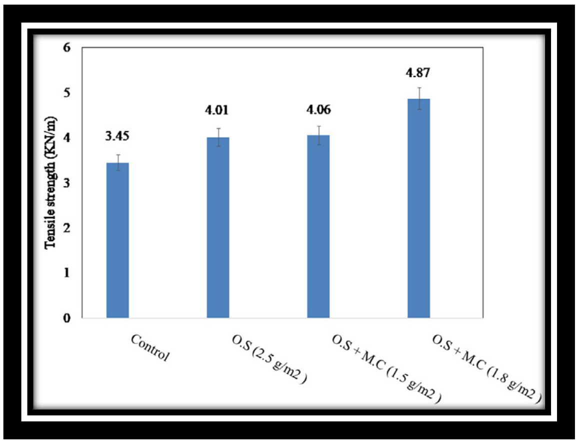

Figure 2: Effect of surface sizing on tensile strength of the paper

Figure 3 illustrates the influence of surface sizing on the burst factor of the handsheets. The unsized handsheets exhibited a burst factor of 23.7. The burst factor increased significantly following surface-sizing treatment. Oxidized starch applied at a surface sizing dose of 2.5 g/m² resulted in a 34.5% improvement over the unsized handsheets. The improvements in burst strength using oxidized starch as surface sizing chemical was earlier reported by various researchers (14,15). Interestingly, a blend of oxidized starch/ modified cellulose applied at only 1.5 g/m² produced a comparable increase of 36.3%. This suggests that the addition of modified cellulose enhances the efficiency of the surface sizing formulation, allowing similar strength development at a lower coating dose. A further increase in the coating dose of the oxidized starch/modified cellulose blend to 1.8 g/m², the burst factor increased by 63.7% relative to the unsized handsheets and by 21.6% compared with the handsheets surface-sized using oxidized starch alone, as shown in Figure 3.

The enhancement in burst factor can be attributed to the penetration of the low viscosity surface sizing solution into the porous paper structure, where it forms a sub polymeric network within the fiber matrix. At the same time, a portion of the sizing material remains on the paper surface, creating a continuous film layer. These combined effects strengthen the fiber network by increasing the extent of hydrogen bonding between the cellulose fibers and the sizing components. Moreover, the presence of modified cellulose introduces additional functional groups that may facilitate both hydrogen and covalent bond formation with the cellulosic fibers. As a result, improved inter-fiber bonding, pore filling, and surface film formation collectively contribute to the observed increase in burst factor and other strength-related properties of the paper (12, 16).

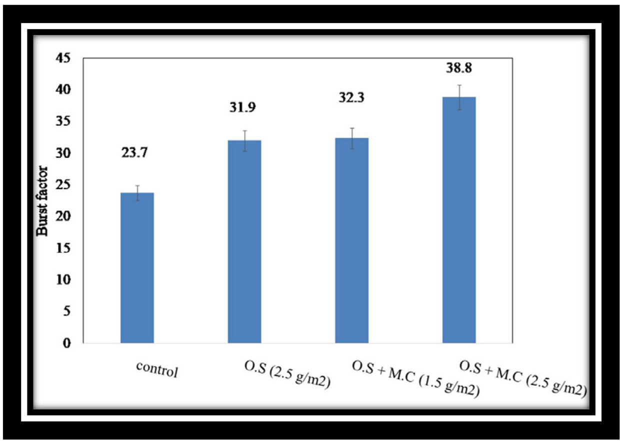

Figure 3: Effect of surface sizing on burst factor of the paper

The effects of surface sizing on the elongation and tensile energy absorption (TEA) index of the handsheets showed in Figures 4 and 5, respectively. The unsized handsheets exhibited an elongation of 3.3 mm and TEA index of 840 J/kg. Surface sizing with oxidized starch at a coating dose of 2.5 g/m² increased the elongation and TEA index by 67.2% and 58.5%, respectively, compared with the unsized handsheets. Surface sizing with an oxidized starch/ modified cellulose blend further enhanced both properties. At a coating dose of 1.8 g/m², the elongation and TEA index increased by 21.5% as compared to the handsheets surface sized with oxidized starch alone, as shown in Figures 4 and 5.

The observed improvements in elongation and TEA index may be attributed to enhanced inter-fiber bonding resulting from the penetration of the surface sizing solution into the paper structure and the formation of a continuous film on the paper surface. The presence of modified cellulose likely promotes additional hydrogen bonding interactions, thereby improving the paper's ability to absorb and dissipate energy before failure.

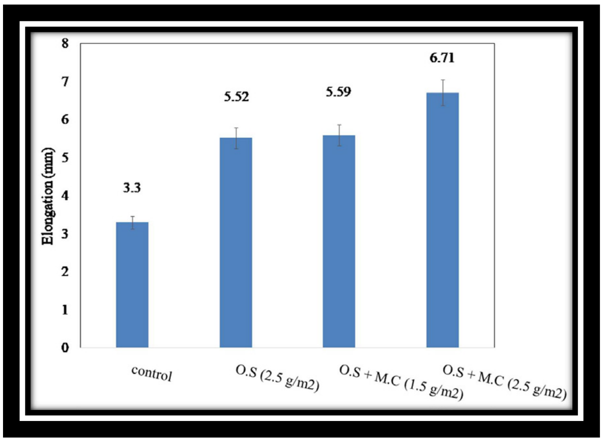

Figure 4 Effect of surface sizing on elongation of the paper

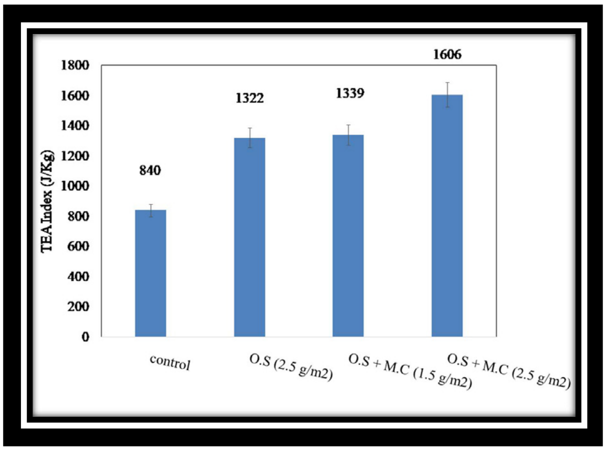

Figure 5 Effect of surface sizing on TEA Index of the paper

The effect of surface sizing on the porosity of the handsheets is shown in Figure 6. The control handsheets exhibit high porosity due to the voids in the fiber network. The SEM images shows that after surface sizing by both surface sizing solutions the voids area of paper is reduced as shown in Figure 7. Surface sizing with oxidized starch fill the pores of the surface and allows the fibers to bond more, reducing porosity. The porosity is further reduced by adding modified cellulose to the oxidized starch. The blended solution of oxidized starch and modified cellulose forms the denser structure and fills the pores which further makes the fiber network stronger through hydrogen bonding.

4.0 Potential industrial application of the present study

The developed oxidized starch/ modified cellulose formulation has strong potential for industrial implementation in paper mills manufacturing writing, printing, and other cultural paper grades. The formulation can be applied through existing size-press systems without major process modifications, making its adoption economically feasible. The enhanced tensile strength, burst strength, energy absorption, and reduced porosity achieved at lower coating dosages can help mills reduce starch consumption while improving paper quality and printability. Furthermore, as both oxidized starch and modified cellulose are renewable and biodegradable materials, the technology supports sustainable papermaking practices and can contribute to reducing dependence on synthetic surface treatment chemicals.

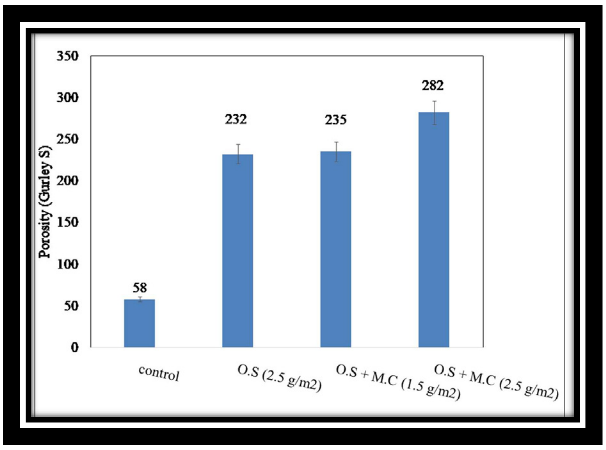

Figure 6 Effect of surface sizing on porosity of the paper

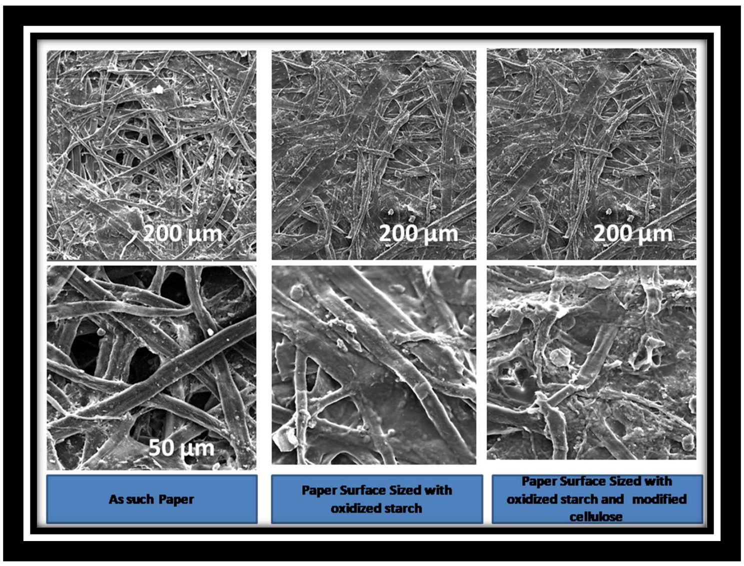

Figure 7 SEM images of Paper Handsheets

Conclusion

The results demonstrate that surface sizing significantly improves the mechanical and structural properties of paper. While surface sizing with oxidized starch alone enhanced tensile strength, burst factor, elongation, TEA index, and reduced porosity, the incorporation of modified cellulose into the oxidized starch formulation produced even greater improvements, particularly at a coating dose of 1.8 g/m². The oxidized starch/ modified cellulose blend increased tensile strength by 41.1%, burst factor by 63.7%, and further improved elongation and TEA index compared with oxidized starch alone, while also reducing paper porosity by improving the pore filling and densification of the fiber network. These improvements are attributed to the formation of a continuous surface film, increased fiber-fiber bonding, and the additional hydrogen bonding interactions provided by the functional groups of modified cellulose. The findings indicate that modified cellulose is an effective co-binder for surface sizing formulations and can achieve superior paper strength and barrier properties at lower coating dosages than conventional oxidized starch sizing.

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