Effect of Polyelectrolyte Molecular Weight and Structure on the Rheological Properties of Cellulose Nanofibril Suspensions

Jaeho Ryu,[a] and Hye Jung Youn [b,c,*]

The effect of polyelectrolyte addition on the rheological behavior of cellulose nanofibril (CNF) suspensions was examined at a CNF consistency of 1.5%. Cationic and anionic polyelectrolytes with different molecular weights and structures were employed, and rheological behavior was evaluated using steady shear and strain-controlled measurements, with particular emphasis on viscosity hysteresis and yield stress as indicators of network deformation and recovery. At this consistency, steady-state viscosity exhibited little response to polyelectrolyte addition. In contrast, viscosity hysteresis and yield stress showed strong dependence on polyelectrolyte molecular weight and structure, reflecting differences in CNF network deformation and reformation behavior. High molecular weight polyelectrolytes restricted nanofibril mobility and altered network recovery after shear, whereas low molecular weight polyelectrolytes acted through electrostatic interactions. Branched cationic polyelectrolytes enhanced network strength, as evidenced by increased yield stress, while linear and anionic polyelectrolytes reduced network strength by limiting effective inter-fibrillar contacts. These results indicate that, at a CNF consistency of 1.5%, rheological responses are governed predominantly by network-scale interactions associated with polyelectrolyte molecular weight and architecture rather than by surface charge alone. These findings clarify the deformation and recovery behavior of CNF suspensions under conditions relevant to practical handling and processing.

DOI: 10.15376/biores.21.2.4045-4056

Keywords: Nanofibrillated cellulose; Nanocellulose suspensions; Highly associated suspensions; Rheological property; Thixotropy; Yield stress

Contact information: a: R&D Center, Greeneple Inc., 21 Kentech-ro, Naju, 58330, Korea; b: Department of Agriculture, Forestry and Bioresources, College of Agriculture and Life Sciences, Seoul National University, 1, Gwanak-ro, Gwanak-gu, Seoul, 08826, Korea; c: Research Institute of Agriculture and Life Sciences, Seoul National University, 1, Gwanak-ro, Gwanak-gu, Seoul, 08826, Korea;
** Corresponding author: page94@snu.ac.kr*

INTRODUCTION

Cellulose nanofibrils (CNFs), also known as nanofibrillated cellulose, have attracted considerable attention as a promising, renewable material for industrial applications since their introduction in the early 1980s (Herrick *et al*. 1983; Turbak *et al*. 1983). The CNFs are produced by several mechanical processes, including high-pressure homogenization, microfluidization, and grinding, often combined with chemical or enzymatic pretreatments (Pääkkö *et al*. 2007; Nakagaito and Yano 2008; Siró and Plackett 2010). Mechanical fibrillation typically yields nanofibrils with widths of 10 to 100 nm and lengths of several micrometers, forming an entangled, percolating network due to their high aspect ratio (Broadbent and Hammersley 1957; Pääkkö *et al*. 2007).

Because of their network structure and strong hydrophilicity, CNF suspensions exhibit high viscosity at low shear rates and pronounced shear-thinning behavior with increasing shear rate (Ono *et al*. 2004; Iotti *et al*. 2011; Hubbe *et al*. 2017; Xu *et al.* 2024). Similar rheological behavior is observed in pulp fiber suspensions, where flocculation and network restructuring under shear govern macroscopic flow properties (Bennington *et al*. 1990; Hubbe 2007). In such systems, cationic polyelectrolytes are commonly used to control flocculation *via* electrostatic interactions or polymer bridging, depending on molecular weight and charge density (Lindström 1989; Rojas and Hubbe 2004).

The rheological properties of cellulose fiber suspensions are strongly influenced by individual fiber characteristics, such as elasticity and aspect ratio, which directly influence the modulus and yield stress of the network (Tatsumi *et al*. 2002; Youn and Lee 2002). However, most previous rheological studies have focused on the linear viscoelastic regime or low shear conditions, where the network structure remains largely intact (Lowys *et al*. 2001; Pääkkö *et al*. 2007; Agoda-Tandjawa *et al*. 2010). Consequently, the dynamic restructuring and recovery behavior of CNF networks under steady shear is poorly characterized at elevated consistencies.

The influence of electrostatic interactions and polyelectrolyte addition on CNF dewatering behavior has been examined in previous studies. In the authors' earlier work, CNF dewatering under vacuum and pressure conditions was systematically investigated, demonstrating that optimal dewatering occurred when the zeta potential approached the isoelectric point through controlled addition of cationic polyelectrolytes (Ryu *et al*. 2014). That study clarified the electrostatic basis of CNF dewatering but did not address the rheological response or network dynamics of CNF suspensions under shear.

In contrast, this study focuses on the rheological behavior of CNF suspensions under conditions where network interactions are pronounced. Most rheological studies on CNF have been conducted in dilute or semi-dilute regimes (typically ≤ 0.5%), where inter-fibrillar interactions are relatively weak and structural rearrangements can be readily isolated. The CNF suspensions at higher consistencies (approximately 1.0 to 1.5%), which are more relevant to industrial handling, transport, and processing, exhibit stronger network interactions, yet their response to polyelectrolyte addition is not established.

At 1.5% consistency, CNF suspensions form a strongly interconnected network in which nanofibril–nanofibril contacts are frequent, while remaining below the regime where macroscopic jamming or solid-like behavior dominates. Under such conditions, it remains unclear whether electrostatic interactions alone are sufficient to explain rheological modification, or whether steric constraints and molecular entanglement introduced by high-molecular-weight or structurally complex polyelectrolytes play a dominant role.

The objective of the present study is therefore to determine the effect of polyelectrolyte molecular weight and structural characteristics on the rheological behavior of CNF suspensions at a consistency of 1.5%. The CNF suspensions containing various cationic and anionic polyelectrolytes were investigated using steady shear and strain-controlled measurements. Particular attention was paid to viscosity hysteresis and yield stress as indicators of network deformation, recovery, and integrity under shear. Through separating electrostatic effects from network-scale rheological behavior, this study aimed to identify the dominant factors governing the deformation and recovery of CNF networks under high-consistency conditions.

EXPERIMENTAL

Materials

The CNFs were prepared from hardwood bleached kraft pulp (Hw-BKP) through mechanical treatment. As a pretreatment, the pulp was beaten to a freeness of 450 mL CSF using a laboratory Hollander (Valley) beater. The beaten pulp suspension (2%) was then subjected to nanofibrillation using a grinder (Super Masscolloider, Masuko Sangyo Co., Kawaguchi, Japan) operated at a stone gap of −50 μm for 15 passes. The resulting CNF suspension consisted mainly of nanofibrils with an average width of approximately 50 nm. Polyelectrolytes with different molecular weights, charge densities, and molecular structures were used to modify the CNF network structure. The characteristics of the polyelectrolytes are summarized in Table 1. All polyelectrolytes were prepared as 1 wt% aqueous solutions using deionized water.

Table 1. Properties of Polyelectrolytes

Structure	Polyelectrolyte	ID	Molecular Weight, M_w (g/mol)	Charge Density (meq/g)	Manufacturer
Branched cationic	Polyethyleneimine	LmPEI	~2,000	+6.4 to 6.5 @ pH 6[a]	Sigma-Aldrich, USA
		HmPEI	~750,000		
Linear cationic	Poly(diallyldimethyl ammonium chloride)	Poly-DADMAC	400,000 to ~500,000	+6.1[b]	Sigma-Aldrich, USA
	Cationic -polyacrylamide	CPAM	2,000,000	+2.5[a]	Songkang, Korea
Linear anionic	Poly(sodium 4-styrene sulfonate)	LmPSS	200,000	-4.7[a]	Sigma-Aldrich, USA
		HmPSS	1,000,000		

[a] Measured by PCD-02
[b] According to the supplier

Addition of Polyelectrolyte to CNF Suspension

Polyelectrolyte solution with a concentration of 1 wt% was added to the 2% CNF suspension under mechanical agitation using an overhead impeller stirrer at 1300 rpm. After addition, the suspension consistency was adjusted to 1.5%. Because polyethyleneimine (PEI) is pH-sensitive, CNF suspension containing PEI were adjusted to pH 6. Other polyelectrolytes, relatively insensitive to pH, were added to the CNF suspension at pH 7. Polyelectrolyte dosage was varied at 1, 3, 5, 7, and 10 wt% based on the oven-dried weight of CNF.

Measurement of Rheological Properties

The steady-state rotational viscosity was measured using a stress-controlled Bohlin rheometer (Malvern Instruments, Malvern, UK) with parallel-plate geometry (40 mm diameter). The plate gap was 1 mm. The shear rate was swept from 0.07 to 100 s^{-1}. The viscosity at the midpoint shear rate (1 s^{-1}) during the entire sweep was used to evaluate the thixotropic index, as defined in Eq. 1,

$$\text{Thixotropic index} = A - B \tag{1}$$

where A is the viscosity at 1 s^{-1} while shear rate increases and B is the viscosity at 1 s^{-1} while shear rate decreases. In rheological measurements of fibrous or nanofibrillar suspensions using smooth parallel-plate geometries, wall slip or fiber migration near the plate surfaces may occur, potentially leading to underestimation of viscosity. In the present study, the relatively high CNF consistency (1.5%), small plate gap (1 mm), and pre-equilibration under the measurement geometry were adopted to minimize such effects.

Oscillatory rheology was performed using a strain-controlled rheometer (ARES, TA Instruments, New Castle, DE, USA) with parallel-plate geometry (25 mm diameter). The plate gap was approximately 1 mm. Prior to each measurement, the samples were equilibrated for 5 min under the measurement geometry to ensure structural equilibration. Strain sweep tests were conducted from a strain range of 0.1 to 300% at a fixed angular frequency of 1 Hz. The yield stress and yield strain were determined from the strain sweep curves (Fig. 1). The yield stress was determined using the tangent intersection method. Tangent lines were fitted to the initial linear region and the subsequent shear-thinning region of the curve, and their intersection was defined as the yield point. The stress and strain at this intersection were defined as the yield stress and yield strain, respectively.

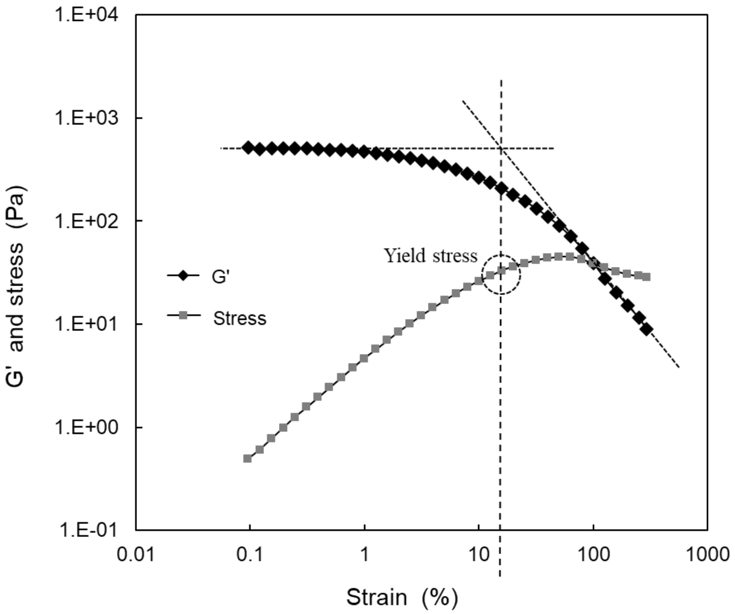

Fig. 1. Representative strain sweep curve

RESULTS AND DISCUSSION

Effect of Polyelectrolyte on Viscosity of CNF Suspension

The CNF suspensions exhibited shear-thinning behavior due to shear-induced rearrangements of the fibrillar network, as widely reported (Lowys *et al.* 2001; Agoda-Tandjawa *et al.* 2010; Iotti *et al.* 2011). Similar rheological responses in pulp suspensions were attributed to network restructuring under shear (Chen *et al.* 2002). Because polyelectrolyte addition alters inter-fibrillar interactions, its effect on CNF viscosity was examined, and the results for cationic and anionic polyelectrolytes are shown in Fig. 2.

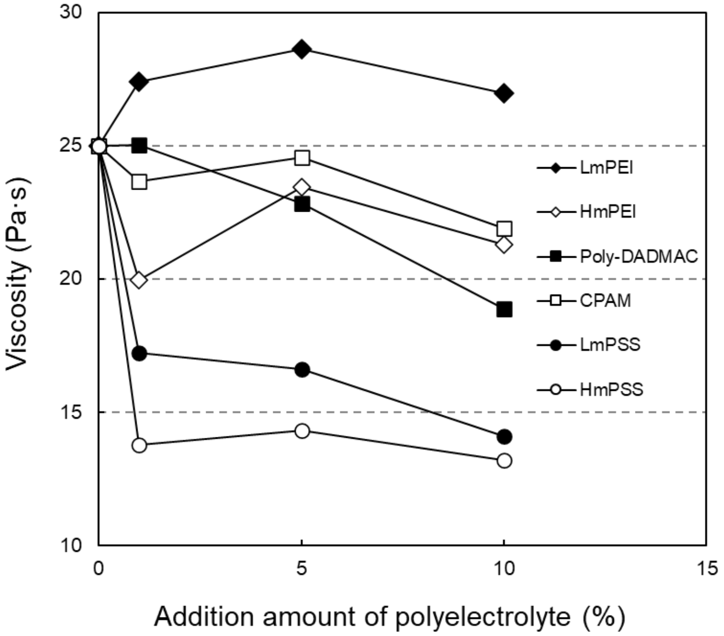

Fig. 2. Viscosity of the CNF suspension with addition of polyelectrolytes at the shear rate of 1 s^{-1}

Overall, the viscosity at the shear rate of 1 s^{-1} showed only minor changes with increasing polyelectrolyte addition. This weak dependence was attributed to the relatively high consistency (1.5%) used in this study, where CNF formed a highly associated and compact network structure. The influence of polyelectrolyte-induced flocculation on viscosity is reduced compared to lower consistency systems. In contrast, Karppinen *et al.* (2011) reported increased viscosity with polyelectrolyte addition at 0.5% consistency. Despite the limited overall variation, clear molecular weight-dependent trends were observed. The addition of low molecular weight PEI (LmPEI) largely maintained viscosity, consistent with limited polymer bridging and enhanced local interactions through patch attraction, which preserved CNF flocs as network fragments. In contrast, high molecular weight PEI (HmPEI) reduced viscosity, suggesting that steric effects from extended polymer chains outweighed electrostatic attraction. At low dosage, high-molecular-weight cationic polyelectrolytes may form large but fragile floc structures separated by a low-viscosity continuous phase, leading to reduced apparent viscosity. Such broken-floc behavior is less likely for LmPEI because of its small molecular size, which limits bridging; instead, its action is consistent with localized charged-patch interactions.

Anionic polyelectrolytes caused a more pronounced decrease in viscosity, especially at higher molecular weight. In the absence of electrostatic attraction, these polymers likely induced electrostatic and steric stabilization of the suspension, reducing effective inter-fibrillar contacts and weakening the CNF network. They may also have acted as lubricants between closely spaced nanofibrils, consistent with previous reports (Peterson 1994; Beghello 1998). The CPAM showed only a minor effect on viscosity, while linear poly-DADMAC may have induced limited bridging; however, excessive charge compensation reduced surface charge heterogeneity and inter-fibrillar friction. Overall, these results demonstrated that under high consistency conditions, the viscosity response of CNF suspensions to polyelectrolyte addition was governed primarily by molecular weight rather than charge alone, reflecting the dominant role of steric effects within a highly associated network structure.

Effect of Polyelectrolyte on Hysteresis

The CNF suspensions exhibited shear-thinning behavior under steady-state flow conditions. Although this behavior has been widely reported (Ono *et al.* 2004; Lasseuguette *et al.* 2008; Iotti *et al.* 2011), viscosity hysteresis between increasing (up-curve) and decreasing (down-curve) shear rates has received relatively limited attention, despite its importance for understanding the deformation and reformation of CNF network structures (Iotti *et al.* 2011). In this study, a CNF suspension with a consistency of 1.5% was examined. The viscosity decreased logarithmically with increasing shear rate. In contrast to previous studies conducted at lower consistency (≈0.5%), the characteristic "shoulder" in the flow curve was not clearly observed. Earlier studies reported shoulders in the shear-rate ranges of 0 to 50 s^{-1} or 2 to 20 s^{-1}, which were attributed to structural transitions within the CNF network (Lasseuguette *et al.* 2008; Iotti *et al.* 2011; Karppinen *et al.* 2011). The absence of a distinct shoulder here is attributed to the higher consistency, which leads to a more compact and highly associated network. Consequently, structural evolution was evaluated through viscosity hysteresis rather than shoulder behavior.

Viscosity hysteresis developed during shear-rate ramping, with the down-curve lying below the up-curve due to energy dissipation associated with network deformation at high shear rates. Hysteresis was quantified by the gap between the up- and down-curves at 1 s^{-1}. Flow curves of CNF suspensions containing 5% PEI with different molecular weights are shown in Fig. 3.

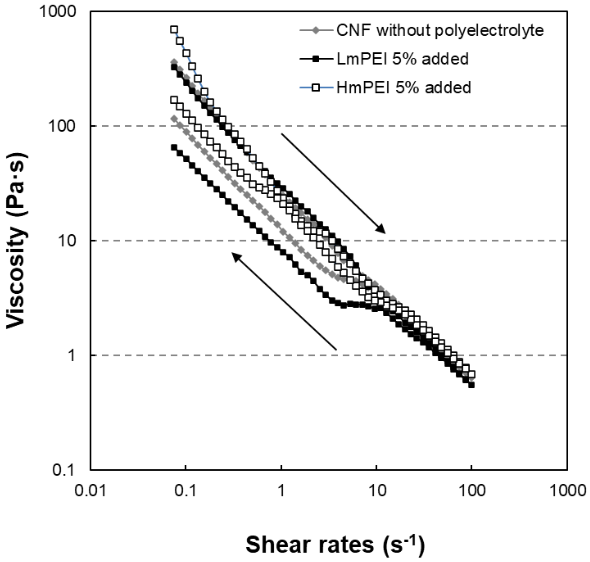

Fig. 3. Viscosity of the CNF suspension and that by addition of PEI with 5% and different molecular weight

At 1 s^{-1}, the hysteresis gap varied strongly with polyelectrolyte type. The thixotropic index of the neat CNF was 12.5 Pa·s. After the addition of 5% low molecular weight PEI (LmPEI), the index increased to 20.6 Pa·s, whereas the addition of 5% high molecular weight PEI (HmPEI) reduced it to 2.5 Pa·s. Because the up-curves were similar for all samples, the hysteresis behavior was primarily governed by the down-curves. A higher down-curve corresponded to a smaller hysteresis, indicating faster structural recovery. Overall, viscosity hysteresis provides insight into the reformation dynamics of the CNF network. A smaller thixotropic index reflects more rapid structural recovery after shear, suggesting efficient reformation of the CNF network once nanofibrils were aligned along the flow direction at high shear rates. The pronounced reduction in hysteresis for certain systems also suggests that shear disrupted electrostatic charged-patch interactions within the CNF network during flow. Subsequent recovery behavior therefore depended on the mobility and molecular characteristics of the added polyelectrolyte. Such behavior is consistent with reported network restructuring and flocculation in nanocellulose suspensions (Hubbe *et al.* 2017).

The thixotropic indices obtained for different polyelectrolytes are summarized in Fig. 4. The effect of PEI molecular weight is shown in Fig. 4(a). LmPEI is a branched cationic polyelectrolyte with a low molecular weight, whereas HmPEI is branched with a high molecular weight. A pronounced difference in hysteresis was observed, particularly at 5% addition. This result indicated that molecular weight critically controls CNF network reformation under applied shear. For poly-DADMAC, increasing molecular weight progressively decreased the thixotropic index with increasing dosage. Moreover, CPAM, which has a higher molecular weight but lower charge density than poly-DADMAC, produced an even more rapid reduction.

Together, these results indicated that molecular weight played a key role in the structural reformation of CNF suspensions. In particular, the presence of high-molecular-weight polyelectrolytes restricted structural deformation by limiting nanofibril mobility within the network. At addition levels above the isoelectric point—typically above 1% (and above 5% for CPAM)—surface charge reversal occurred, and electrostatic repulsion between nanofibrils became dominant. Under these conditions, electrostatic effects alone cannot explain the hysteresis behavior. Instead, molecular weight-dependent physical constraints become increasingly important. When the molecular weight of the polyelectrolyte exceeded approximately 500,000 g/mol, physical entanglement or polymer bridging between CNF and polyelectrolyte chains was likely, leading to restricted network reformation. In contrast, LmPEI, with a molecular weight below 2,000 g/mol, lacked the ability to induce entanglement or bridging and therefore increased hysteresis primarily through electrostatic effects.

For anionic polyelectrolyte PSS, thixotropic index decreased with increased addition, as shown in Fig. 4(b). Although hysteresis also decreased for cationic polyelectrolytes, the underlying mechanism for PSS is fundamentally different and is primarily associated with electrostatic repulsion between CNF and PSS. Because PSS is assumed not to adsorb efficiently onto negatively charged CNF, it did not contribute to flocculation or network reinforcement. Instead, PSS occupied the continuous phase and modifies the local environment between nanofibrils. Electrostatic repulsion prevented close association between CNF and PSS chains, while the polymer chains occupied the interstitial space within the suspension. This effect hindered inter-fibrillar contact and slowed the reformation of the CNF network after shear. As a result, the CNF network exhibited reduced hysteresis and apparent structural recovery accelerated. These observations suggest that, rather than acting as a structural component of the network, anionic polyelectrolytes, such as PSS, influence thixotropic behavior indirectly by altering inter-fibrillar interactions and spatial constraints within the suspension.

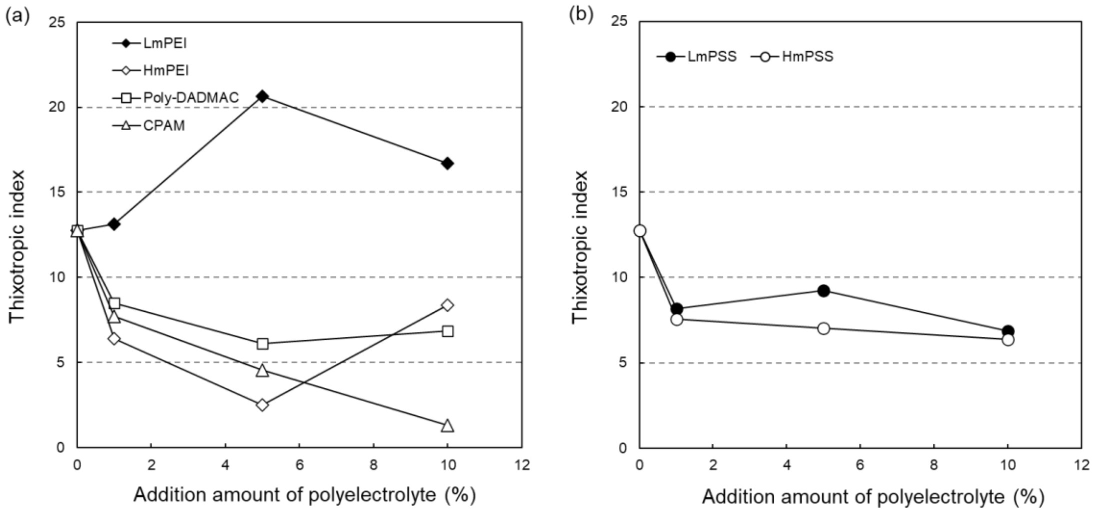

Fig. 4. Thixotropic index of the CNF suspension with (a) cationic and (b) anionic polyelectrolyte

Effect of Polyelectrolyte on Network Strength of CNF Suspension

During strain sweeps, the CNF network yielded when applied strain exceeded a critical value. Because the suspensions were subjected to mechanical agitation (1300 rpm) during polyelectrolyte addition prior to rheological testing, the measured yield stress represents a recovered network strength rather than the initial undisturbed yield stress. Network failure occurred after elastic deformation of the CNF network, indicating that yield behavior reflected the strength of the interconnected nanofibril network. Yield stress as a function of polyelectrolyte addition is presented in Fig. 5. Network failure occurred after elastic deformation of the CNF network, indicating that yield behavior reflected the strength of the interconnected nanofibril network. The mechanical strength of the CNF network is primarily governed by the interlocking of elastically bent nanofibrils, which corresponds to the floc strength of the system (Bennington *et al*. 1990). Accordingly, changes in yield stress provided insight into how polyelectrolytes modify the network structure and strength.

As shown in Fig. 5(a), branched polyelectrolytes increased network strength with increasing molecular weight, whereas linear polyelectrolytes had a weaker effect. Because electrostatic attraction between nanofibrils induced by cationic polyelectrolytes is largely suppressed at addition levels above 1%, the observed enhancement in network strength is attributed mainly to structural modification of the CNF network rather than electrostatic interactions. In this context, HmPEI induced stronger flocculation than LmPEI, leading to a higher yield stress. In contrast, linear polyelectrolytes, such as poly-DADMAC, despite its high charge density, and CPAM, which possesses high molecular weight but low charge density, resulted in weaker flocculation and lower network strength (Fig. 5(b)).

While CNF flocculation and dewatering behavior are maximized near the isoelectric point under dilute or moderately associated conditions (Ryu *et al*. 2014), the present results show that, in highly associated suspensions at 1.5% consistency, network strength was not governed primarily by isoelectric-point-driven flocculation. Instead, polymer molecular weight, architecture, and steric constraints became dominant.

This behavior implied an optimal balance between molecular weight and charge density for network reinforcement. Similar trends were reported by Swerin (1998), who observed that cellulosic fiber suspensions flocculated with cationic polyacrylamides exhibit an optimum addition amount and charge density for maximizing the elastic shear modulus. The present results indicated that this concept is also applicable to CNF systems.

When comparing network strength with respect to polyelectrolyte structure, branched polyelectrolytes produced higher yield stress than linear ones. Network strength depended on both floc strength and floc density. Polyelectrolyte adsorption *via* patch attraction or bridging determined floc density. Flat adsorption produced denser flocs than long-range bridging. Because CNF dimensions are much smaller than those of pulp fibers, the adsorption behavior depended strongly on polymer structure and charge density. For pulp fiber systems, Swerin (1998) reported that fiber flocculation is influenced by both the addition amount and the degree of substitution of the polyelectrolyte. For anionic polyelectrolyte PSS, electrostatic flocculation was not expected. High molecular weight PSS (HmPSS) reduced network strength, consistent with the viscosity results shown in Fig. 5. Thus, anionic polyelectrolytes weaken the CNF network rather than reinforcing it.

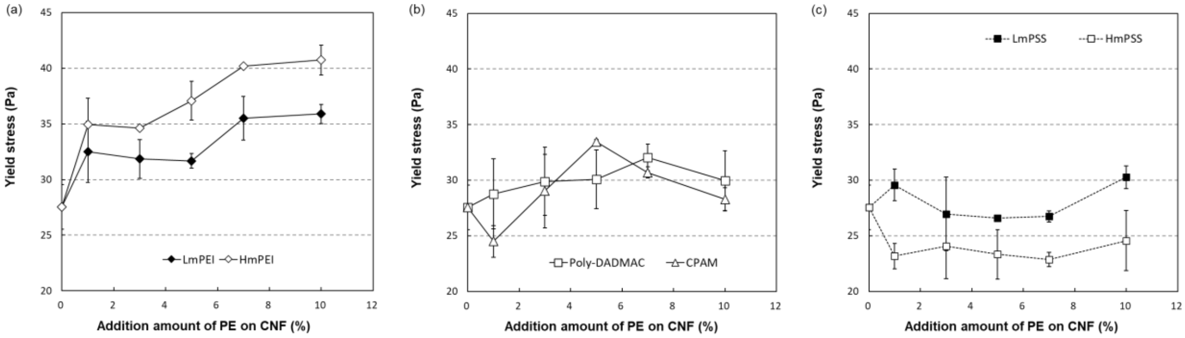

Fig. 5. Yield stress of CNF suspensions with added polyelectrolytes

CONCLUSIONS

1. The rheological behavior of highly associated cellulose nanofibril (CNF) suspensions (1.5% consistency) is governed primarily by interactions at the network scale rather than by surface charge alone. Viscosity, hysteresis, and yield stress collectively characterize the deformation and recovery dynamics of the interconnected CNF network under shear.
2. Molecular weight and structural characteristics of polyelectrolytes govern CNF network behavior. High molecular weight polyelectrolytes impose steric constraints and polymer–nanofibril entanglement, restricting nanofibril mobility and altering viscosity, hysteresis, and network strength, whereas low molecular weight polyelectrolytes act through system through electrostatic interactions.
3. Network strength, as reflected by yield stress, increases with the molecular weight of branched cationic polyelectrolytes, even under conditions where electrostatic attraction is largely suppressed. This demonstrates that, in highly associated CNF suspensions, polymer architecture and steric reinforcement dominate over isoelectric-point-driven flocculation mechanisms in determining network integrity.

ACKNOWLEDGMENTS

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (No. 2012011091).

Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

REFERENCES CITED

Agoda-Tandjawa, G., Durand, S., Gaillard, C., Garnier, C., and Doublier, J. L. (2010). "Rheological behaviour and microstructure of microfibrillated cellulose suspensions/low-methoxyl pectin mixed systems: Effect of calcium ions," *Carbohydrate Polymers* 87(2), 1045-1057. https://doi.org/10.1016/j.carbpol.2011.08.021

Beghello, L. (1998). "Some factors that influence fiber flocculation," *Nordic Pulp & Paper Research Journal* 13(4), 274-279. https://doi.org/10.3183/npprj-1998-13-04-p274-279

Bennington, C. P. J., Kerekes, R. J., and Grace, J. R. (1990). "The yield stress of fiber suspensions," *The Canadian Journal of Chemical Engineering* 68(5), 748-757. https://doi.org/10.1002/cjce.5450680503

Broadbent, S., and Hammersley, J. (1957). "Percolation processes I: Crystals and mazes," *Mathematical Proceedings of the Cambridge Philosophical Society* 53(3), 629–641. https://doi.org/10.1017/S0305004100032680

Chen, B., Tatsumi, D., and Matsumoto, T. (2002). "Floc structure and flow properties of pulp fiber suspensions," *Nihon Reoroji Gakkaishi* 30(1), 19-25. https://doi.org/10.1678/rheology.30.19

Herrick, F. W., Casebier, R. L., Hamilton, J. K., and Sandberg, K. R. (1983). "Microfibrillated cellulose: Morphology and accessibility," *Journal of Applied Polymer Science: Applied Polymer Symposium* 37, 797–813.

Hubbe, M. A. (2007). "Flocculation and redispersion of cellulosic fiber suspensions: A review of effects of hydrodynamic shear and polyelectrolytes," *BioResources* 2(2), 296-331.

Hubbe, M. A., Tayeb, P., Joyce, M., Tyagi, P., Kehoe, M., Dimic-Misic, K., and Pal, L. (2017). "Rheology of nanocellulose-rich aqueous suspensions: A review," *BioResources* 12(4), 9556-9661. https://doi.org/10.15376/biores.12.4.Hubbe

Iotti, M., Gregersen, Ø. W., Moe, S., and Lenes, M. (2011). "Rheological studies of microfibrillar cellulose water dispersions," *Journal of Polymers and the Environment* 19(1), 137-145. https://doi.org/10.1007/s10924-010-0248-2

Karppinen, A., Vesterinen, A.-H., Saarinen, T., Pietikäinen, P., and Seppälä, J. (2011). "Effect of cationic polymethacrylates on the rheology and flocculation of microfibrillated cellulose," *Cellulose* 18, 1381-1390. https://doi.org/10.1007/s10924-010-0248-2

Lasseuguette, E., Roux, D., and Nishiyama, Y. (2008). “Rheological properties of microfibrillar suspension of TEMPO-oxidized pulp,” *Cellulose* 15(3), 425-433. https://doi.org/10.1007/s10570-007-9184-2

Lindström, T. (1989). “Some fundamental chemical aspects on paper forming,” in: *Fundamentals of Papermaking*, C. F. Baker, and V. W. Punton (eds.), Mechanical Engineering Publications, London, UK, pp. 309–412.

Lowys, M. P., Desbrières, J., and Rinaudo, M. (2001). “Rheological characterization of cellulosic microfibril suspensions: Role of polymeric additives,” *Food Hydrocolloids* 15(1), 25-32. https://doi.org/10.1016/S0268-005X(00)00046-1

Nakagaito, A. N., and Yano, H. (2008). “The effect of fiber content on the mechanical and thermal expansion properties of biocomposites based on microfibrillated cellulose,” *Cellulose* 15(4), 555-559. http://doi.org/10.1007/s10570-008-9212-x

Ono, H., Shimaya, Y., Sato, K., and Hongo, T. (2004). “^{1}H spin–spin relaxation time of water and rheological properties of cellulose nanofiber dispersion, transparent cellulose hydrogel (TCG),” *Polymer Journal* 36(9), 684-694. https://doi.org/10.1295/polymj.36.684

Pääkkö, M., Ankerfors, M., Kosonen, H., Nykänen, A., Ahola, S., Osterberg, M., Ruokolainen, J., Laine, J., Larsson, P. T., Ikkala, O., and Lindström, T.. (2007). “Enzymatic hydrolysis combined with mechanical shearing and high-pressure homogenization for nanoscale cellulose fibrils and strong gels,” *Biomacromolecules* 8(6), 1934-1941. https://doi.org/10.1021/bm061215p

Peterson, D. E. (1994). “Nuclear density consistency meter evaluation,” in: *Proceedings of the TAPPI Process Control Symposium*, TAPPI Press, Atlanta, GA, USA, pp. 9-18.

Rojas, O. J., and Hubbe, M. A. (2004). “The dispersion science of papermaking,” *Journal of Dispersion Science and Technology* 25(6), 713-732. https://doi.org/10.1081/DIS-2000355485

Ryu, J., Sim, K., and Youn, H. J. (2014), “Evaluation of dewatering of cellulose nanofibrils suspension and effect of cationic polyelectrolyte addition on dewatering,” *Journal of Korea TAPPI* 46(6), 78-86. https://doi.org/10.7584/ktappi.2014.46.6.078

Siró, I., and Plackett, D. (2010). “Microfibrillated cellulose and new nanocomposite materials: A review,” *Cellulose* 17(3), 459-494. https://doi.org/10.1007/s10570-010-9405-y

Swerin, A. (1998). “Rheological properties of cellulosic fibre suspensions flocculated by cationic polyacrylamides,” *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 133(3), 279-294. https://doi.org/10.1016/S0927-7757(97)00212-4

Tatsumi, D., Ishioka, S., and Matsumoto, T. (2002). “Effect of fiber concentration and axial ratio on the rheological properties of cellulose fiber suspensions,” *Nihon Reoroji Gakkaishi* 30(1), 27-32. https://doi.org/10.1678/rheology.30.27

Turbak, A. F., Snyder, F. W., and Sandberg, K. R. (1983). “Microfibrillated cellulose, a new cellulose product: Properties, uses, and commercial potential,” *Journal of Applied Polymer Science: Applied Polymer Symposium* 37, 815-827.

Xu, J., Wang, P., Yuan, B., and Zhang, H. (2024). “Rheology of cellulose nanocrystal and nanofibril suspensions” *Carbohydrate Polymers* 324, article 121527. https://doi.org/10.1016/j.carbpol.2023.121527

Youn, H. J., and Lee, H. L. (2002). "An experimental investigation of the effect of pulp types, mechanical treatments, and crill contents on fibre network strength," *Nordic Pulp & Paper Research Journal* 17(2), 187-192. https://doi.org/10.3183/npprj-2002-17-02-p187-192

Article submitted: January 14, 2026; Peer review completed: February 28, 2026; Revised version received and accepted: March 6, 2026; Published: March 20, 2026.
DOI: 10.15376/biores.21.2.4045-4056