Synergistic Biomass-based Dual-polymer System of Cationic Guar Gum and Gum Arabic for Enhanced Retention and Drainage in Papermaking

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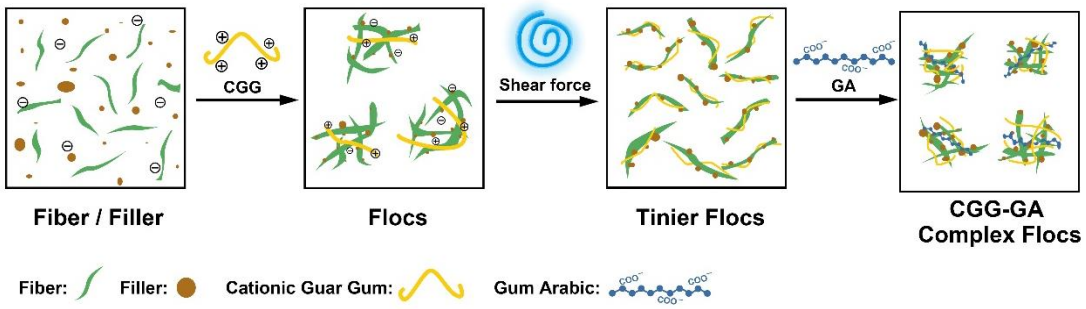
GRAPHICAL ABSTRACT



Synergistic Biomass-based Dual-polymer System of Cationic Guar Gum and Gum Arabic for Enhanced Retention and Drainage in Papermaking

Mingjie Tan,[a,*] Weitao Zhang,[a] Enhao Fu,[b] and Fei Yang [b]

Gum arabic (GA), a natural plant-derived polysaccharide renowned for its adhesive properties, is widely utilized across various industries, particularly in papermaking. This study systematically investigated the retention and drainage performance of pulp when treated with cationic guar gum (CGG) and GA individually and further explored the development of a biomass-based dual-polymer retention and drainage aid system. The CGG-GA system outperformed single-component additives by initially forming larger flocs with CGG, which were subsequently stabilized by GA, leading to enhanced retention and drainage efficiency. Additionally, the dual-polymer system reduces the cationic demand of white water, highlighting a synergistic interaction between CGG and GA. Scanning electron microscopy (SEM) analysis revealed a more uniform floc distribution in the dual-polymer system, while physical property tests confirm improved tensile strength in handsheets. This study presents a sustainable and effective alternative to conventional retention and drainage aids, offering both environmental benefits and enhanced paper quality.

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Keywords: Dual-polymer System; Cationic guar gum; Gum arabic; Retention and drainage; Handsheet property

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INTRODUCTION

To ensure the retention of fine fibers and fillers during paper production and achieve a smooth drainage process, retention and drainage aids are frequently added to the pulp. However, achieving an optimal balance among retention, drainage, and drying with just one additive proves challenging (Zhang and Hu 2004; Hou *et al.* 2004; Antunes *et al.* 2008). Thus, dual-polymer retention and drainage aid systems have been introduced (Fu *et al.* 2011). In modern dual-polymer retention and drainage systems (*e.g.*, for packaging or printing paper), the primary component is a cationic coagulant with high charge density and low molecular weight, such as polyamine, polydiallyl dimethyl ammonium chloride, polyethyleneimine, or cationic starch, while the secondary component is a water-soluble anionic flocculant with low charge density and high molecular weight, such as anionic polyacrylamide (Hu *et al.* 2008). The first cationic polymer added adsorbs to the surface of the fibers and fine components, imparting a positive charge characteristic of a cationic charge patch.

The cationic charge patch provides sites for attaching anionic polyelectrolytes, which subsequently interact with the patch's positive charge *via* high molecular weight anionic polymers. Consequently, the repulsion from nearby negative charges causes the anionic polymer to extend into the surrounding water phase. This facilitates its attachment to the positively charged area on another particle's surface, creating a bridging effect that promotes the aggregation of these particles (Gill 1991). Commonly, this method employs cationic polymers along with negatively charged inorganic particles. Examples include the Compozil system (cationic starch + colloidal silica; Sonehara and Andersson 2001); the Hydrocol system (cationic polyacrylamide + modified bentonite; Langley and Litchfield 1986); the modified dicyandiamide–formaldehyde polymer–bentonite system (Zhang *et al.* 2019); the cationic starch - anionic nanosilica system (Khosravani *et al.* 2010); and the micropolymer system, which incorporates spherical, highly branched anionic polymer microparticles—synthesized *via* microemulsion into a filamentary micronetwork structure—alongside cationic components (Honig *et al.* 2000). However, conventional methods are insufficient for producing paper requiring food-contact compliance, such as baking paper and coffee filter papers. This is because even minimal residual additives in these products may still present health risks through chronic exposure or chemical migration (Geueke *et al.* 2024).

Currently, research on biomass-based dual-polymer systems for retention and drainage assistance remains limited. Guar gum, being a natural polymer, has long been utilized in studies of retention and drainage aids due to its ability to induce the flocculation of fine fibers and fillers in slurries (Barua *et al.* 1994). Additionally, cationic guar gum (CGG) has been demonstrated to be a more effective flocculant than natural guar gum (Levy *et al.* 1995). Yang *et al.* (2020) investigated the system's retention performance when combining cationic guar gum and amphoteric polyacrylamide (PDAA), finding that optimal retention at a PDAA/CGG ratio of 1:1. As a natural plant-derived polysaccharide, gum arabic (GA) is widely utilized in the paper industry due to its excellent adhesive properties. Incorporating GA into bleached kraft pulp has been shown to improve the paper's tensile strength. The addition of 3% GA to the pulp mixture *via* bulk incorporation can enhance tensile strength, yielding a 17.1% improvement in tensile index relative to the control (Ferreira *et al.* 2022). However, GA's contribution to retention and drainage remains underexploited in conventional systems. The combination of CCG and GA to form a dual-polymer system for retention and drainage holds promise for addressing some of the limitations of biomass-based dual-polymer systems.

This research investigated the impact of individual CGG and GA additives on retention and drainage efficiency, ultimately leading to the development of a CGG-GA biomass-based dual-polymer system for retention and drainage. This novel system not only enhances drainage and retention but also positively affects the physical characteristics of handsheets. Its application in paper production offers an innovative approach to develop an additive system specifically designed for papers requiring food contact, leveraging its natural polysaccharide composition and reduced leaching risk to address regulatory guidelines on chemical migration. This study aims to provide a more sustainable and food-safe alternative for the paper industry while simultaneously improving paper performance (21 CFR §184.1330).

EXPERIMENTAL

Equipment

The experimental equipment comprised specialized instruments for each processing stage. Fiber disintegration was performed using a laboratory Hollander beater (FFiber-Beating 30; Guangdong Fiber Technology Research Co., Ltd., China). Drainage properties and retention rate were quantified *via* a dynamic retention/drainage jar (DFR-05; BTG PLC, Eclépens, Switzerland). Handsheets were formed according to ISO 5269-1 standards using a Messmer 255 former (Testing Machines Inc., USA). Zeta potential of fiber suspensions was measured using a zeta potential analyzer (SZP-10; BTG PLC, Eclépens, Switzerland). Material characterization included: air permeability measurements (L&W 166; Lorentzen & Wettre, Kista, Sweden), SEM imaging (SU5000; Hitachi High-Tech, Tokyo, Japan), thickness profiling (Schopper-type YQ-Z-13; Hangzhou Nodin, China), tensile strength testing (L&W CE062; Lorentzen & Wettre, Sweden), and formation analysis (2D LAB F/SENSOR; Techpap, Gières, France).

Materials

Cationic guar gum (2×10^6 Da, 0.35 meq/g) was supplied by Cargill (USA). Gum arabic (2×10^5 Da, -0.05 meq/g) was supplied by Nexira (France). Bleached softwood pulp was sourced from Canfor (Canada). Bleached hardwood pulp was sourced from Fibria (Brazil). Fiber dimensions and fines content were measured using an Morfi Compact Techpap Fiber Analyzer. The essential features of pulps used in the experiment are shown in Table 1. Precipitated calcium carbonate (PCC, with particle sizes in the range 0.1 to 1 μm) provided by China Tobacco Mauduit (Jiangmen) Paper Industry Co., Ltd.

Table 1. Properties of the Pulps Utilized in the Experiment

Parameter	Bleached Softwood Pulp	Bleached Hardwood Pulp
Fiber length*; mm	0.713	0.697
Fiber width; μm	23.0	17.9
Coarseness; mg/m	0.1160	0.0074
Fines; %	6.83	6.92

*Weighted fiber length

Methods

Beating

Beating was performed using a laboratory Hollander beater at a consistency of 1.57% with 23 L pulp volume. Softwood pulp with a Schopper-Riegler Degree of 90 °SR and hardwood pulp with a Schopper-Riegler Degree of 35 °SR were obtained.

Measurement of drainage time

The process began by preparing a 1000 mL stock suspension at 1 wt% total solids concentration in a defibrizer for 8,000 rpm. The fiber furnish comprised a 50:50 blend of softwood and hardwood pulp (dry weight basis), with 30% PCC added as filler. The prepared suspension was transferred to a dynamic retention/drainage jar equipped with a 60 mesh/0.17 mm filter and drainage module. Once the first additive was introduced, the dynamic retention/drainage jar started by operating at 700 rpm for 10 s. Following this interval, the second additive was injected prior to secondary shearing at 800 rpm for 10 s, and then water filtration commenced. Subsequently, the device autonomously logged both

the filtration duration and the filtrate's quality. Drainage performance was evaluated through the temporal progression of filtrate mass accumulation, recorded automatically by the dynamic retention/drainage jar 's gravimetric sensor (precision: ±0.1 g).

Measurement of filler/fines retention

The pulp was configured in the same way as the measurement of drainage time, and the supporting equipment for the dynamic retention/drainage jar was replaced with a 24 mesh/0.35 mm filter and a retention module. Once the first additive was introduced, the dynamic retention/drainage jar started by operating at 700 rpm for 10 s. Following this interval, the second additive was injected prior to secondary shearing at 800 rpm for 10 s. Upon opening the valve, the instrument automatically determined the overall retention and filler/fine fiber retention. The retention rate is measured in accordance with the TAPPI T261 cm-94 standard. The specific calculation formula is as follows,

$$\text{Retention } (\%) = (1 - \frac{C}{C_0}) \times 100 \quad (1)$$

where C_0 is the initial solid concentration (g/L) of the pulp before filtration; C is the solid content concentration (g/L) in the filtrate. C is determined by the Mütek^{TM}RET-20 laboratory concentration module of the dynamic retention/drainage jar (DFR-05) using the optical peak method.

Preparation of handsheets and measurement of properties

The handsheets with a target 60 g/m^2 grammage were formed using a MESSMER 25 handsheet former following the TAPPI T205 (2024) standard method. The fiber furnish comprised a 50:50 blend of softwood and hardwood pulp (dry weight basis), with 30% PCC added as filler. Blank control samples were prepared identically without any additives. Given consistent pulp composition across all specimens, subsequent sample nomenclature refers specifically to additive type and dosage (*e.g.*, '0.05% CGG' denotes sheets with 0.05% cationic guar gum). The physical properties of handsheets were determined by the following standards: ISO 536:2019 (grammage), ISO 534:2011 (thickness), ISO 1924-2:2008 (tensile strength), ISO 5636-5:2013 (air permeability).

Scanning electron microscope

The instrument used was a SU5000 scanning electron microscope (SEM) made in Japan, with a set accelerating voltage of 3.0 kV, and secondary electron signal imaging. Before imaging, the samples were fixed with conductive adhesive, then sprayed with gold. The experimental equipment was debugged according to the instructions, and the SEM images were observed and photographed at the appropriate magnification.

RESULTS AND DISCUSSION

Impact of Biomass-Based Additives on the Drainage and Retention Properties of Pulp

Impact on retention performance

Figure 1 illustrates how varying levels of CGG, GA, and dual polymer system impacted the retention of filler. Solely incorporating CGG enhanced retention, as its cationic groups were able to create flocs with anionic elements in the pulp, thereby

retaining cellulosic fines and filler particles. Cationic guar gum, known for its strong hydrophilicity and bridging ability, interacts electrostatically with PCC particles and other negatively charged substances, improving their retention in the suspension. The CGG-GA dual-polymer system showed the most significant improvement in retention over the two single additives, and the maximum retention of 73.9% was achieved at an additive composition of 0.15% CGG-0.15% GA (total 0.30%). This occurs because in the dual-polymer system, CGG initially created sizable flocs with delicate fibers and fillers, scattered into tinier flocs due to increased shear force. Subsequently, the anionic groups on the molecular chain of the introduced gum arabic, namely COO^- (Isobe *et al.* 2020), joined with the unattached cationic sites on the CGG's molecular chain, resulting in a composite structure with the smaller flocs and enhancing their strength. This enhancement boosted the robustness and steadiness of the flocs, thereby significantly boosting the retention efficiency of fillers and fine fibers.

It is worth noting that in Fig. 1 there were slight fluctuations in retention at certain dosages. Despite minor experimental deviations, the overall trend remains clear, and the dual-polymer system showed significantly better retention performance than individual additives.

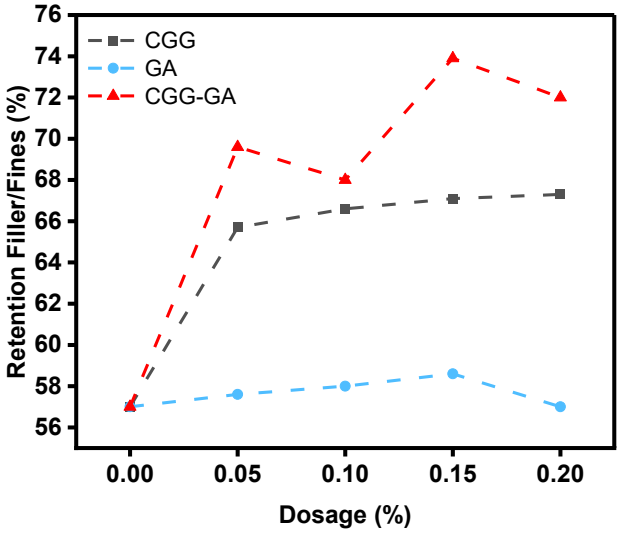

Fig. 1. Variation of filler/fines retention with additive dosage (%, w/w per component)

Impact on drainage performance

Figure 2 displays a comparative analysis of optimal water filtration curves for CGG, GA, and dual-polymer system treated pulp, along with the duration required to gather 700 mL of filtrate. The diagram illustrates that, in terms of enhancing drainage efficiency, the filtering assistance provided by individual additives—whether CGG or GA—was limited. Enhanced drainage efficiency was achieved through the dual-polymer system, where the cationic moieties of CGG electrostatically neutralized the surface anionic charges on fibers and fillers, diminished electrostatic repulsion, and encouraged particle aggregation, thereby boosting drainage efficiency. Concurrently, the extended molecular chains of CGG bridged fibers and fillers *via* polymer entanglement, forming larger flocs that enhanced drainage efficiency by facilitating rapid water removal through interconnected pores. The GA is capable of enhancing the flocs' robustness and steadiness, amplifying intermolecular interactions, and amplifying the impact of retention and drainage support. Furthermore,

incorporating GA increases the pulp's bulk, improves its flow properties, and promotes the aggregation of fibers and fillers.

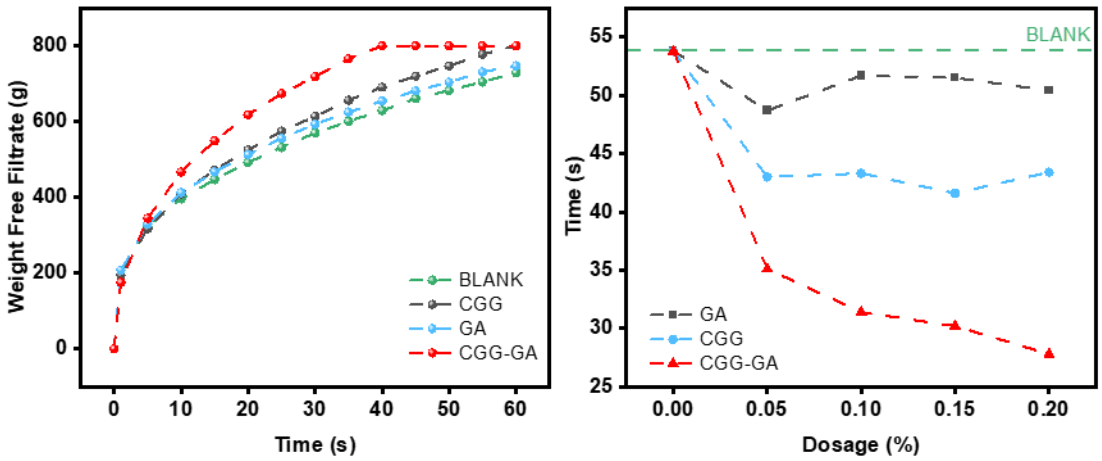

Fig. 2. Variation of filtrate weight with time (a) and time taken to collect 700 mL of filtrate (b)

Analysis of the Mechanism of CGG-GA Dual-polymer System

Zeta potential measurements were performed on pulp samples treated with CGG, GA, and their combination to better understand the charge distribution and the mechanisms of retention and drainage. The unmodified pulp had an initial zeta potential of -30.9 mV. Upon the addition of CGG, the zeta potential increased with rising dosages, indicating the positive charge of CGG, which enhances retention by promoting stronger interactions between the pulp fibers and the cationic polymer. In contrast, GA, an anionic polymer, led to a slight decrease in zeta potential, reflecting the negative COO^- groups in GA.

When CGG and GA were combined, the zeta potential stabilized at around 0.15% CGG-0.15% GA, which corresponded to the optimal retention and drainage performance observed. This suggests a synergistic effect between the two polymers, where CGG, with its positive charge, attracts negatively charged fines and fillers, while GA stabilizes the flocs formed by CGG. This balance of charges in the dual-polymer system enhanced both retention and drainage efficiency, with the optimal zeta potential values supporting the improved performance of the pulp system.

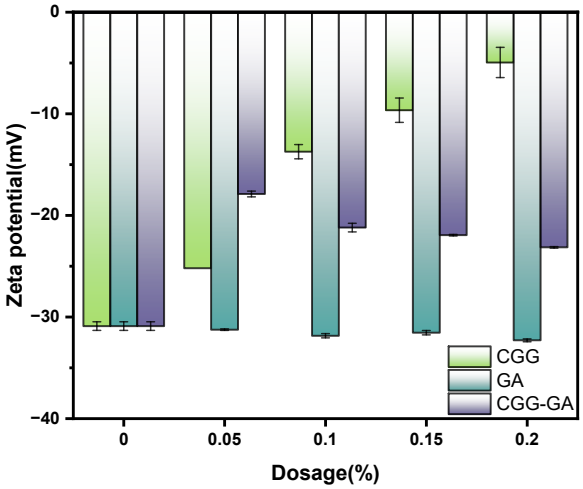

Fig. 3. Change of zeta potential with additive dosage (%, w/w per component)

Impact on Handsheets

SEM analysis

The SEM images reveal the microscopic morphology of the paper's surface after the addition of various additives. Observations revealed that without any additive, PCC was spread evenly over the surfaces of paper's fibers without noticeable flocculation. When GA was introduced, the spreading of PCC across the paper's surface became more pronounced due to the repulsion of anionic groups in GA, a result of the negative charges from both the fine fibers and the filler. Following the addition of CGG, calcium carbonate flocculation emerged on the handsheets' surface. This is attributed to CGG generating a positive charge in water, counterbalancing the negative charge between some filler and fine fibers. Consequently, particle repulsion diminished, leading to particle cohesion during collisions. Subsequently, these flocs were transformed into smaller flocs due to shear, but once the shear force vanished, they were able to reassemble. Under the combined influence of CGG-GA, the handsheets displayed clear flocs on their surface, composed of calcium carbonate and fine fibers, yet the spread across these flocs was more even. While SEM provides high-resolution morphological insights, the analyzed areas represent <0.01% of the total sheet surface. This localized sampling may not capture bulk heterogeneity, particularly in paper where fiber distribution varies spatially. Nevertheless, comparative analysis of these microstructural variations rationalizes the systematic trends in tensile strength and air permeability observed macroscopically (Fig. 4).

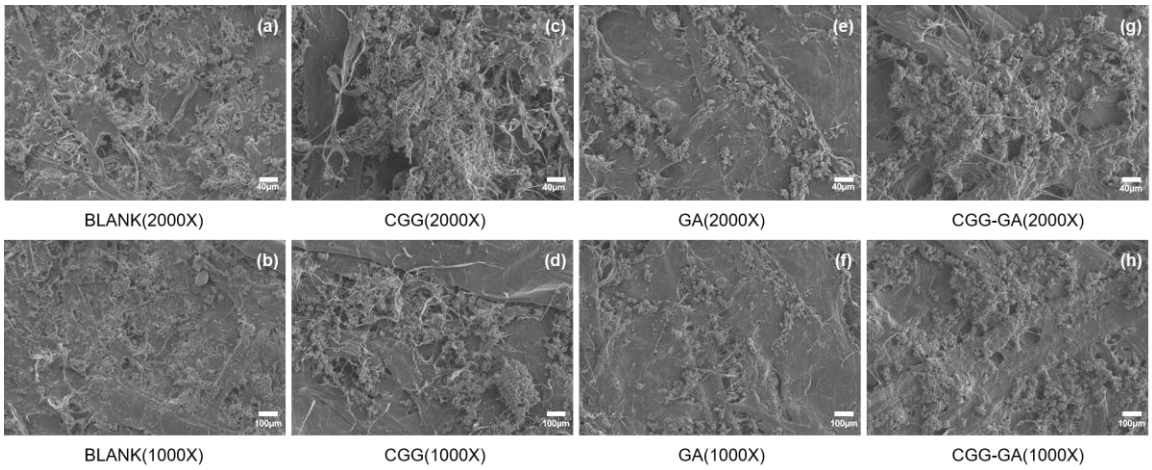

Fig. 4. SEM images of handsheets under different additive treatments

Impact on formation uniformity

Retention and drainage processes often lead to subpar paper uniformity due to flocculation creating flocs on the paper's surface. The elevated light transmission (L.T) of CGG-added paper sheets suggests a decline in uniformity, aligning with prior research findings. Paper treated with GA exhibited a minor rise in uniformity relative to the control group, due to the uniform distribution of calcium carbonate and fine fibers on the paper's surface, a result of their enhanced mutual repulsion. The presence of flocs led to a reduction in the paper's uniformity within the dual-polymer additive system relative to the control group, which is inevitable. Moreover, fiber flocculation itself plays a critical role in determining sheet formation quality. In addition to the aggregation of fines and fillers, polymer-induced fiber–fiber interactions can lead to the formation of fiber flocs, which have a more pronounced impact on paper uniformity. Higher polymer dosages or high

molecular weight additives may promote fiber bridging, forming larger flocs that deteriorate formation. However, the enhanced uniformity, as opposed to just single CGG, suggests that introducing GA intensifies the repulsion among the flocs, allowing for a slight dispersion among them.

Despite the increased flocculation observed with the CGG–GA dual-polymer system, paper formation was nevertheless improved compared to that with CGG alone. This seemingly paradoxical outcome can be explained by the formation of compact, shear-resistant flocs stabilized by electrostatic interactions between CGG and GA. These stable flocs were less susceptible to breakage and re-flocculation during the drainage process, resulting in a more uniform distribution of fibers and fillers. Thus, the enhanced formation in the dual-polymer system is not due to a reduction in flocculation, but rather to improved floc stability and moderate dispersibility, which together promote better spatial uniformity in the final sheet structure.

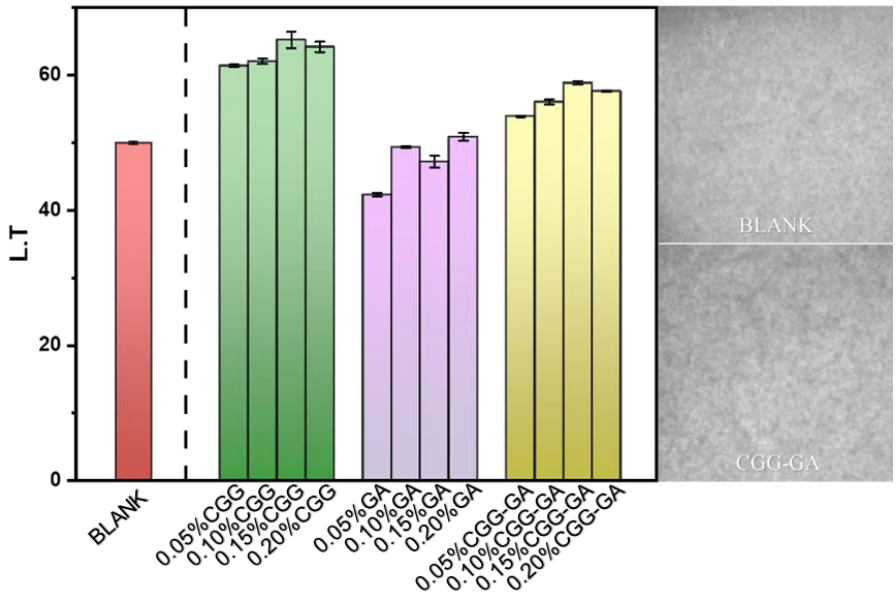

Fig. 5. Formation variation of handsheets under different additive treatments and images taken by the formation tester

Impact on tensile strength

The GA polymer, having a rich content of hydroxyl groups (Grein-Iankovski *et al.* 2018), can establish hydrogen bonds with the cellulosic surfaces after addition to pulp, thereby reinforcing the tensile strength when mixed with GA. Adding CGG led to a reduction in the tensile index. The addition of retention aid promotes the flocculation of fillers and fine fibers, yet improper control of this flocculation can result in the development of large and uneven flocs. The integration of these flocs with the fiber network might be suboptimal, leading to a paper structure that is irregular. The irregular distribution of flocs in paper can lead to points of stress concentration within the paper when it is subjected to tensile stress. This can heighten its susceptibility to damage and reduce its tensile strength. With the dual-polymer additive system, the tensile index of the paper increased and exceeded that of the blank and CGG-added groups. Compared to the addition of a single CGG additive, a composite structure is created between gum arabic and the flocs due to the dual-polymer system's influence. Anionic groups on the gum arabic

molecular chain cause increased repulsion among the flocs, causing a more uniform spread of the flocs and a more consistent paper structure. Simultaneously, the residual hydroxyl groups along the molecular chain of gum arabic engage in hydrogen bonding with plant fibers, thereby enhancing the strength of the handsheets. Furthermore, when used alone, the negatively charged gum arabic tends to act as a dispersant for the mineral filler, thereby lowering the amount of PCC in the paper, which may also contribute to the observed increase in tensile strength.

It should be noted that fiber orientation was not controlled in this study. All handsheets were prepared under the same forming conditions, and thus tensile strength data are presented for relative comparison between treatments rather than as absolute indicators of mechanical performance.

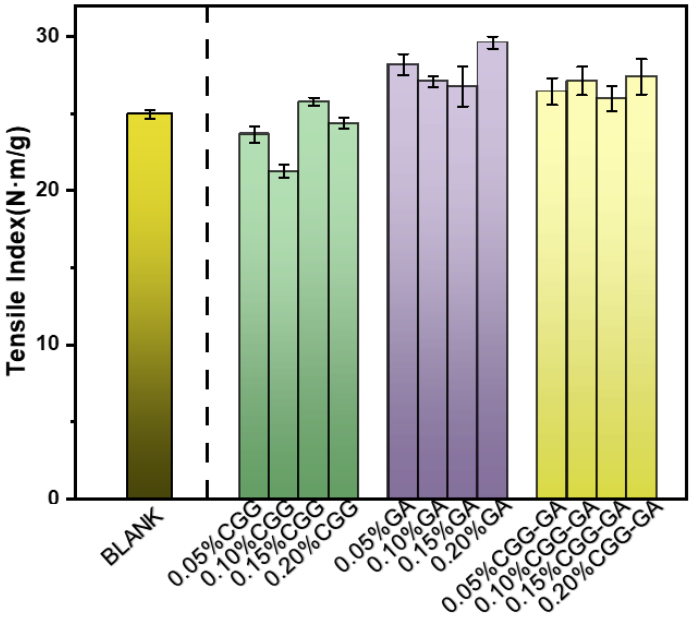

Fig. 6. Tensile strength variation of handsheets under different additive treatments

Impact on air permeability

Following the introduction of CGG, the uneven clustering of fillers and fine fibers can lead to localized blockages in the fiber network's pores. The reduction in air permeability upon the incorporation of GA suggests that the presence of its anionic groups facilitated a more refined and even dispersion of fillers throughout the paper, culminating in a denser structure.

The CGG-GA dual-polymer system combines the characteristics of the two additives and obtains a balance between them. Once GA attaches to the cationic sites of CGG, flocs composed of CGG, fine fibers, and filler flocs develop into more robust flocs. Due to the presence of anionic groups on the gum arabic molecular chain and the inherent negative charge of the filler, electrostatic repulsion occurs among the flocs, promoting a more uniform distribution of these aggregates within the paper matrix.

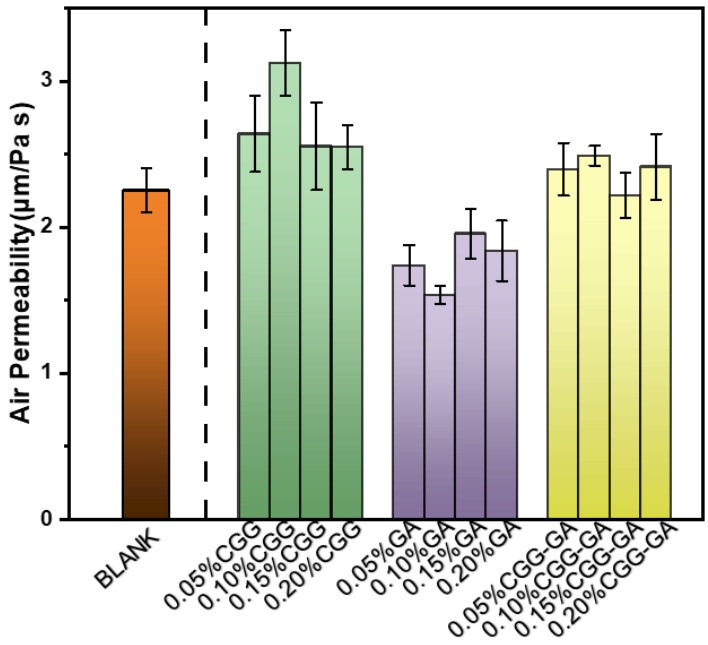

Fig. 7. Air permeability variation of handsheets under different additive treatments

CONCLUSIONS

1. The biomass-based cationic guar gum – gum arabic (CGG-GA) dual-polymer retention and drainage aid system, comprising 0.15% CGG and 0.15% GA, demonstrated superior retention performance and improved pulp drainage efficiency. This was attributed to the synergistic interplay between CGG and GA, which effectively flocculated fine fibers and fillers and stabilized the resulting flocs, thereby boosting the retention rate.
2. The zeta potential measurements revealed that the combination of CGG and GA led to an optimal zeta potential, indicating a stable interaction between the cationic CGG and anionic GA. This synergistic effect improved the retention and drainage properties by stabilizing the flocs formed during the retention and drainage processes. The optimal performance observed was directly linked to the charge balance between CGG and GA, as evidenced by the zeta potential data.
3. The addition of GA promoted the formation of hydrogen bonds with cellulose, thereby enhancing paper tensile strength. The CGG-GA system not only optimized the paper structure, but it also achieved a more uniform dispersion of flocs, resulting in a more even distribution of calcium carbonate and fine fibers on the paper surface compared to using CGG alone. Through balancing the properties of CGG and GA, the system formed robust flocs through electrostatic repulsion, leading to a more consistent distribution of aggregates within the paper matrix.

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