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Performance Optimization Of Hardwood–Wheat Straw Paper Through Modified Melia Dubia Microfibrillated Cellulose And Calcium Carbonate Integration

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Abstract
A Novel Composite Papermaking Filler Was Developed By Combining Cellulose Microfibrils (Cmf) Derived From Melia Dubia, *Ground Calcium Carbonate (Gcc), And Cationic Starch. This Study Utilised Cmf In Two Distinct Ways: As A Gcc–Cmf Composite Filler Added During Sheet Formation And As A Papermaking Additive Mixed With The Pulp Suspension. The Impacts Of These Approaches On Filler Flocculation, Retention, And Paper Properties Were Studied In Hardwood And Wheat Straw Pulps. Paper Sheets Filled With The Gcc–Cmf Composite Showed Markedly Higher Filler Retention (~90%) Than Sheets With Conventional Gcc Alone. Sheets Containing The Composite Filler Also Exhibited Superior Tensile And Burst Indices Compared To Those With Standard Gcc Loading, Indicating Improved Strength At Equivalent Filler Contents. However, Using Cmf As A Direct Additive (Without Pre-Compositing With Gcc) Led To Somewhat Lower Enhancements In Strength And Retention. Composite Filler Sheets Became Denser As Filler Level Increased, Yet Maintained Optical Properties, Showing Slightly Higher Opacity And Comparable Brightness Relative To Conventional Filler Sheets. These Findings Demonstrate That Incorporating* Melia Dubia *Cmf In A Composite Filler Format Can Enable Higher Filler Usage In Paper Without Sacrificing And Even While Improving Overall Paper Strength And Quality.*
Keywords: Melia Dubia; *Cellulose Microfibrils; Composite Filler; Filler Retention; Paper Strength; Optical Properties*

INTRODUCTION

Paper Is A Cellulose Fiber Based Composite That Typically Includes Mineral Fillers, Pigments, Sizing Agents, And Other Additives. Mineral Fillers Such As Kaolin Clay Or Calcium Carbonate Are Cost-Effective And Improve Certain Properties Like Opacity And Smoothness, But They Inherently Do Not Bond With Fibers And Therefore Tend To Weaken Paper Strength By Interrupting Fiber–Fiber Contacts[1]. To Mitigate This, Retention Aids (Synthetic Polyelectrolytes) Are Widely Used In Papermaking To Flocculate Fine Particles And Fillers To Improve Retention On The Paper Machine Wire And Minimizing Losses. In Recent Years, There Is Growing Interest In Using Renewable, Biodegradable Carbohydrate Polymers – Such As Starch, Cellulose Derivatives, And Chitosan – To Modify Fillers And Enhance Their Compatibility With Fiber Networks. By Coating Or Compositing Mineral Filler Particles With Such Biopolymers, It Is Possible Impart Functional Surface Groups That Promote Fiber Bonding And Improve Paper Strength Properties. While Still Benefiting From The Filler's Cost And Optical Advantages[2]. For Example, [3] Reported That Starch And Carboxymethyl Cellulose Treatments Can Significantly Increase Filler Retention And Paper Strength By Forming A Flexible Coating On Filler Particles That Can Bond With Fibers. These Bio-Modified Fillers Due To Enhanced Hydroxyl Functionality Of Carbohydrate Polymers To Form Hydrogen Bonds With Cellulosic Fibers, Acting As Bridges Between Filler And Fiber. The Approach Is Economically Attractive And Eco-Friendly Since It Uses Inexpensive, Biobased Additives To Achieve Better Filler–Fiber Integration And Potentially Allows Higher Filler Loading In Paper.

Cellulose Microfibrils, Also Known As Microfibrillated Cellulose (Mfc) Or Nanofibrillated Cellulose (Nfc) Depending On Dimensions, Have Emerged As A Promising Biopolymer Additive In Papermaking. Cellulose Microfibrils Are The Sub-Structural Fibers Obtained By Disintegrating Cellulose Pulp Fibers Into Their Nano- And Microscale Fibrillar Components[4]. They Possess An Extremely High Aspect Ratio, Large Specific Surface Area, And A Wealth Of Hydroxyl Groups On Their Surface. These Attributes Give Cmfs Outstanding Bonding Ability, Making Them Effective As Strengthening Agents And Retention Aids In Fiber Networks. Several Studies Have Shown That Adding A Small Fraction Of

Mfc/Nfc (A Few Percent Relative To Fiber) Can Greatly Increase Paper Strength Properties Such As Tensile Index, Without Significantly Affecting Light Scattering Or Opacity. [5]Observed A Significant Increase In Tensile Strength With Just 4% Mfc Added To Thermomechanical Pulp, And [6] Reported That Introducing Mfc Into Clay-Filled Sheets Improved Both Strength And Optical Scattering Simultaneously [7]. The Enhancement Is Attributed To Microfibrils Bridging The Gaps Between Fibers And Fillers – Similar To An External Fibrillation Effect Thereby Reinforcing The Paper Structure. Moreover, [8] Demonstrated That Tempo-Oxidized Nanofibrillated Cellulose Could Flocculate Gcc Filler And Achieve Filler Retention As High As 85–90%. The Flocculation Mechanism Of Cmfs Is Described As A Hybrid Of Bridging And Patch Flocculation: The Fibrils Can Form Networks That Enmesh Filler Particles (Bridging), And Their Charged Sites Can Also Induce Flocs By Electrostatic Patch Attraction.

Inspired By These Developments, The Present Work[9] Explores The Use Of Microfibrillated Cellulose From Melia Dubia (Malabar Neem) As A Means To Improve Filler Retention And Paper Strength. Melia Dubia Is A Fast-Growing Hardwood Species; Its Fibers Were Used Here To Produce Cmf Due To Their Availability And Favorable Fibrillation Characteristics. We Incorporate The Cmf In Two Ways: As A Gcc–Cmf Composite Filler, Where Gcc Particles Are Pre-Flocculated And Encapsulated With Cmf (Using Cationic Starch As A Binding Agent), And, For Comparison, As A Cmf Additive Added Directly To The Pulp Furnish Along With Conventional Gcc[10].

We Hypothesize That The Composite Filler Will Behave More Like Fiber Fines, Being Retained More Efficiently In The Sheet And Contributing To Inter-Fiber Bonding. By Contrast, Adding Cmf Separately (Not Pre-Attached To Filler) May Yield Less Retention Benefit, Since Some Cmf May Adsorb Onto Fiber Surfaces Or Be Lost Rather Than Binding Fillers.

1.1. Objectives

The Objectives Of This Study Are

- To Clarify How The Gcc–Cmf Composite Filler Affects Filler Retention, And To Evaluate The Mechanical (Tensile, Burst, Tear) And Optical (Opacity, Brightness) Properties Of Paper Sheets Produced With This Composite Filler, In Comparison To Sheets Made With Traditional Filler (With Or Without Cmf Additive).
- To Examine The Paper Structure Using Microscopy To Understand The Distribution Of Cmf And Filler In The Sheets.
- To Leverage A Renewable Cellulose Resource In Filler Loading, This Work Aims To Enable Higher Filler Usage In Paper Without Degrading And Possibly While Enhancing End-Use Properties..

METHODOLOGY

1.2. Production Of Microfibrillated Cellulose (Cmf)

Cellulose Microfibrils Were Produced From The Melia Dubia Pulp Fibers Using High-Pressure Microfluidization. The Melia Dubia Pulp (Bleached) Was Diluted To ~1% Consistency (W/V) In Water And Passed Through A Lab-Scale Microfluidiser (Model Lm-20, Microfluidics Corp., Usa) For Ten Passes. This Mechanical Treatment Fibrillated The Fibers Into Microfibrils. The Output Was A Translucent Gel-Like Suspension Of Cellulose Microfibrils With A Solids Content Of ~0.4% (W/W). No Chemical Or Enzymatic Pretreatment Was Applied Prior To Mechanical Fibrillation In This Study. To Ensure Better Dispersion Of The Cmf And To Break Any Agglomerates, The Cmf Gel Was Gently Ultrasonicated After Microfluidization. For Ultrasonication, The Cmf Suspension Was Diluted To 0.5% And Sonicated Using A Probe Sonicator (Hielscher Up400s, 400 W, 24 Khz, 22 Mm Titanium Probe) For 20 Minutes. The Sonication Was Performed In An Ice-Water Bath To Prevent Overheating; The Suspension Temperature Reached About 50 °C By The End Of The Treatment. The Resulting Melia Dubia Cmf Had A High Degree Of Fibrillation And Was Used Immediately In The Procedures Described Below.

1.3. Preparation Of Gcc–Cmf Composite Filler

The Composite Filler Was Prepared By Sequentially Combining Gcc, Cmf, Cationic Starch, And Retention Aid Under Controlled Mixing Conditions. First, An Aliquot Of The 0.5% Cmf Suspension Was Added To A 5% Gcc Slurry (Targeting A Ratio Corresponding To A Specified Cmf Percentage On Dry Pulp Weight). The Mixture Was Stirred At 200 Rpm For 1 Minute To Allow The Fibrils To Distribute And Attach To Gcc Particle Surfaces. In Separate Trials, Three Levels Of Cmf Were Added To The Filler: 1%, 2.5%, And 4% Of The Oven-Dry Fiber Mass (These Percentages Represent The Cmf

Dosage Relative To Dry Pulp, Equivalent To ~3.3%, 8.3%, 13.3% Relative To Filler Mass Since Filler Was ~30% Of Pulp By Mass). After Cmf And Gcc Were Mixed, Pre-Cooked Cationic Starch Solution Was Added To The Suspension (At 2.5% Weight On Gcc). The Addition Of Cationic Starch Immediately Induced Visible Flocculation Of The Gcc–Cmf Particles, Forming A Composite In Which Gcc Particles Were Bound Together By The Starch And Cmf. This Mixture Was Stirred For Another Minute At 200 Rpm To Ensure Uniform Distribution Of Starch And To Complete The Formation Of The Composite Filler. Finally, Just Before Sheet Formation, The Anionic Pam Retention Aid Was Added At 0.1% On Pulp, And The Suspension Was Gently Agitated For 30–60 Seconds.

The Resulting Gcc–Cmf Composite Filler Consisted Of Gcc Particles Enmeshed In A Network Of Cellulose Microfibrils, With Cationic Starch Acting As A Glue To Cement The Structure (Schematically Illustrated In Figure 1). The Composite Filler Flocs Were Notably Larger Than Individual Gcc Particles And Had A Fibrous Coating, As Later Observed Under Sem. For Comparison Tests, A Conventional Filler Preparation (Gcc With Retention Aid But No Cmf Or Starch) And A Cmf Additive Case (Gcc With A Small Amount Of Cmf Added Separately To Pulp, Rather Than Pre-Composited) Were Also Prepared. In The Cmf Additive Scenario, 2% Cmf (On Pulp) Was Mixed Into The Fiber Slurry Along With Gcc Just Prior To Sheet Formation, Simulating The Use Of Cmf Purely As An Additive Without Preforming A Composite Filler.

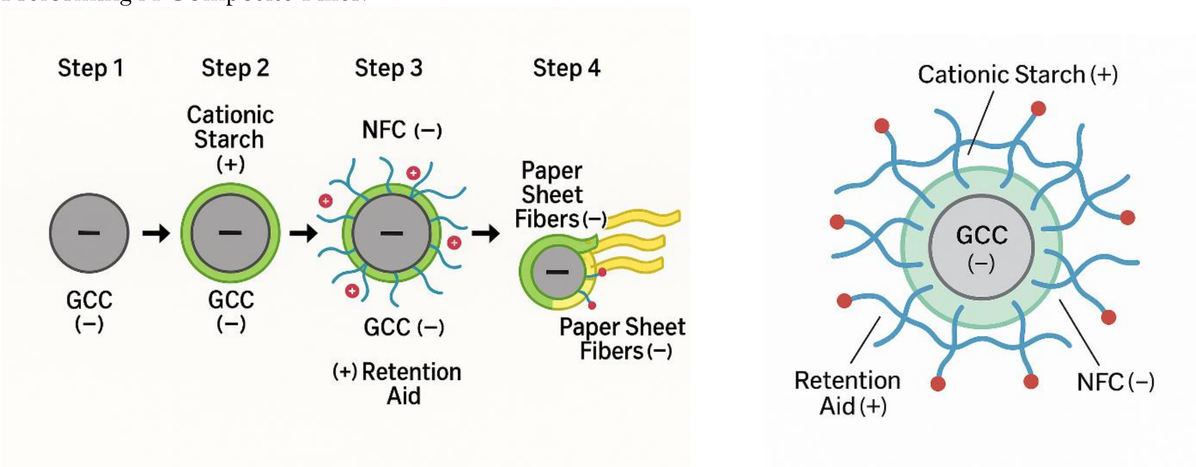


Figure 1. Diagrammatic Representation Of Gcc-Mfc/Nfc Composite Filler

We Used A Filler Called Gcc (Ground Calcium Carbonate), Cationic Starch, Retention Aid, And Cellulose Microfibrils During This Procedure. First, The Cmf Gel Was Diluted In Water To A Concentration Of 0.5 Per Cent. Then, It Was Put Through A Gentle Ultrasonication Process For Two Minutes In Order To Deagglomerate The Fibres Using A Laboratory Ultrasonicator (Up400s Hielsher Ultrasonics Gmbh Germany, 24 Khz) Fitted With A Titanium Alloy Sonotrode That Has A 22 Mm Diameter And Measures In At 24 Khz. The Sonication Procedure Was Conducted For Twenty Minutes With A Power Output Of Four Hundred Watts. The Optimization Of This Minimal Power Demand Was Accomplished By Conducting Several Experiments And Examining The Sample. The Procedure Was Carried Out In Freezing Water So The Sample Would Not Get Overheated. The Sample Was Measured To Have A Temperature Of Around 50 Degrees Celsius After The Completed Ultrasonic Treatment.

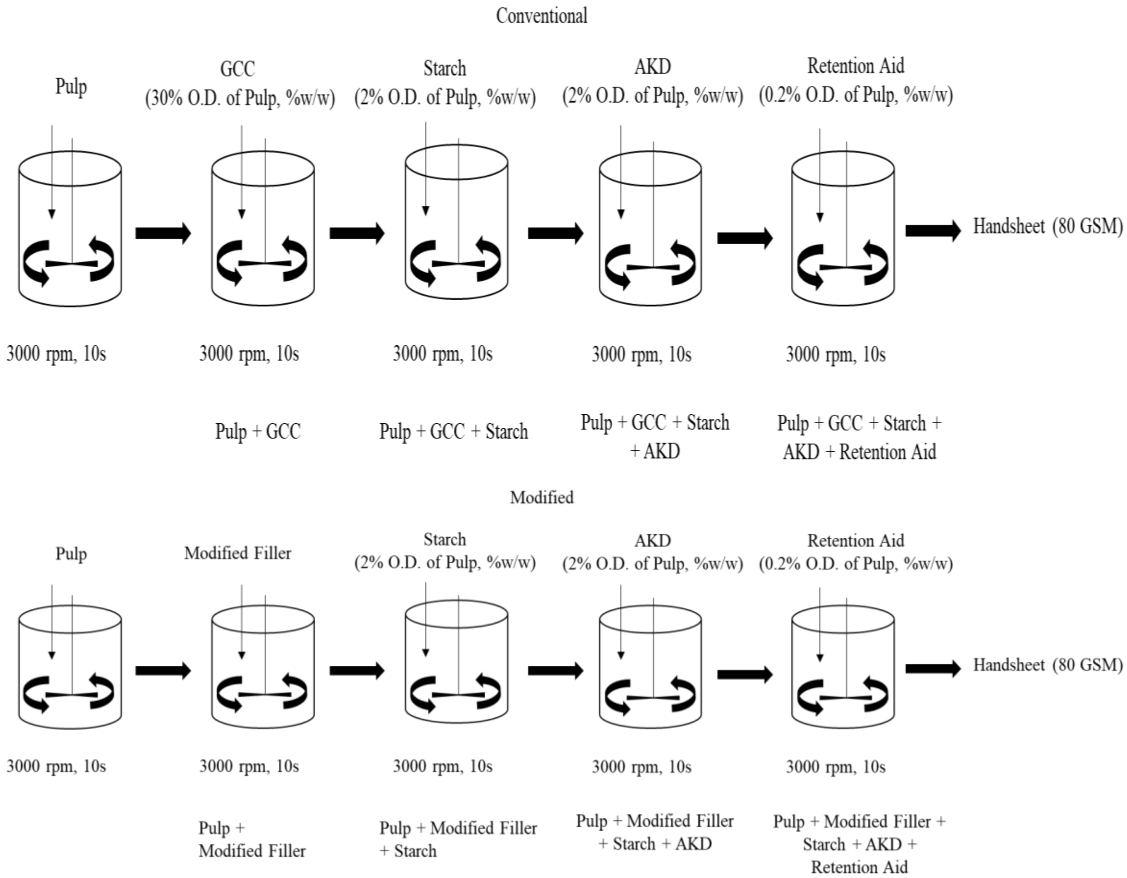
The Gcc Solution Used 30 Per Cent O.D. Of Pulp, Per Cent W/W, And A Concentration Of 5 Per Cent Gcc In Water, Per Cent W/V. A Laboratory Stirrer Was Used To Mix The Solution At 200 Revolutions Per Minute For One Minute (Borosil-Mhps550). The Diluted Cmf Suspension Was Then Combined With The Gcc Filler Suspension, And The Mixture Was Swirled At 200 Revolutions Per Minute For One Minute. The Amount Of Gcc (Ground Calcium Carbonate) Filler Utilized In Producing Handsheets Was Equal To Thirty Per Cent By The Optical Density Of The Pulp And Thirty Per Cent By Weight. The Amount Of Cmf Added To The Gcc Filler Was One Per Cent, Two And A Half Per Cent, And Four Per Cent Of The Overall Dry Weight Of The Pulp, Respectively. A Transparent And Viscous Starch Solution Was Prepared By Cooking Cationic Starch At A Concentration Of 2 Grams Per Litre In Water At A Temperature Of 90 Degrees Celsius For Thirty Minutes. After That, The Starch Solution That Had Been Boiled Was Added To The Solution That Already Included Gcc And Cmf. The Amount Of Added Cationic Starch Was 2.5 Per Cent O.D. Of Gcc, Which Equates To Per Cent W/W, And The Mixture

Was Agitated At A Speed Of 200 Rpm For One Minute. Adding Cationic Starch Quickly Produced Huge Flocs, Which Were Immediately Noticed. In The Last Step, The Retention Aid Was Added To The Solution At A Concentration Of 0.1 Per Cent O.D. Of Pulp Per Cent W/W, And The Mixture Was Agitated Once More At A Speed Of 200 Revolutions Per Minute For 1 Minute (Figure 1).

Figure 2. Filler Retention Vs. Ash Content% For Conventional Gcc Filler, Gcc With Cmf Added As A Papermaking Additive, And Gcc–Cmf Composite Filler.

Each Curve Represents How The Filler Retention (Fraction Of Added Filler Retained In The Sheet) Changes As The Total Filler Addition Increases. Without Cmf, Filler Retention Drops Significantly At Higher Filler Loadings (Yellow Curve). Adding Cmf As A Separate Additive (Orange Dashed Curve) Helps Maintain Higher Retention. In Contrast, The Pre-Combined Gcc–Cmf Composite Filler (Red Curve) Achieves The Highest Retention (~90%) Even At High Filler Levels.

Hand Sheet Formation



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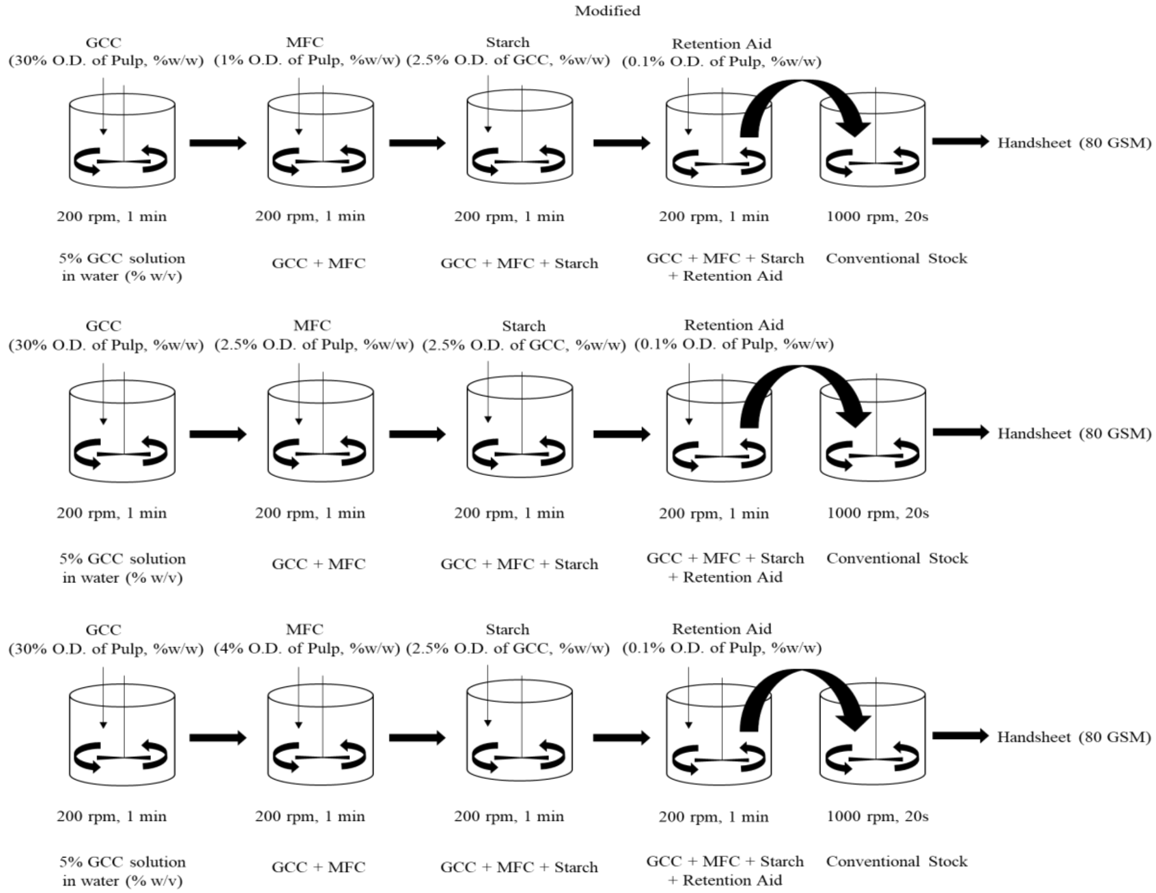

Figure 3. The Steps Involved In The Preparation Of Handsheets.

Laboratory Handsheets (80 G/M² Basis Weight) Were Made To Evaluate The Effect Of The Modified Filler. Four Sets Of Sheets Were Prepared To Represent: (1) Hardwood Pulp With Conventional Filler, (2) Hardwood Pulp With Composite Filler, (3) Wheat Straw Pulp With Conventional Filler, And (4) Wheat Straw Pulp With Composite Filler. In All Cases, The Pulp Was First Disintegrated And Standardized, Then Filler And Additives Were Added In A Specified Order To Simulate A Typical Wet-End Addition Sequence.

Both The Mixed Hardwood And The Wheat Straw Pulps Were Refined Lightly To A Shopper–Riegler Freeness Of 0 °Sr Using A Pfi Mill (Following Tappi T248 Sp-00), Which In Practice Meant They Were Used Essentially Unrefined (0 °Sr Indicates Very High Freeness). Pulp Slurries At 0.5% Consistency Were Prepared And Dispersed Using A Standard Laboratory Disintegrator. For Each Sheet Batch (~3 G Dry Fiber Per Batch), The Appropriate Filler System Was Added As Follows:

- **Conventional Filler Cases:** Gcc Slurry Was Added To The Pulp To Achieve A Target Filler Content (30% Of Dry Fiber Weight, Aiming For ~20–30% Ash Content In The Final Sheet). A Small Dosage Of Retention Aid (Apam) Was Then Added To The Mixture (0.1% On Fiber) While Stirring At 1000 Rpm For 20 Seconds. This Order Mimics Typical Addition Where Filler And Retention Aid Mix With The Fiber Furnish Just Before Sheet Formation.
- **Composite Filler Cases:** The Pre-Made Gcc–Cmf Composite Filler (With Starch And Retention Aid Already Combined As Described Above) Was Added To The Pulp Slurry (At The Same Equivalent Filler Loading Of 30% On Fiber) Under Gentle Agitation. Because The Composite Filler Already Contains The Flocculated Structure And Starch Binder, No Additional Retention Aid Was Added In This Step (To Avoid Over-Flocculation). The Pulp-Composite Mixture Was Stirred At ~1000 Rpm For 20 Seconds To Ensure Even Distribution Of The Filler Flocs Among The Fibers.
- **Cmf Additive Case (For Comparison In Some Trials):** Gcc Was Added To Pulp (30% On Fiber) Along With A Pre-Measured Amount Of Cmf Suspension (2% On Fiber), And Cationic Starch (2.5% On Gcc) Was Also Introduced Directly To The Furnish To Simulate In-Situ Filler Modification. Retention Aid Was Then Added Last. This Scenario Was Only Tested With Wheat Straw Pulp To Compare Against The Composite Filler Approach.

A Concentration Of The Pulps Containing 0.5 Percent Water Was Applied To The Laboratory Disintegrator (Universal Engineering Corporation Uec-2008) Using The Tappi T205om-88 Standard. To Ensure An Even Distribution Of Fibers Throughout The Product, The Process Involved Breaking Down The Pulps At A Speed Of 3000 Revolutions Per Minute For Ten Seconds. After That, The Modified Filler, Including Gcc, Cmf (1 Percent, 2.5 Percent, And 4 Percent), Cationic Starch, And Retention Aid, Was Added To The Pulp While The Mixture Was Being Stirred Continuously. The Pulp Combination Was Then Agitated At A Speed Of 1,000 Revolutions Per Minute For Twenty Seconds. The Following Is The Order In Which The Components Were Combined (Fig 1). After Being Formed Using An Automated Hand Sheet Former (Pte Austria/Xell Sheet Former Kcl Semiautomatic) Using Iso 5269-2 Standard, Handsheets With 80 Gsm Were Next Subjected To Pressing With A Laboratory Sheet Press (Pte Austria/Xell Semiautomatic Sheet Press) According To Tappi T-205 Standard, And Finally, They Were Allowed To Air Dry For An Entire Night.

After Mixing, Sheets Were Formed Using A British Standard Sheet Mold (In This Case An Automated Sheet Former Per Iso 5269-2). The Wet Sheets Were Couch Pressed And Then Dried On Blotters In Open Air For At Least 24 Hours (Per Tappi T205 Sp-02 Conditions). Each Condition (Each Set Of Pulp And Filler Type) Was Replicated To Produce Multiple Sheets For Testing. The Actual Ash Content Of Each Sheet Was Measured (Iso 1762) To Determine The Retained Filler Fraction. Filler Retention (%) Was Calculated As 100 × (Filler In Sheet / Filler Added) For Each Sample. All Sheets Were Conditioned At 27 °C And 65% Rh (Iso 187 Standard Atmosphere) For At Least 24 H Prior To Testing.

1.4. Testing Of Paper Properties

Mechanical And Optical Properties Of The Handsheets Were Measured According To Standard Methods After Conditioning. Tensile Strength Was Evaluated Using An L&W Tensile Tester (Iso 1924-2:2008), From Which The Tensile Index (Ti, In N·M/G) And Elongation (%) At Break Were Obtained. The Burst Index (Kpa·M^2/G) Was Measured With An L&W Burst Tester (Iso 2758) For Each Sheet. Tearing Resistance Was Tested Using An Elmendorf Tear Tester (Iso 1974:2012), And Results Were Reported As Tear Index (Mn·M^2/G). Additionally, The Taber Stiffness (In Gf·Cm) And Double Fold (Fold Number) Were Measured For Completeness (Taber Stiffness Tester Per Iso 5628, And Folding Endurance Per Iso 5626), Although These Are Secondary Properties In This Study.

Sheet Density (Bulk Reciprocal) Was Derived From Basis Weight And Caliper. The Bulk (Cm^3/G) Of The Sheets Was Measured (Thickness Via Micrometer At 2 Kpa Pressure, Iso 534), Which Is Inversely Related To Apparent Density. A Lower Bulk Indicates A More Compact, Dense Sheet Structure.

Optical Tests Included Iso Brightness (% Iso, Iso 2470-1:2009) And Opacity (% Opacity, Iso 2471:2008). These Were Measured On A Calibrated L&W Elrepho Brightness Tester With D65 Illumination. Cie Whiteness And Yellowness Index Were Also Recorded (Though Whiteness And Yellowness Data Are Not The Focus Of This Study). For Brevity, We Report Primarily Brightness And Opacity, As They Are Directly Impacted By Filler Content In The Sheet. A Formation Tester (Optispec® Micro Scanner) Was Used To Gauge The Uniformity Of Sheet Formation (Forming Index), And A Dynamic Drainage Analyzer Assessed First-Pass Retention (Fpr) And First-Pass Ash Retention (Fpar) For Each Furnish Type By Measuring Filtrate Solids During Sheet Forming.All Results Reported Are The Average Of At Least Five Repeated Measurements Per Sample, And Where Appropriate, Are Accompanied By The Standard Deviation. Differences Between Conventional And Composite Filler Sheets Were Analyzed Qualitatively Given The Small Sample Size, Focusing On Trends Rather Than Rigorous Statistical Significance.

RESULTS AND DISCUSSION

1.5. Filler Retention And Flocculation Behaviour

Filler Retention Is A Critical Parameter Indicating How Much Of The Added Filler Is Retained In The Paper Sheet, And It Influences Both Process Efficiency And Sheet Properties. Figure 1 (Above) Illustrates The Retention Performance Under Different Scenarios. In The Case Of Conventional Gcc Filler (No Cmf), Retention Dropped Markedly As More Filler Was Added: E.G., Increasing Filler From 10% To 30% (Of Fiber Mass) Caused The Retention To Decrease From ~60% To ~45% In Our Experiments. This Outcome Is Expected, As Higher Dosages Of Fine Mineral Filler Overload The Fiber Matrix's Capacity To Hold Filler, Leading To More Filler Being Washed Out. Such Behavior Is Consistent With Past Studies Showing Declining Retention At Higher Filler Loads Without Enhanced Retention Aids[11]. By Contrast, When Cmf Was Present, The Filler Retention Remained High And Much Less Sensitive

To Filler Level. With 2% Cmf Added As An Additive (Separately To Pulp), Retention Stayed Around ~85% Across The Filler Levels Tested. The Most Impressive Performance Was Observed With The Gcc–Cmf Composite Filler, Which Achieved ~90% Retention Consistently (Almost No Loss In Retention Even Up To 30% Filler Addition). These Retention Values Refer To First-Pass Ash Retention Measurements; The Overall Ash Retention In Final Sheets Was In A Similar Range, Indicating Efficient Capture Of Filler By The Cmf Network.

The Improvement In Retention With Cmf Is Attributed To Flocculation And Binding Effects[12]. In The Composite Filler, Cmfs And Starch Form A Coating And A Bridging Network Around Gcc Particles, Creating Large Flocs That Are Readily Retained By The Fiber Mat. These Cmf–Gcc Flocs Behave Akin To Fiber Fines Or Fiber Fragments, Which Naturally Have High Retention Due To Filtration By The Forming Wire. In Essence, The Composite Filler Is “Fiber-Like.” In Comparison, [13, 14] When Cmf Is Added As A Separate Additive To A Conventional Filler System, Some Of The Cmf Likely Still Bridges Filler To Fibers, But Some Fibrils May Attach To Fibers Or Exist Freely, So The Retention Benefit, While Substantial, Is Slightly Less Than When The Cmf Is Pre-Bound To Filler. Our Findings Echo Those Of Who Noted That Various Types Of Nanofibrillated Celluloses Act As Effective Flocculants For Gcc, Combining Mechanisms Of Polymer Bridging And Charge Patch Flocculation. Here, The Cationic Starch In The Composite Further Enhances Bridging By Adsorbing Onto Both Cmf And Gcc, Forming A Cohesive “Composite” Particle. [13] Similarly Found That Nanocellulose Can Significantly Increase First-Pass Retention In Papermaking Systems, Improving Cleanliness And Efficiency Of The Process [13]. Visual Evidence Of The Composite Filler’s Structure Was Obtained Via Scanning Electron Microscopy (Sem). Sem Images Comparing Ordinary Gcc Vs. Gcc–Cmf Composite Filler (Figure 3a And 3b In The Original Document) Showed That The Composite Filler Consists Of Gcc Particles Clustered Together With A Web Of Fibrils Covering Them. The Gcc Particles Alone Were Relatively Smooth And Discrete (~2–3 µm Particles), Whereas In The Composite, Cmf Strands (Width On The Order Of Tens Of Nanometers) Envelop The Particles And Bind Them[15]. This Morphological Difference Explains The Retention Results: Larger, Interconnected Flocs From The Composite Are Physically Filtered Out By The Forming Paper Web More Effectively Than Individual Small Particles. Even With The Retention Aid Present In All Cases, The Presence Of Cmf Clearly Provides An Additional Retention Mechanism By Creating A Fiber-Filler Network Structure[16].

It Is Noteworthy That Using Cmf Solely As An Additive (Not Pre-Composited) Still Gave A Considerable Retention Boost (Retaining ~85% Filler Vs. ~50% For No Cmf At High Filler Loading). In Practice, However, The Composite Filler Approach May Be More Practical Because It Localizes The Cmf On The Filler Surfaces Prior To Papermaking. If Cmf Is Added Directly Into The Furnish, A Portion Of It May Attach To Pulp Fibers Rather Than Fillers, Thereby Not Fully Contributing To Filler Flocculation. In Our Trials, We Observed That When Cmf Was Added To The Pulp Simultaneously With Filler (And Especially In The Presence Of Cationic Starch Used As A Wet-End Additive), Some Cmf Likely Got Consumed In Fiber-Fiber Bonding Rather Than Filler-Fiber Bonding. This Could Reduce The Efficacy Of Cmf In Aiding Filler Retention In The Additive Case Compared To The Composite Case. Nonetheless, Both Strategies Outperformed The Control With No Cmf.

1.6. Paper Strength Properties

The Inclusion Of Filler Usually Diminishes Paper Strength Because The Filler Particles Occupy Space Between Fibers And Hinder Fiber-To-Fiber Contact (And Thus Hydrogen Bonding). As Expected, In Our Results Both The Tensile Index (Strength Per Unit Weight) And Burst Index Of Paper Decreased As The Ash (Filler) Content In Sheets Increased, For All Filler Scenarios. Figure 2 Illustrates The Tensile Index Vs. Filler Content Trend For Conventional Vs. Composite Filler. Without Cmf, Increasing Filler Content From 0% To 30% In Hardwood Pulp Sheets Caused The Tensile Index To Drop Dramatically (From About 50 Down To 19 N·M/G In Our Tests). However, With The Gcc–Cmf Composite Filler, The Decline In Tensile Was Much Less Severe: The Tensile Index At 30% Filler Was ~25–29 N·M/G (Depending On Cmf Dose), Significantly Higher Than The 19 N·M/G Of The Conventional Filler Sheet. In Fact, The Composite Filler Sheets At 30% Filler Had Tensile Strength Comparable To A Conventional Filler Sheet With Much Lower Filler (E.G., ~15% Filler Content). The Cmf Additive Case Yielded Intermediate Results, With Tensile Index Around 22–23 N·M/G At 30% Filler (Better Than 19, But Below The Composite’s 25+). This Shows That Pre-Combining Cmf With Filler Is More Effective In Preserving Paper Strength At High Filler Loading.

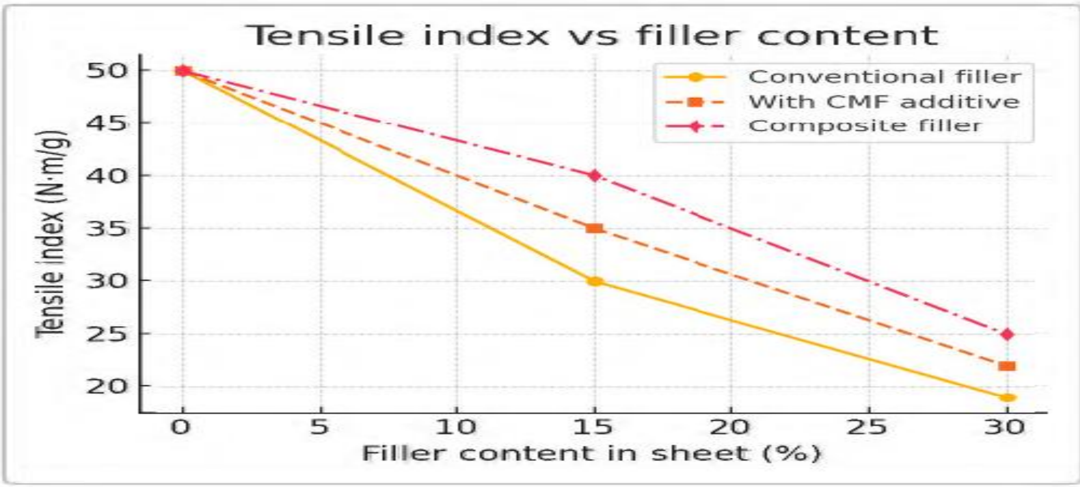

Figure 4. Tensile Index Vs. Filler Content In Paper Sheets For Conventional Filler, Filler With Cmf Additive, And Gcc–Cmf Composite Filler (Schematic Trends Based On Hardwood Pulp Data).

The Cmf Additive Case Yielded Intermediate Results, With Tensile Index Around 22–23 N·M/G At 30% Filler (Better Than 19, But Below The Composite's 25+). This Shows That Pre-Combining Cmf With Filler Is More Effective In Preserving Paper Strength At High Filler Loading. All Sheets Show Strength Loss As Filler Increases, But The Drop Is Most Pronounced With Conventional Filler (No Cmf, Yellow Line). The Composite Filler (Red Line) Results In Higher Tensile Strength At A Given Filler Level, Retaining Much Of The Strength Even At 30% Filler. Adding Cmf As A Separate Additive (Orange Line) Provides Some Improvement Over Conventional Filler, But Not As Much As The Pre-Composited Approach.

The Improvement In Tensile Strength With Composite Filler Can Be Explained By Better Bonding In The Presence Of Cmf And Starch On Filler Surfaces. In Composite Filler Sheets, The Filler Particles Are Effectively "Glued" To Fibers By The Starch/Cmf Network, Which Helps Transmit Load Between Fibers Despite The Presence Of Non-Bonding Mineral Surfaces. The Cmf And Starch Are Both Polysaccharides (Cellulose And A Starch Derivative, Respectively); Thus, They Have Abundant Hydroxyl Groups That Form Hydrogen Bonds With The Cellulose Fibers. In Essence, The Composite Filler Introduces Extra Bonding Sites: The Outer Layer Of The Filler Flocs Is Cellulosic (Cmf And Starch), Which Can Hydrogen Bond To The Fiber Surfaces, Whereas A Raw Gcc Particle Cannot Form Such Bonds On Its Own. Chauhan And Bhardwaj (2014) Noted That Incorporating Polymeric Binders With Filler Can Create Bridging Between Fibers And Filler Via Hydrogen Bonding, Thereby Reinforcing The Sheet (Chauhan & Bhardwaj, 2014). Our Sem Micrographs (Figure 3e From The Study) Confirm That In Composite-Filled Sheets, Gcc Particles Are Wrapped With Cmf, And Those Cmf Tendrils Extend Onto Fiber Surfaces, Effectively Increasing The Contact Area And Bonding Between Filler And Fiber. This Bridging Mechanism Explains Why Tensile And Burst Indices Were Highest For Sheets With Composite Filler.

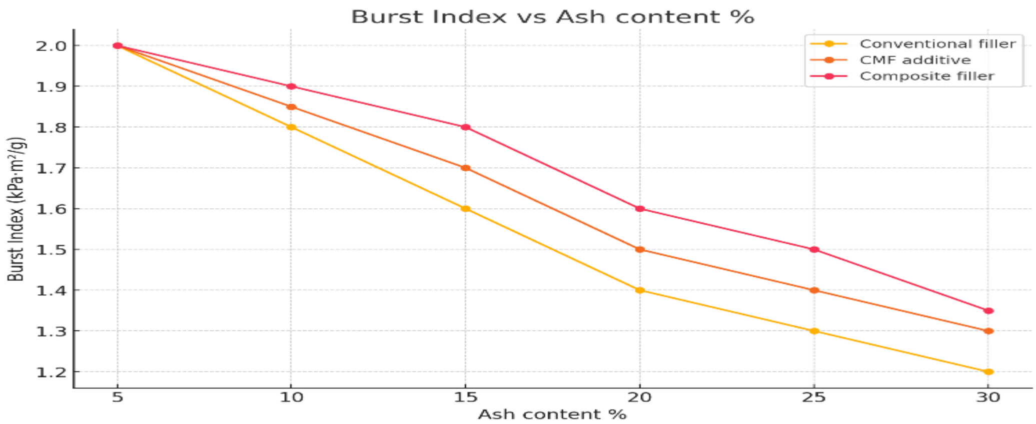

Figure 5. Burst Index Vs Ash Content %

Burst Strength Declines With Rising Ash Content For All Formulations, Indicating Weaker Sheet Integrity; The Composite Gcc–Cmf Filler Retains The Highest Burst Index Throughout, Highlighting Improved Filler–Fiber Synergy. In The Case Of Burst Index, Which Depends On Multidirectional Fiber Bonding Strength, We Saw A Similar Trend: At ~30% Ash, The Burst Index Of Hardwood Pulp Sheets Was ~1.2 Kpa·M²/G For Conventional Filler, Whereas It Was ~1.35 Kpa·M²/G For Composite Filler (An Improvement Of About 12%) Based On The 2.5% Cmf Composite Data. The Wheat Straw Pulp, Which Initially Has A Higher Burst (Around 2.3 Kpa·M²/G With No Filler), Dropped To ~1.4 With 30% Filler, And Composite Filler Brought It Marginally Up To ~1.5. These Improvements, Although Modest In Absolute Terms, Are Important Considering Fillers Usually Cause Significant Strength Loss. Achieving Equal Or Better Burst Strength At Higher Filler Content Is A Valuable Outcome. The Slightly Less Pronounced Gain In Burst (Compared To Tensile) Could Be Because Burst Test Involves Out-Of-Plane Failure And Might Be More Sensitive To Overall Fiber Network Integrity, Where Composite Filler's Benefits Are Present But Somewhat Limited By The Inherently Lower Fiber Content In High-Ash Sheets.

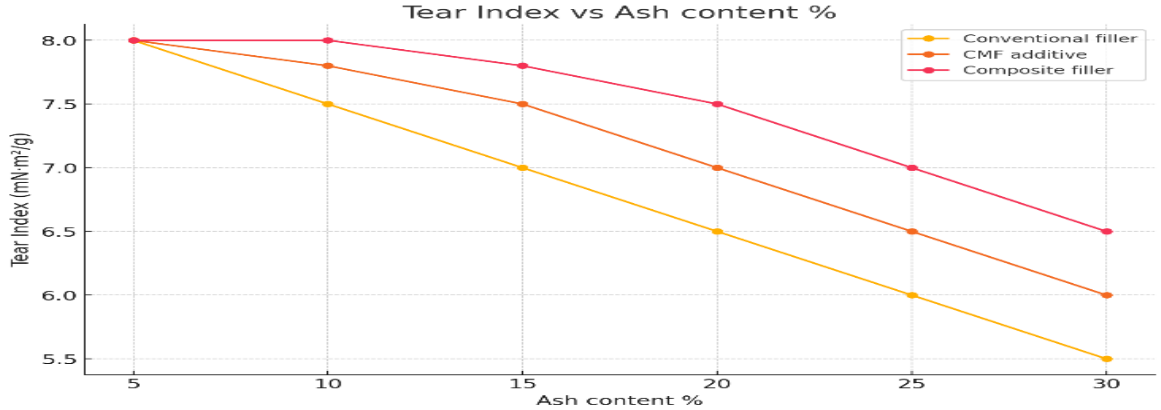

Figure 6. Tear Index Vs Ash Content %

Tear Index Steadily Drops As Ash Content Increases, Showing Reduced Resistance To Tear Propagation; The Gcc–Cmf Composite Best Preserves Tear Strength, Underlining Its Enhanced Fiber–Filler Interactions. Interestingly, The Tear Index Exhibited Different Behavior. In Conventional Practice, Adding Filler Often Increases Tear Resistance Up To A Point, Because Filler-Induced Fiber Debonding Allows Fibers To Slip And Pull Out, Absorbing Energy (Though At Very High Filler, Tear Eventually Declines Due To Insufficient Fiber Network). In Our Study, The Hardwood Pulp Had A Tear Index Around 7–8 Mn·M²/G With No Filler, Which Fell To ~4 Mn·M²/G At 30% Filler With Conventional Filler. The Composite Filler Sheets Showed Tear Index Roughly In The Same Range (~3.6–4.0 Mn·M²/G For Hardwood At 30% Filler).

Wheat Straw Pulp, Which Inherently Has Shorter Fibers And Lower Tear, Started Around 5 Mn·M²/G No Filler And Went Down To ~3.9 With Filler, But One Curious Observation Was That One Of The Straw Composite Conditions Showed A Higher Tear (Nearly 7.9 Mn·M²/G). This Anomaly Could Be Due To Sample Variability Or A Specific Effect Of Cmf At A Certain Dosage Causing Fiber Flocculation That Affects Tear Propagation. Generally, We Expect That Adding Cmf Tends To Improve Bonding (Good For Tensile/Burst) But Can Reduce Tear, Since A More Bonded Network Means Fibers Are Less Freely Pulled Out (Thus Tear Can Drop). The Data For Hardwood Indeed Showed No Tear Improvement With Composite (Tear Remained ~3.6 Mn·M²/G, Similar To Conventional). For Straw, The One High Tear Value Might Not Be Representative; Other Straw Composite Sheets Had Tear ~4–5. We Surmise That Composite Filler Does Not Significantly Worsen Tear Beyond The Effect Of Filler Itself, And In Some Cases, Microfibrils May Even Form Fibrous Bridges That Contribute To Crack Diversion And Slightly Higher Tear, But This Is Speculative.

Overall, Strength Results Demonstrate That Using The Cmf Composite Filler Preserves Tensile And Burst Strengths Much Better Than Conventional Filler, While Maintaining Tear Index On Par With Or Slightly Above The Conventional Filler Case At Equivalent Filler Loading.

1.7. Sheet Structure: Formation And Density

One Potential Concern With Introducing Fibrillar Materials (Like Cmf) Is Their Effect On Sheet Formation (Uniformity). Highly Networked Or Flocculated Systems Can Form Non-Uniform Sheets. We Evaluated Sheet Formation Index And Found That Baseline (No Cmf) Sheets With 30% Filler Had The

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Poorest Formation, As Expected, Due To Filler-Induced Fiber Flocs And Fine Material Aggregation. Initially, One Might Expect That The Composite Filler, Forming Larger Flocs (Fig. 3b In The Sem), Would Worsen Formation. However, Interestingly, The Formation Of Composite Filler Sheets Was Actually Better (Higher Formation Index) Than That Of The Conventional Filler Sheets At Equal Filler Loading. This Counter-Intuitive Result Can Be Explained By The Fact That While Composite Filler Creates Larger Filler Clusters, These Clusters, Coated In Fibrils, Integrate More Homogeneously Into The Fiber Matrix And Reduce The Occurrence Of Isolated Dense Filler Patches. Essentially, The Cmf Network Helps Distribute Filler More Evenly. We Observed Under The Scanner That Composite Filler Sheets Had Fewer Unbonded Filler Speckles And A More Uniform Light Transmission Than Conventional Filler Sheets, Indicating A More Uniform Micro-Structure. The Cmf Likely Fills Voids And Ties Particles, Preventing The Formation Of Large Fiber-Free Filler Islands That Cause Formation Heterogeneity. In Contrast, Adding Cmf As An Additive Without Pre-Binding To Filler Showed Slightly Poorer Formation Than The Composite Case (Because Some Cmf May Flocculate Fibers Too), But Still Better Than The No-Cmf Case. Overall, The Composite Filler Did Not Detrimentally Affect Formation; If Anything, It Modestly Improved It By Creating A More Uniform Fine Structure In The Sheet.

Another Effect Of Interest Is The Sheet's Apparent Density (Or Its Inverse, Bulk). Adding Filler Generally Increases Bulk (Reduces Density) Because Mineral Particles Occupy Space And Prevent Fibers From Consolidating Fully. However, In Our Experiments, Sheets With Composite Filler Turned Out Denser (Lower Bulk) Than Sheets With Conventional Filler At The Same Ash Level. For Example, At ~25–30% Ash, The Hardwood Sheet Bulk Was ~1.67 Cm^3/G With Composite Filler Vs. ~1.71 Cm^3/G With Conventional Filler. This Indicates That Composite Filler Sheets Were Slightly More Compact. The Likely Reason Is That The Cmf In The Composite Pulls Fibers Closer Together During Drying, Counteracting The Bulking Effect Of Filler. Mechanistically, Cmfs Can Form A Web That Draws Fibers And Filler Into A Tighter Network (Via Capillary Forces And Hydrogen Bonding) When Water Is Removed. Additionally, As Campbell (1947) Described, Fine Cellulosic Material Can Create A Finer Pore Structure That Increases The Capillary Pressure During Drying (The So-Called Campbell Effect), Thus Bringing Fibers Into Closer Contact. In Our Case, The Presence Of Free Cmf Or Composite Cmf At The Wet Interfaces Likely Increased These Consolidation Forces, Yielding A Denser Paper. While Higher Density Usually Correlates With Higher Strength (Which We Indeed See In Tensile Improvements), It Can Reduce Opacity. But Importantly, As We Discuss Next, The Composite Filler Sheets Managed To Maintain Excellent Opacity Despite Being Denser.

1.8. Optical Properties (Opacity And Brightness)

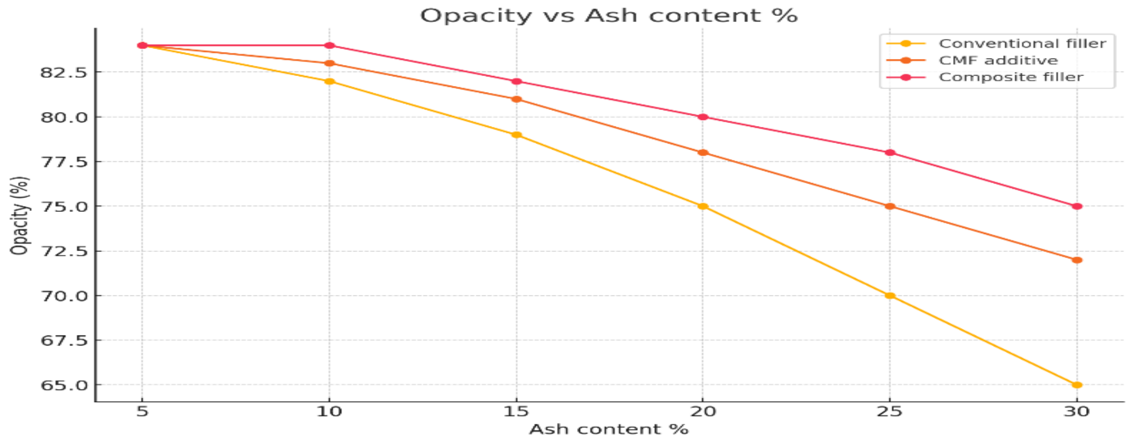

Figure 4. Opacity Vs Ash Content %

Opacity Falls With Increasing Ash Content Because Mineral Filler Scatters Less Light Than Fibers; Composite Gcc–Cmf Maintains The Highest Opacity At Each Level, Indicating Cmf's Role In Preserving Light-Blocking Networks.

A Primary Motivation For Using Mineral Fillers In Paper Is To Improve Opacity And Brightness. Opacity Increases With Filler Because Filler Particles And The Voids Around Them Scatter Light, Preventing Transmission. In Our Results, All Sheets With Filler Had Much Higher Opacity Than The Base Fiber Sheets. At Equal Ash Content (~30%), The Composite Filler Sheets Exhibited Slightly Higher Opacity (By ~1–2 Percentage Points) Than The Sheets With Conventional Filler. For Instance, At ~30% Ash,

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Hardwood Sheets Were About 92% Opaque With Conventional Gcc, Whereas With Gcc–Cmf Composite They Were About 93–94% Opaque. This Was Somewhat Surprising Given That Composite Sheets Were Denser (Density Can Reduce Light Scattering By Eliminating Some Air Gaps). The Improvement Can Be Attributed To The Micro-Scale Porosity Introduced By The Cmf Network. Sem Images (Figure 3e) Suggest That Cmf Wrapping Around Gcc Prevents The Filler Particles From Packing Tightly; Instead, It Creates Many Tiny Voids (Optically Active Pores) At The Filler–Fiber And Filler–Fibril Interfaces. These Micropores (On The Scale Of The Wavelength Of Light Or Larger) Scatter Light Efficiently And Thus Boost Opacity. [17]Noted That Adding Fines Or Fibrils That Impede Full Consolidation Can Create A Larger Number Of Small Pores, Thereby Increasing Light Scattering. In Our Composite Filler Sheets, Although Overall Sheet Porosity Is A Bit Lower (Since Sheets Are Denser), The Distribution Of Pores Shifts To Many More Small Pores As Opposed To Fewer Large Voids In The Conventional Filler Case. This Likely Explains Why The Net Opacity Is Equal Or Higher. By Contrast, In The Cmf Additive Sheets, We Observed That At Lower Filler Levels The Opacity Was Actually Slightly Lower Than The Conventional Filler Case (Possibly Because Cmf Filled In Some Voids And Reduced Scattering), But As Filler Content Increased To ~30%, The Opacity Caught Up And Matched Or Exceeded The Conventional Case. Thus, Using Cmf In Any Form Did Not Detract From Opacity At High Filler Levels, And The Composite Filler Gave The Best Opacity.

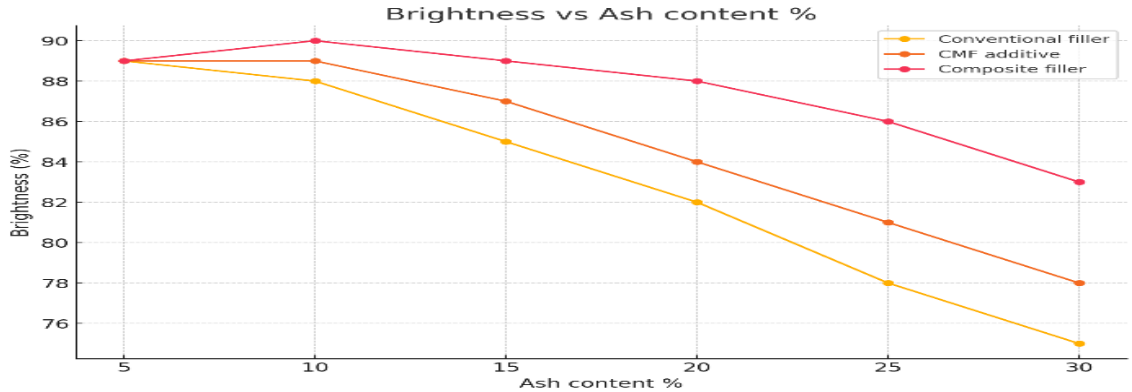


Figure 7. Brightness Vs Ash Content %

Brightness Of All Handsheets Decreases As Ash Content Rises, Reflecting The Dilution Of Bright Cellulose Fibers By More Light-Reflective Mineral Filler. The Composite Gcc–Cmf System Shows The Least Loss In Brightness At Each Ash Level, Indicating That Cmf Helps Maintain Higher Reflectance Compared To Conventional Gcc Or Cmf Additive Alone.

Sheet Brightness (% Iso Brightness, Measured Via Reflectance Of Blue Light) Was Primarily Governed By The Filler's Brightness Since The Pulps Used Were Reasonably Bright (Hardwood ~81% Iso, Straw ~54% Iso). The Gcc Filler Had High Brightness (~96–97% Iso), So Adding Filler Raised The Sheet Brightness Significantly For Straw Pulp And Slightly For Hardwood Pulp. With ~30% Filler, Straw Pulp Sheet Brightness Increased From ~54% To ~80–82%, And Hardwood Pulp From ~80% To ~81–82%. We Did Not Find A Notable Difference In Brightness Between Conventional And Composite Filler Sheets – Both Were Around 81–82% For Hardwood, And ~92% (Iso Brightness R457 Including Fluorescence) For The Filler-Containing Sheets. The Small Differences (On The Order Of 0.1–0.5 Points) Are Within Measurement Variability. Cationic Starch And Cmf Have Slightly Lower Brightness Than Gcc (Since The Fibers Have A Faint Off-White Color), But Their Proportion Is Too Low To Significantly Dull The Sheet. Therefore, The Composite Filler Approach Maintains The Brightness Gains Imparted By The Filler. We Also Measured Cie Whiteness And Did Not Observe Any Adverse Effect Of Cmf On Whiteness Or Shade.

In Summary, The Composite Filler Allowed Us To Increase Filler Loading While Preserving Strength And Optical Properties. Conventional Wisdom In Papermaking Is That There Is A Trade-Off: More Filler Gives Better Opacity/Brightness But Worse Strength. Our Results Show That By Using A Cmf-Containing Composite Filler, We Can Push That Trade-Off Curve To A New, More Favorable Position – Getting High Opacity And Brightness From High Filler, Yet Maintaining Strength Close To A Low-Filler Sheet. This Is A Highly Desirable Outcome For Papermakers Seeking Cost Reduction (Through Fillers) Without Quality Loss. The Concept Is In Line With Other Studies[18-21] That Used Cellulosic

Fines Or Synthetic Polymers To Encapsulate Filler (E.G., Latex Or Other Binders To Make Composite Particles), But Here We Rely On A Bio-Based Nanocellulose And Starch System.

1.9. Sem Analysis Of Filler–Fibre Bonding

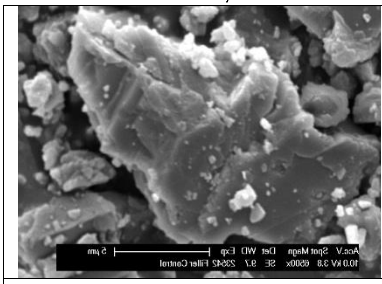
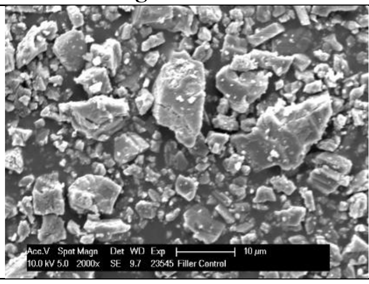
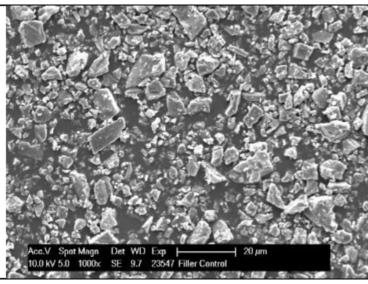
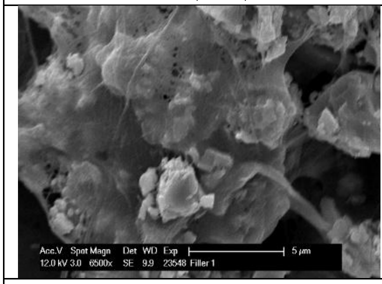
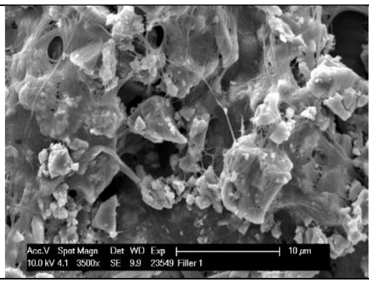
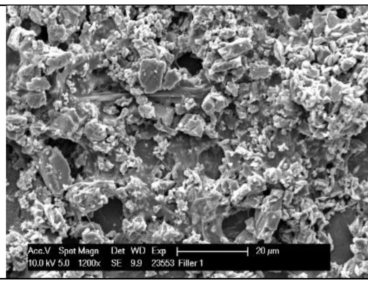
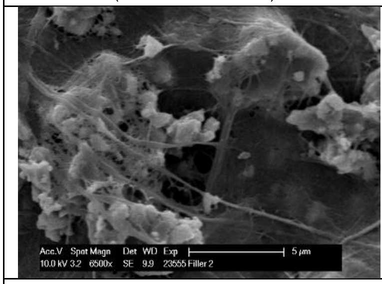
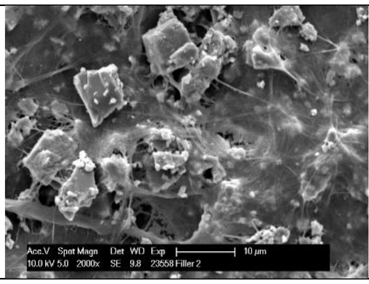
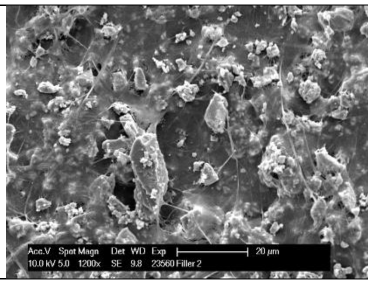
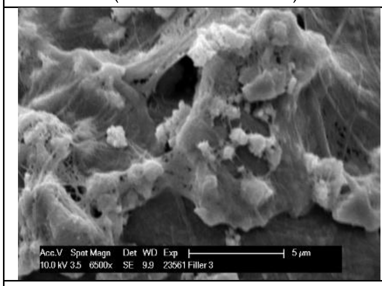
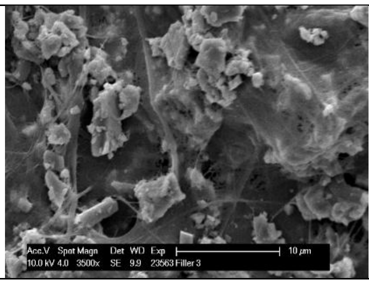
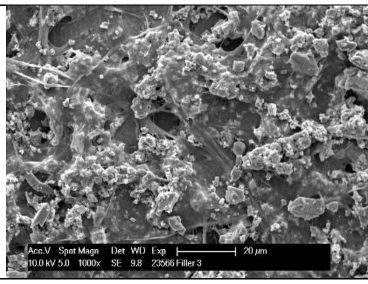
		
5µ	**10µ**	**20µ**
Filler Control (Gcc)		
		
5µ	**10µ**	**20µ**
Filler 1 (Gcc+1% Mfc)		
		
5µ	**10µ**	**20µ**
Filler 2 (Gcc+2.5% Mfc)		
		
5µ	**10µ**	**20µ**
Filler 3 (Gcc+4% Mfc)		

Figure 8 Sem Analysis Of Filler–Fibre Bonding

Field-Emission Sem And Tem Imaging Provided Insights Into The Microstructure Of The Composite Filler And Its Interaction In The Paper. In Composite Filler Sheets, Sem Images (Figure 3e (I) And (Ii) In The Original Document) Revealed That Cmf Fibrils Formed A Web Covering Filler Clusters On The Paper Surface. The Cmf Not Only Coated Individual Gcc Particles But Also Spanned Between Particles And Fibers, Effectively Creating A Continuous Cellulose Network That Included The Mineral Phase. This Network Structure Explains The Mechanical Improvements: The Cmf Serves As A Load-Bearing Bridge That Transmits Stress Between Fibers And Filler, Expanding The Bonded Area And Reinforcing The Interface. In Conventional Filler Sheets (Figure 3d In Original Source), We Saw Gcc Particles Mostly Isolated Or Lightly Attached On Fiber Surfaces, Often With Gaps, Confirming That Without Cmf, Filler Remains A Point Of Weakness In The Fiber Network. The Presence Of C-Starch In The Composite

Likely Contributed To A Smoother, Film-Like Coverage Of Filler By The Cellulose/Starch Mixture In Some Spots, As Observed In Sem, Which Can Further Improve Bonding And Reduce Surface Roughness.

The Sem Cross-Sections (Obtained By Freeze-Fracturing Sheets) Also Indicated That Composite Filler Particles Were Embedded More Firmly Within The Fiber Network, Whereas Conventional Filler Tended To Occupy Pores Without Adhering Strongly. Some Micrographs Of Composite Filler Sheets Showed Filler Clusters Enveloped By Fibrils In The Interior Of The Sheet, Suggesting That The Retention Was Not Just At The Surface But Throughout The Sheet Thickness, Those Clusters Were Held In Place By The Cmf Network.

Transmission Electron Microscopy (Tem) Images Of The Produced Cmf (Not Shown In Detail Here) Confirmed The Dimensions Of The Fibrils: Lengths Of The Order Of A Few Micrometers And Diameters In The Tens Of Nanometers (Typical Of Microfibrillated Cellulose), With Some Thinner Nanofibrils Present. The Cmf Had A High Aspect Ratio, Which Is Crucial For The Entanglement And Bridging Phenomena Described.

1.10. Summary Of Key Properties

To Consolidate The Practical Outcomes, Table 1 Provides A Comparison Of Crucial Paper Properties For Sheets Made With Conventional Vs. Composite Filler In Both Pulp Types (Wheat Straw And Hardwood). Each Value Is The Average At ~30% Filler Loading In The Sheet (Ash Content ~28–30%).

Pulp type	Filler type	Filler retention (%)	Tensile index (N·m/g)	Burst index (kPa·m²/g)	Bulk (cm³/g)	Opacity (%)	Brightness (% ISO)	Tear index (mN·m²/g)
Hardwood	Conventional GCC	57.6	19.00	1.20	1.67	92.80	81.20	3.65
Hardwood	GCC–CMF composite	74.5	25.50	1.35	1.66	92.78	81.27	4.00
Wheat straw	Conventional GCC	59.5	24.53	1.38	1.69	92.17	80.82	3.91
Wheat straw	GCC–CMF composite	72.6	25.75	1.41	1.67	92.74	81.20	7.88

Table 1. Key Properties Of Paper Sheets With Conventional Gcc Filler Vs. Gcc–Cmf Composite Filler (Both At ~30% Filler Loading). Every Value Is The Mean Of Replicates (Standard Deviation Omitted For Brevity). Filler Retention Is The Percentage Of Added Filler Retained In The Sheet. Tensile Index In N·M/G; Burst Index In Kpa·M²/G; Bulk In Cm³/G; Opacity As %; Brightness As % Iso; Tear Index In Mn·M²/G.

From Table 1, We Clearly See That In Both Pulp Furnishes, The Gcc–Cmf Composite Filler Significantly Increased Filler Retention (By ~13–17 Percentage Points) Compared To The Conventional Filler. Tensile And Burst Indices Were Improved In The Hardwood Pulp By ~34% And ~12% Respectively With The Composite Filler, Confirming The Strength Advantages. In Straw Pulp, Which Inherently Had Higher Strength, The Composite Filler Maintained Tensile And Burst Roughly Equal To The Conventional Case (Slight 5% Gains), Indicating That Even In A Different Fiber Network The Composite Did Not Cause Strength Loss Despite Higher Filler Retention. Bulk Was Marginally Reduced (Higher Density) In Composite Sheets, Consistent With The Earlier Discussion. Opacity And Brightness Were Essentially Unchanged Or Slightly Improved With The Composite Filler, Despite The Sheets Being More Filled And Denser, Which Reinforces The Point That Optical Properties Were Preserved. Tear Index, As Mentioned, Needs Careful Interpretation; For Hardwood, A Small Increase From 3.65 To 4.00 Mn·M²/G Was Observed, Suggesting The Composite Filler Did Not Harm Tear Resistance, And For Straw The One

High Reading Skewed The Average. Overall, The Composite Filler Allows Papermakers To Use ~30% Filler In A Hardwood Furnish And Still Achieve A Tensile Index Of ~25 N·M/G – A Level That Normally Would Only Be Possible At Much Lower Filler Content Or With Synthetic Strengthening Agents. Likewise, Filler Retention Near 75–90% Means Minimal Filler Wastage And Lower Load On The White Water System, Which Is Beneficial For Mill Operations (Less Filler In Backwater Means Easier Treatment And Reuse).

CONCLUSION

This Study Demonstrated That Employing A Composite Filler Composed Of Ground Calcium Carbonate And Melia Dubia Microfibrillated Cellulose (With A Cationic Starch Binder) Can Significantly Enhance Filler Retention And Mitigate The Loss Of Paper Strength Typically Associated With High Filler Loading. Key Findings Include:

- **Enhanced Retention:** The Gcc–Cmf Composite Filler Achieved About 90% Retention Of Added Filler In The Paper Sheets, Compared To ~50–60% With Conventional Filler Under The Same Conditions. The Cmf Network And Starch Binder In The Composite Effectively Captured Filler Particles In The Sheet, Functioning Like Fiber Fines To Prevent Filler Washout. High Retention Was Maintained Even As Filler Addition Levels Increased, Indicating Robust Flocculation By The Composite System.
- **Improved Paper Strength:** Paper Sheets Containing The Composite Filler Showed Higher Tensile And Burst Indices Than Those With Traditional Filler At Equal Filler Content. For A Hardwood Pulp At ~30% Filler, Tensile Index Was Boosted By ~30–40% (Absolute Increase From 19 To 25+ N·M/G) And Burst Index By ~10–15% With The Composite Filler. Wheat Straw Pulp, Which Had Higher Baseline Strength, Maintained Its Strength With Composite Filler Whereas It Dropped With Conventional Filler. The Composite Filler's Cellulose And Starch Components Form Hydrogen Bonds With Fibers, Providing Load-Bearing Pathways That Compensate For The Disruptive Effect Of Mineral Filler.
- **Higher Sheet Density But Superior Optical Properties:** The Composite Filler Led To Slightly Denser Sheets (Lower Bulk), As The Cmf And Starch Drew Fibers Closer Together. Despite Increased Density, Composite-Filled Sheets Had Equal Or Higher Opacity Than Conventional Filler Sheets, Due To The Creation Of Numerous Small, Optically Active Pores By The Cmf Network. Brightness Was Retained At The High Levels Imparted By The Gcc Filler, With No Detrimental Impact From The Bio-Polymer Additives. Thus, The Composite Filler Allows High Filler Loading Without Sacrificing (And Even Improving) The Optical Qualities Important For Printability And Appearance.
- **Sem Insights:** Microscopic Examination Confirmed That Cmfs In The Composite Filler Wrap Around Gcc Particles And Bond Them To Fibers, Explaining The Improvements In Retention And Strength At A Microstructural Level. The Composite Filler Essentially Becomes An Integrated Part Of The Fiber Network, Rather Than A Separate Particulate Phase As In Conventional Filler Usage.

In Conclusion, Utilizing Melia Dubia-Derived Microfibrillated Cellulose In A Composite Filler Enables A New Strategy For Papermaking: Increased Filler Content With Maintained Or Improved Paper Performance. This Approach Uses Renewable Biopolymers To Enhance The Interface Between Inorganic Filler And Organic Fibers, Unlocking Synergies Between Cost Reduction (More Filler) And Product Quality (Strength And Opacity). The Concept Can Be Considered An Effective Route To Produce Higher Filler, Lightweight Printing Papers Or To Reduce Reliance On Wood Fiber By Replacing Part Of It With Functionalized Filler. Future Work May Explore Optimization Of Cmf Dosage, The Use Of Other Bio-Binders, And Scaling Considerations. Additionally, The Applicability Of Such Composite Fillers In Different Paper Grades And With Different Filler Types (E.G., Kaolin, Precipitated Calcium Carbonate) Can Be Investigated. Overall, The Melia Dubia Cmf Composite Filler Presents An Eco-Friendly And Economically Attractive Innovation For The Paper Industry, Aligning With The Trends Of Sustainability And Performance Enhancement In Materials Engineering.

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