# Stability and Rheological Behavior of Nanocellulose-modified UF Resin Compositions

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The aim of this work was to investigate the influence of nanocellulose on the storage stability and rheological properties of urea formaldehyde (UF)-based adhesive compositions for wood-based panels. Three types of UF resins characterized by different F/U molar ratios were used for this research. Resin modifications with nanocrystalline cellulose and a nanofibrillated cellulose content of 1.0% to 5.0% by dry weight of resin were prepared. The flow curve characteristics and storage stability were studied. The viscosity values mainly depended upon the type of nanocellulose used, as well as its loading in the composition. The UF resins modified with nanocrystalline cellulose kept their rheological behavior and proper viscosity after 4 weeks in storage, which lowered the percentage of viscosity retention by approximately 1.5 times compared with that of industrial resins. Nanocrystalline cellulose might be used as a stabilizer in resin compositions during long-term storage, while nanofibrillated cellulose might act as a thickening agent through the limited extent of loading in a composition.

*Keywords: Bio-based adhesives; Nanocellulose; Stability; Rheology; UF resin*

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## INTRODUCTION

The growing demand for gaining independence from fossil resources and increasing concern for human health issues have led to increasing attention to developing ecological and renewable bioadhesives, based on starch (Sun *et al.* 2018), soy (Ghahri *et al.* 2018), liquefied wood (Janiszewska *et al.* 2016a,b; Janiszewska 2018), and plant proteins (Yang *et al.* 2006; Ferdosian *et al.* 2017), including nanoadhesives (Kojima *et al.* 2016; Khoramishad and Khakzad 2018). One of the most promising biomaterials with a wide variety of properties and functionalities is nano-scale cellulose. Since 2014, the nanocellulose market has quickly developed. Today, there are more than a dozen commercial-scale nanocellulose production facilities, as well as numerous patents for nanocellulose processing (Crotogino 2012; Olkowski and Laarveld 2013; Nelson *et al.* 2014; Miller 2015). Moreover, because of the lack of international standards for this emerging nanomaterial, the Standardization Task Group ISO/TC 6/TG 1 has been established in order to include cellulose nanomaterials specificity factor in existing TC 6 standards and at the same time to allow sustained development and applications.

At the nano-scale, two materials can be mentioned: nanocrystalline cellulose (NCC) and nanofibrillated cellulose (NFC). Both are good for strength, reinforcement, and rheological modification. They are renewable, lightweight, and are characterized by a high surface area, high mechanical strength modulus, and high tensile strength. Nanocellulose, with its various functionalities, opens up many fields for application.

Intensive research on product development with the use of cellulose nanofibers has been conducted in the fields of paper and packaging, nanocomposites and construction materials, coating and functional surfaces, adhesives, membranes, electronic materials, *etc.* (Roman and Winter 2006; Huq *et al.* 2012; Isogai 2013; Grüneberger *et al.* 2014a; Honorato *et al.* 2015; Gicquel *et al.* 2017, Kargarzadeh *et al.* 2017). It is also worth noting that nanocellulose fibers or crystals are networked and fixed irreversibly, which means that there is no risk associated with nanoparticle release or of a negative impact on human health or the environment (Hubbe *et al.* 2008; Habibi *et al*. 2010; Harlin and Vikman 2010).

Current state of the art technology presents a nanocellulose as a modifying and reinforcing agent for adhesive resins. It is known that urea formaldehyde (UF) resin has a strong adhesion to cellulose-containing materials. That is why cellulosic fibers are appropriate for UF resins modification (Veigel *et al.* 2012). The mechanism of UF resin modification is different for NFC and NCC because of their specific properties. Nanofibrillated cellulose, with a greater length of fibers and aspect ratio, reinforces the UF resin by entanglement of the elongated particles. Nanocrystalline cellulose particles that interact with a polymer matrix have less tendency to become entangled. This is because of the lower length and higher degree of crystallinity. Research on the strength enhancement of particleboard by reinforcing the adhesive with nanocellulose is of great importance. The reinforcement effect in most cases is attributed to the viscosity of the modified adhesives. Mahrdt *et al.* (2016) demonstrated that the addition of microfibrillated cellulose (MFC) to the UF adhesive resulted in a higher viscosity and shear thinning behavior compared with an industrial adhesive. This can be beneficial in terms of its application, as it would have the positive effect of distributing adhesive onto wood particles and improving the adhesion process.

Moreover, nanocellulose-reinforced gluing systems have a more environmentally friendly impact compared with industrial adhesives. The reduction in formaldehyde emissions from UF resin adhesives modified with NFC or silylated NCC was 30% or 13%, respectively, which is major (Zhang *et al.* 2011; Ayrilmis *et al.* 2016). In structural applications, the improvement of mechanical properties of wood-based panels is desirable and depends upon the bonding quality between wood particles. Mahrdt *et al.* (2015) and Veigel *et al.* (2011) investigated the behavior of a nanocellulose-modified adhesive within the wood adhesive interphase. It was shown that the share of adhesive in the glue-line increased because of the addition of nanocellulose and the fracture energy of the adhesive bonds was improved. Adhesive bonds were toughened by up to 45% with the addition of small amounts of nanocellulose (2.0%). Adhesive distribution corresponds well with mechanical board properties. Particleboards and oriented strand boards manufactured with a UF adhesive loaded with 1.0% NFC already showed a reduced thickness swelling and higher bending strength (Veigel *et al.* 2012). Efforts have been made recently to use NFC as a binder in adhesive formulations for particleboards. Amini *et al.* (2017) found that particleboard panels based solely on NFC met the industry requirements for mechanical properties. Improved bonding performance was also shown by Kwon *et al.* (2015) by incorporating the 3.0% of microfibrillated cellulose (MFC) into the UF adhesive. They indicated that the further increment in the MFC content up to 5.0% decreased the tensile shear strength of the panels.

Identifying the rheological properties of adhesives is one of the key factors in determining their technological usefulness in panel manufacturing. Hubbe *et al.* (2017) stated that understanding the rheological phenomena related to the aqueous suspension of nanocellulose is of great importance and it has been recommended that further research be conducted in the flow behavior of systems formed with the use of nanocellulose. So far there has been a lack of profound rheological data for UF adhesives modified with nanocellulose, especially with nanocrystalline cellulose. Thus, to get a deeper understanding of NFC and NCC properties, this study aims to provide an investigation of the rheological characteristics and storage stability of UF-nanobased adhesive compositions for wood-based panels.

## EXPERIMENTAL

### Materials

Three types of industrial UF resins characterized by different formaldehyde/urea molar ratios, solid contents, and viscosities were selected for this research. Two types of biopolymer, namely NFC and NCC, were chosen as a modifying agent for the UF composition. The NFC and NCC were supplied as the water suspension from two leading producers of biochemicals (The Biofore Company UPM, Helsinki, Finland; Melodea Bio Based Solutions, Jerusalem, Israel). The NCC was extracted from plant material under controlled acid hydrolysis with the use of sulphuric acid, which led to the formation of crystals suspended in the water. During treatment, the NCC obtained negative charges on their surface because of the formation of sulphate ester groups, which enhanced their stability in aqueous solutions (Peng *et al.* 2011). According to the producer data sheet, the sulfur content was determined at the level of 113.87 mmol/kg ± 26.11 mmol/kg. In turn, the NFC was essentially produced by a mechanical process and only contained cellulose fibrils and water; the only function groups were hydroxyl groups. The basic physico-chemical properties of the tested UF resin and nanocelluloses are listed in Tables 1 and 2.

**Table 1.** Characteristics of the UF Resins

| Characteristic | Units | UF1 | UF2 | UF3 |
|---|---|---|---|---|
| Appearance | – | Cloudy liquid | White cloudy liquid | White cloudy liquid |
| pH | – | 7.7 | 7.5 | 8.1 |
| Solid Content | % | 67 | 68 | 70 |
| Formaldehyde/urea Molar Ratio [a] | – | 1.11 | 1.01 | 0.85 |
| Viscosity at 200 rpm (Standard Spindle) | mPa·s | 380.4 | 433.0 | 320.4 |
| [a] Provided by the producer | | | | |

**Table 2.** Characteristics of the Nanocelluloses

| Characteristic | | Units | NFC | NCC |
|---|---|---|---|---|
| Product Form | | – | Transparent gel, thixotropic | Transparent gel, thixotropic |
| pH [a] | | – | Neutral | 6.0 |
| Solid Content | | % | 2.63 | 2.02 |
| Viscosity at 200 rpm (Standard Spindle) | | mPa·s | 1230.0 | 127.9 |
| Particle Size [a] | Width | nm | 4 - 100 | 5 - 20 |
| | Length | nm | Several µm | 100 - 500 |
| [a] Provided by the producer | | | | |

## Methods

### *Preparation of the UF-nanocellulose compositions*

The UF resin compositions were prepared as a mixture of industrial UF resin and NCC or NFC at quantities of 1.0% to 5.0% by weight of the solid resin. To achieve a proper distribution of nanocellulose in the resin, the compositions were mechanically mixed at room temperature with the use of a homogenizer (Ultra-Turrax T-25 produced by IKA - Werke, Staufen, Germany) for 10 min at a speed of 8000 rpm to 10000 rpm.

### *Viscosity measurement and rheological characteristics*

The test was performed in comparison to a standard UF resin. The influence of nanocellulose on the viscosity and flow curve characteristics of the UF resins were also determined. The tests were performed using a Brookfield Rheometer LV DV2T EXTRA (Middleboro, U.S.A) at a temperature of 23 °C ± 2 °C in a controlled increasing shear rate mode, which considered the constant volume of the test sample (9 mL) and type of spindle used (SSA-Small Sample Adapter, SC4 31, Middleboro, MA, USA). The viscosity was averaged over a 30-s time span. Three measurements were taken of each sample and averaged.

### *Storage stability*

The UF-nanocellulose compositions were stored in tightly sealed bottles for 4 weeks at room temperature (23 °C ± 2 °C). After storage, the viscosity and flow curves were evaluated, using a rheometer under the conditions described above. Before measurement, each composition was slightly stirred. Three measurements were taken of each sample and averaged. To determine the storage stability, the percentage viscosity retention based on the viscosity measurements before and after storage was calculated.

## RESULTS AND DISCUSSION

Figure 1 illustrates the rheological behavior of standard UF resins and nanocelluloses. All of the resin types exhibited Newtonian flow behavior over the whole measuring range (1 rpm to 100 rpm). The viscosity of the UF resins was in the range of 320 mPa·s to 433 mPa·s, and the highest viscosity was recorded with UF2. In contrast, the NFC and NCC presented a non-Newtonian shear thinning characteristic, which meant that the viscosity of such fluids decreased as the speed increased. The reason for such behavior was that the nanocellulose particles were linear and adopted a random position in the rest state. When shear forces appear, fibers orient in the direction of the flow, which reduces the frictional resistance, and consequently, decreases the viscosity. The nanocellulose NFC was characterized by a high viscosity (above 100 000 mPa·s) at low speeds, while the viscosity noticeably decreased to 1230 mPa·s as the speed increased. In the case of the NCC, the viscosity was lower, starting from 4000 mPa·s to 128 mPa·s, in the speed range of 1 rpm to 200 rpm.

Figures 2a through 4a show the variation in the viscosity of the UF-NFC compositions as a function of the speed. The speed-viscosity curves mainly depend upon the type of nanocellulose used, as well as on the nanocellulose loading in the composition. The modification of the UF resins with NFC noticeably increased the viscosity of the UF resin compositions. The curves of the NFC-modified UF resin compositions, especially at the NFC contents of 4.0 wt.% and 5.0 wt.%, displayed pseudoplastic non-Newtonian behavior. This shear thinning effect was less pronounced in the compositions based on the UF3 resin, which could have been because of the lower viscosity of the standard UF3 resin.

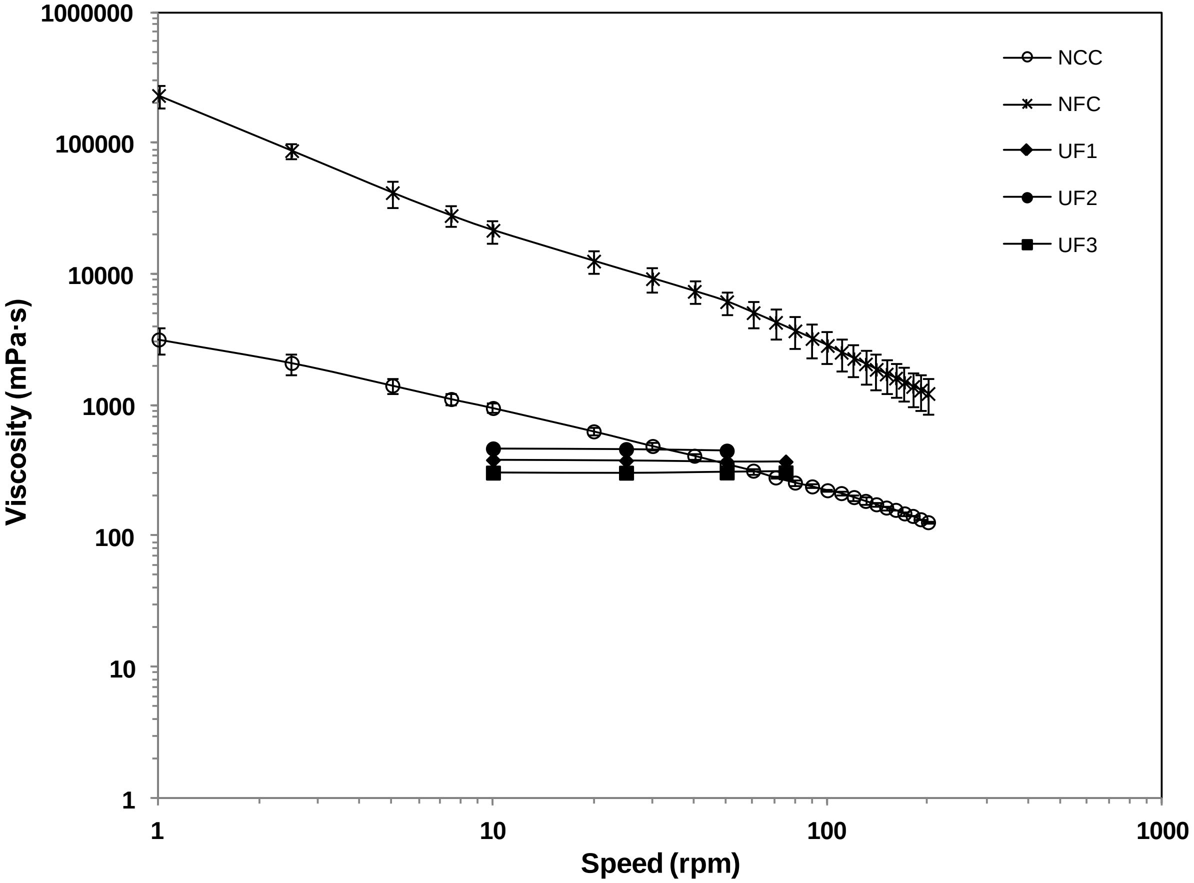

**Fig. 1.** Speed-viscosity curves of the UF resins and nanocelluloses

Different behavior is shown by the UF resin compositions loaded with NCC (Figs. 5a to 7a). The viscosity decreased as the NCC concentration increased. However, it did not lead to a change in the flow characteristics. The UF-NCC compositions remained a Newtonian fluid, as did the standard UF resin.

The speed-viscosity curves of the nanocellulose-modified UF resins after 4 weeks of storage at room temperature are presented in Figs. 2b through 7b. Figures 2b through 4b show the changes in the rheological behavior of the UF resins modified with NFC. The viscosity remarkably increased, and the flow characteristics were affected during storage. All of the compositions showed non-Newtonian behavior, which was most evident in the mixture based on the UF2 resin. Thus, an influence of the resin type on the rheological behavior was observed. When the initial viscosity (before storage) of the UF resin was higher, the viscosity of the compositions was higher after the test.

Storage did not cause changes in the speed-viscosity curves of the NCC compositions and still exhibited Newtonian behavior. It is important and crucial, in terms of technological usefulness, that the viscosity of the tested NCC compositions were increased to a lesser extent than that of the standard UF resin. This implied a high stability for the NCC-modified UF resin compositions.

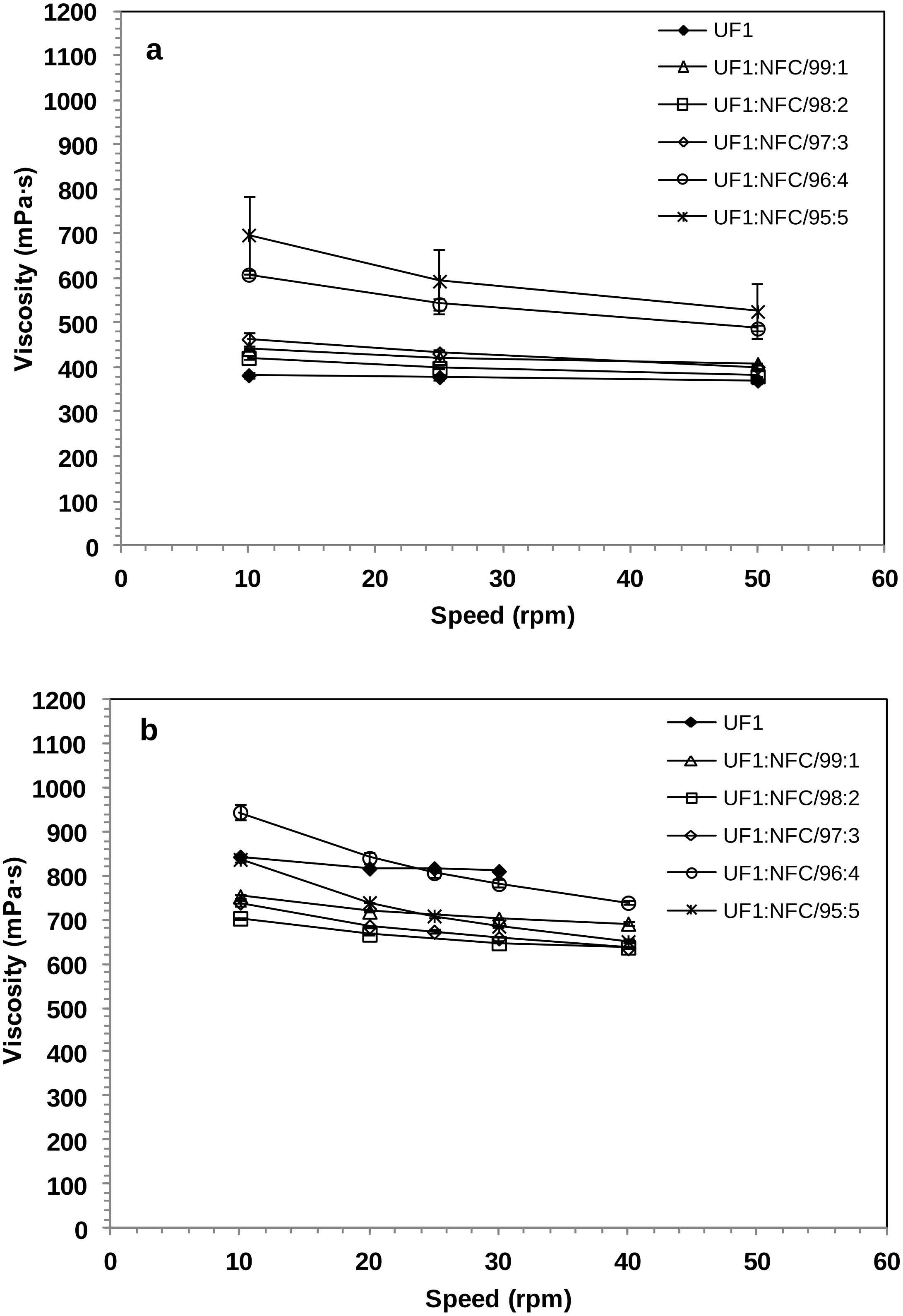

**Fig. 2.** Speed-viscosity curves of the UF1-NFC compositions depending on the NFC loading (a) before and (b) after 4 weeks of storage

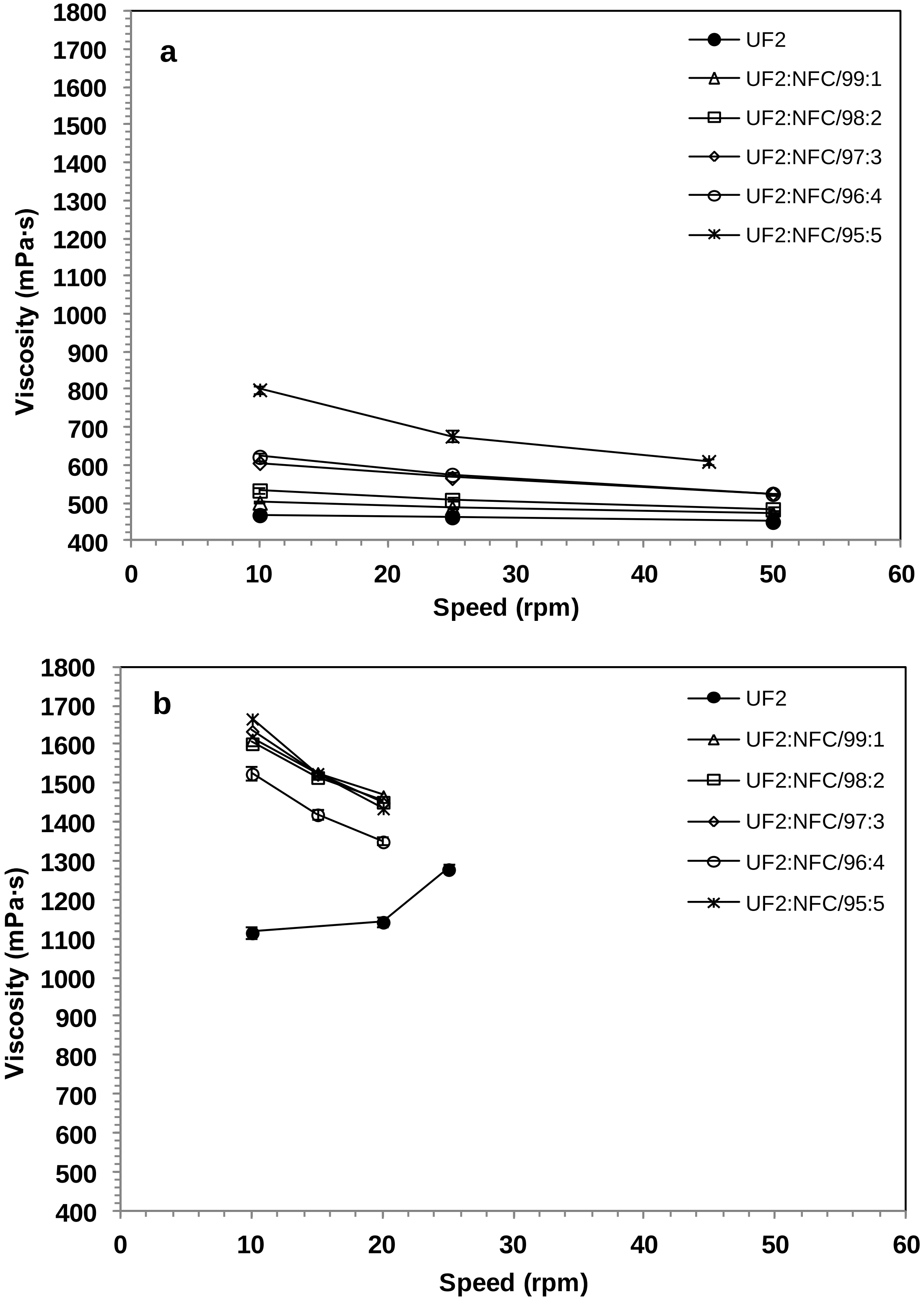

**Fig. 3.** Speed-viscosity curves of the UF2-NFC compositions depending on the NFC loading (a) before and (b) after 4 weeks of storage

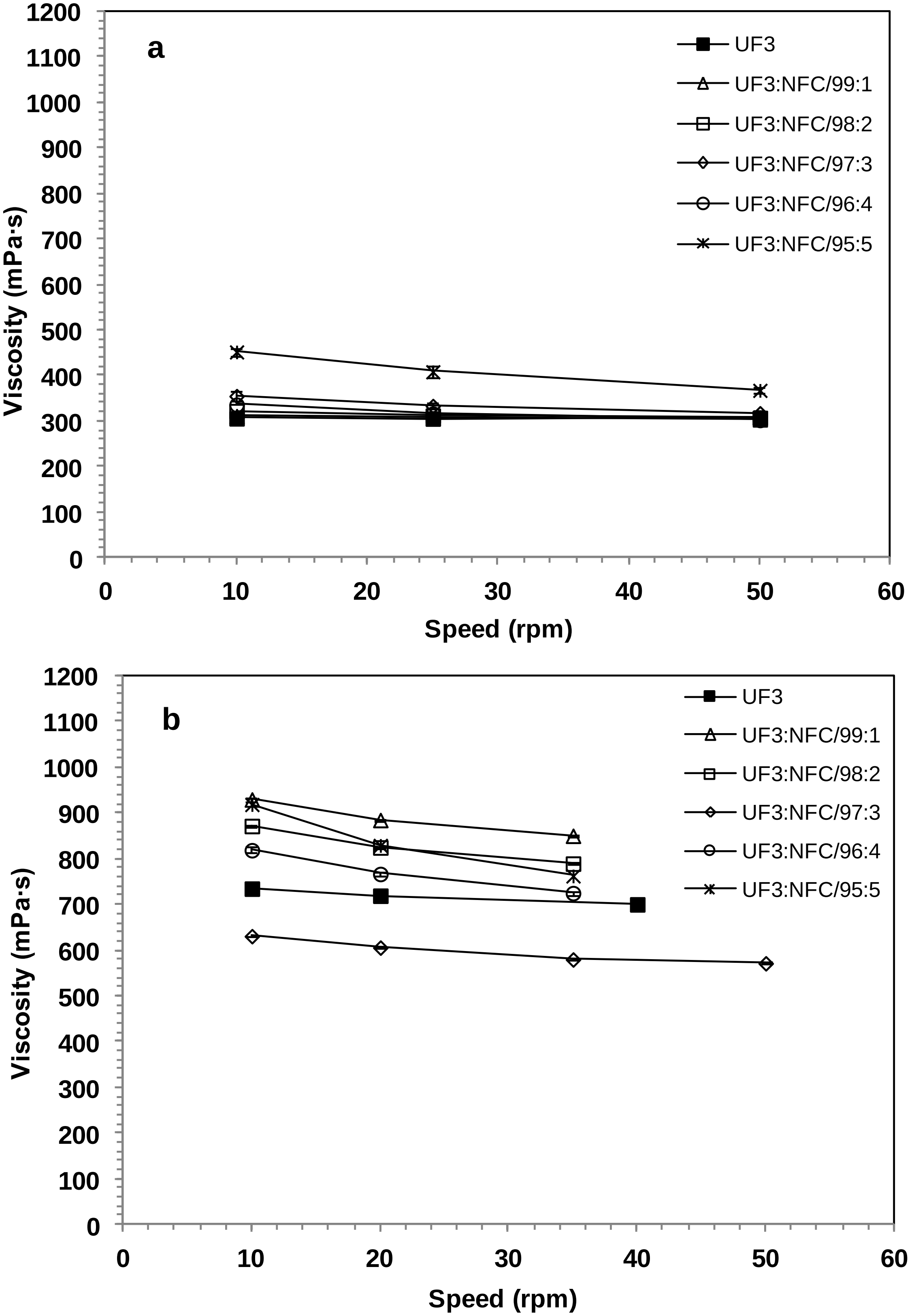

**Fig. 4.** Speed-viscosity curves of the UF3-NFC compositions depending on the NFC loading (a) before and (b) after 4 weeks of storage

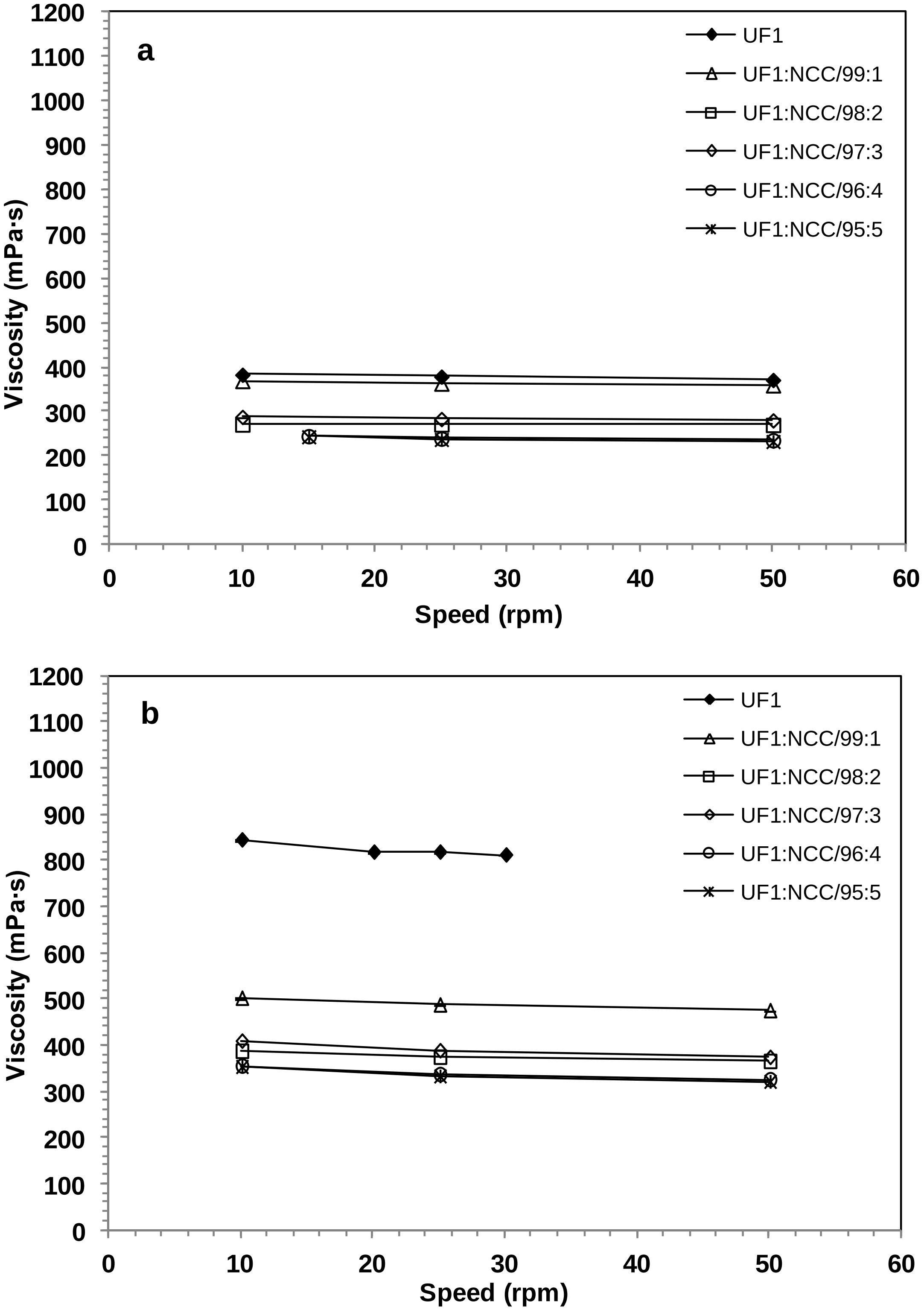

**Fig. 5.** Speed-viscosity curves of the UF1-NCC compositions depending on the NCC loading (a) before and (b) after 4 weeks of storage

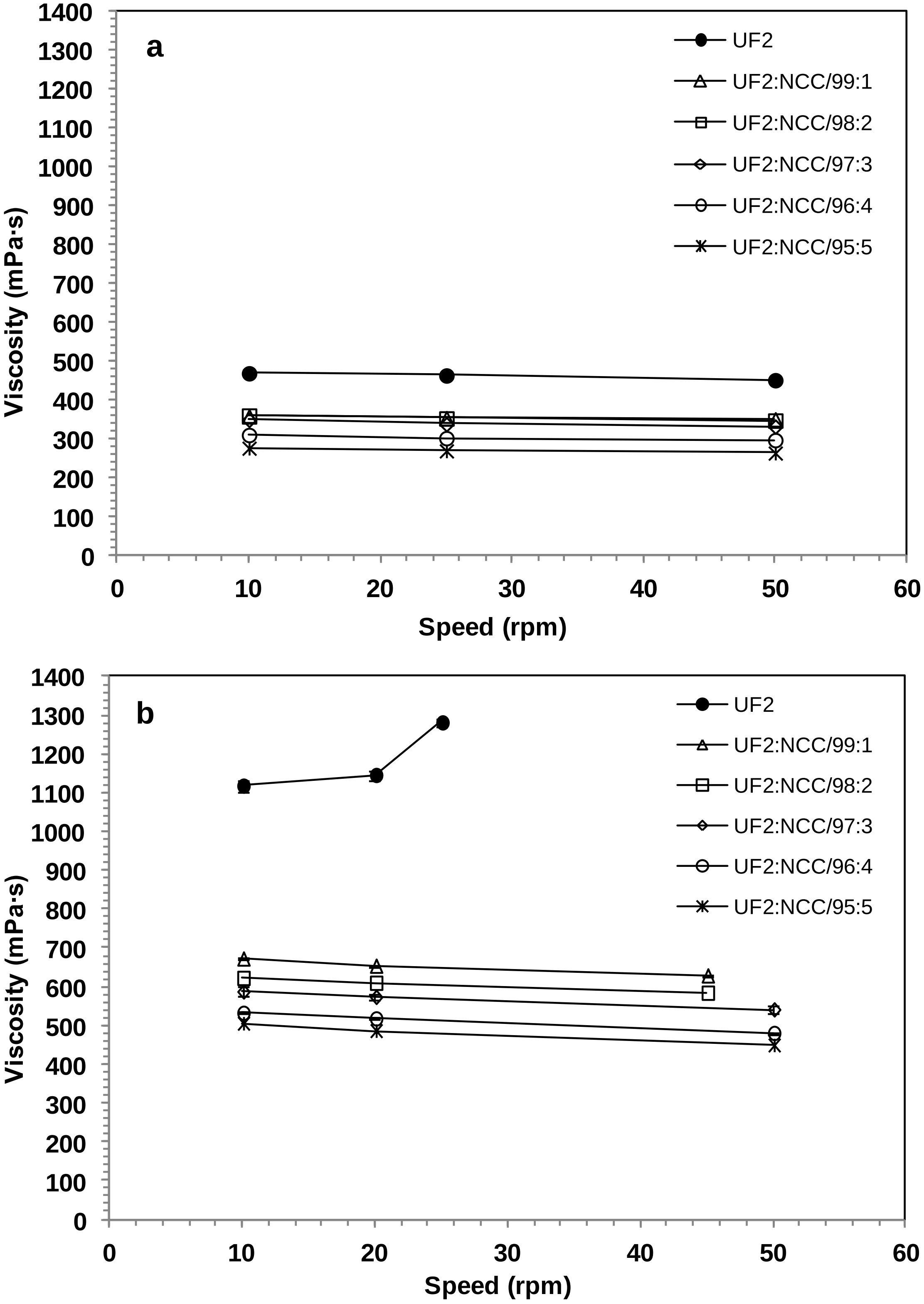

**Fig. 6.** Speed-viscosity curves of the UF2-NCC compositions depending on the NCC loading (a) before and (b) after 4 weeks of storage

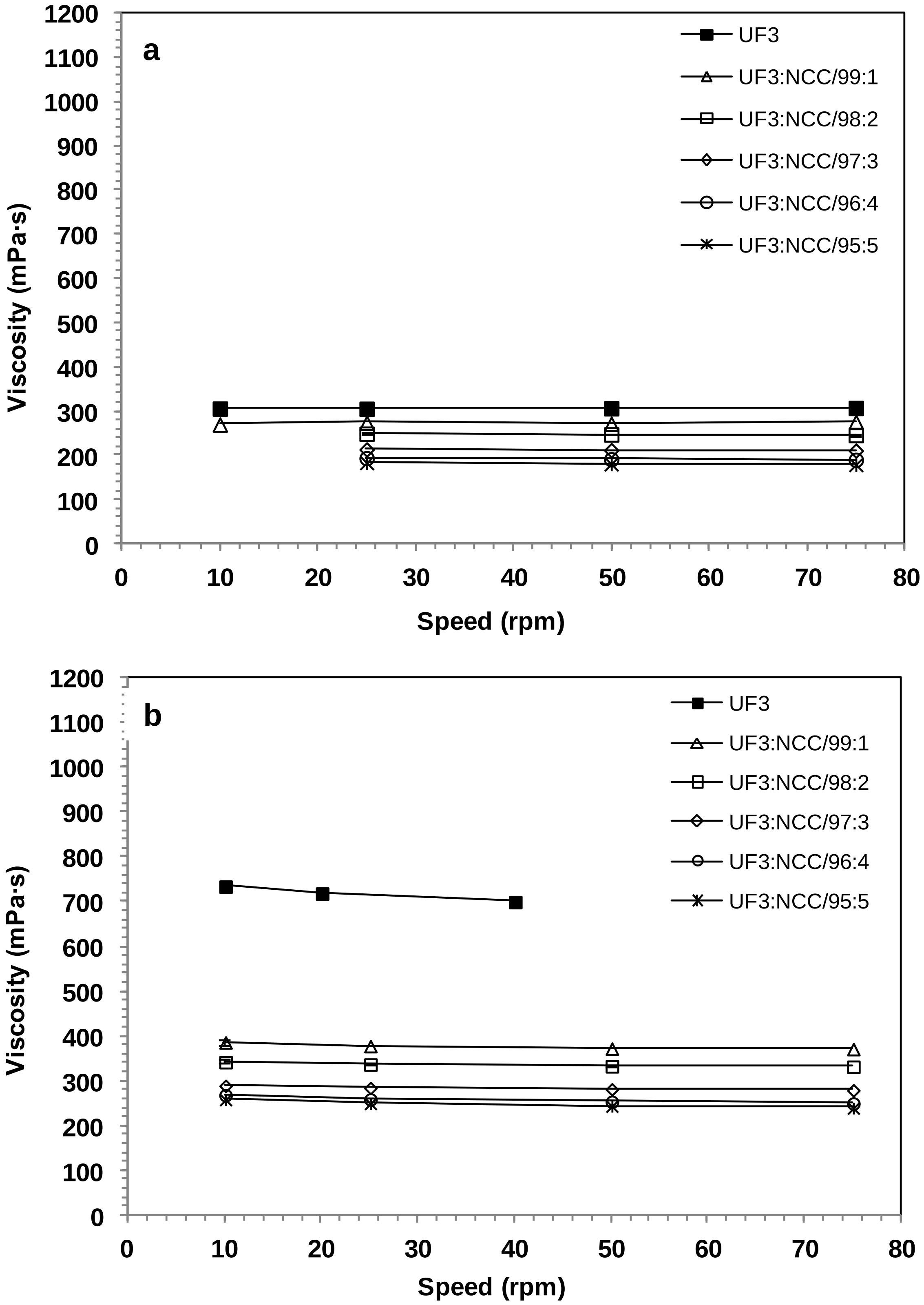

**Fig. 7.** Speed-viscosity curves of the UF3-NCC compositions depending on the NCC loading (a) before and (b) after 4 weeks of storage

The viscosity retention after storage of the standard UF resins and nanocellulose-modified UF resins is summarized in Fig. 8. The modification of the UF resins with NFC made the compositions highly viscous, with the viscosity values decreasing with an increasing NFC loading. The viscosity retention exceeded 150% in the case of the UF1 resin-based compositions and 300% in the case of the UF2 resin with a 1% NFC loading. Stable viscosity retention values were demonstrated for the UF1 resin-based compositions, with a retention of approximately 170%.

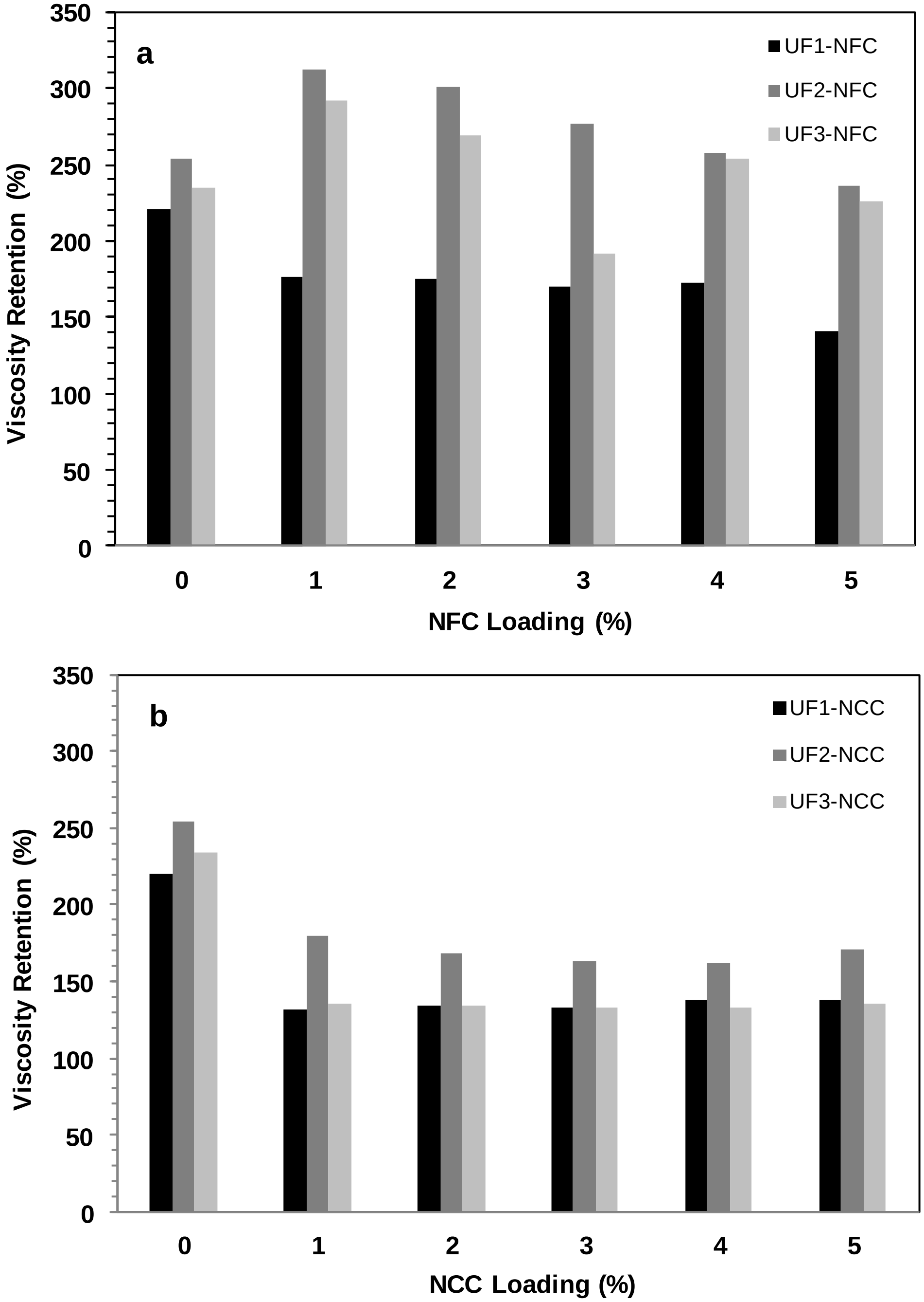

**Fig. 8.** Viscosity retention as a function of the (a) NFC and (b) NCC loading in the UF compositions after 4 weeks of storage

The viscosity retention of all of the NCC-modified UF resins was found to be between 130% and 180%. It is worth highlighting that the standard UF resins showed noticeably lower storage stability values as the viscosity retention exceeded 200%. The impact of the NCC loading was imperceptible.

## Discussion

Rheological effects are important issues that have to be considered when developing new solutions for adhesive system applications. The rheological measurements were also found to be a reliable and fast method to monitor the storage stability of the UF adhesive compositions.

The influence of the resin type on the rheological behavior of the UF-nanocellulose compositions was negligible. The main differences in the viscosities and flow characteristics between the compositions loaded with NFC and NCC resulted from the type of nanocellulose and its properties. The UF-NFC compositions exhibited shear-thinning behavior at a low NFC concentration. It was confirmed that the viscosity values of the UF-NFC compositions that were higher than those of the standard resin were caused by the higher solid content in the compositions and the tendency of NFC particles to become entangled. Grüneberger *et al.* 2014b showed similar behavior for compositions with the addition of NFC but in the case of acrylate polymer emulsions. For UF-MFC, Mahrdt *et al.* (2016) also reported high initial viscosity and shear thinning curve. The other reason for the high viscosity was the water-swellable nature of nanocellulose, especially in its amorphous regions. Veigel *et al.* (2012) demonstrated the rapidly increasing viscosity of NFC-modified UF resins and concluded that NFC should be used with small percentages to keep the viscosity of the adhesive low enough for application to wood surfaces.

It is worth noting that, unlike NFC, the NCC thinned the UF resins. The UF-NCC compositions displayed lower viscosity values and Newtonian behavior. Grüneberger *et al.* (2014b) stressed that thinned polymer emulsions exhibit remarkably lower viscosity and Newtonian flow curves, which can be explained by the fewer interactions between the emulsion particles. This was also a consequence of the crystalline regions of the NCC hydrogel, which are less sensitive to water and thus do not swell (Hubbe *et al.* 2008). Contrary to the authors' findings, Mesquita *et al.* (2018) found that viscosity increased directly with the amount of NCC. Moreover, they stated that 5% of NCC in UF composition is not suitable to produce particleboards, because of the high viscosity. This different effect was attributed to the type of NCC used. They applied freeze-dried NCC with the solid content of 98%, while authors employed aqueous suspensions of nanocellulose (only 2.0% of solids).

The UF resin is characterized by a very short pot-life, not suitable for use on slower technological lines or on plants forced to store resin for a longer time. The stabilization of UF resin properties during long time storage is important for producers and customers (Kurta *et al.* 2004). As a result of the research the modification of UF resins with nanocellulose, there is an opportunity to regulate the rheology and stability. The viscosity-increase of the UF resin after 4 weeks of storage modified with NFC was in good agreement with the study performed by Grüneberger *et al.* (2014b). A lower impact of storage on the viscosity values was observed for the UF-NCC compositions. The explanation for this behavior was found in the research by Moberg *et al.* (2017). The rheological and viscosity properties vary between suspensions depending on the dimensions of the cellulosic elements, as well as on their surface characteristics.

Cellulose nanofibril suspensions with a higher fiber length or aspect ratio are more viscous than cellulose nanocrystal suspensions. Thus, NCC can act as a stabilizer of UF adhesive resin in order to prolong its lifetime.

## CONCLUSIONS

Nanocrystalline cellulose has proven to be promising material in modification of UF adhesive compositions in terms of viscosity and rheological properties. The NCC-modified resins demonstrated a lower viscosity than the unmodified resins. In comparison, the NFC-modified resins showed substantially higher viscosity values, thus limiting the feasible NFC loading level. The shear thinning effect was highly pronounced in the UF compositions modified with NFC. Moreover, the UF resins modified with NCC kept their rheological behavior and proper viscosity after 4 weeks of storage, which lowered the percentage of viscosity retention by approximately 1.5 times compared with industrial resins. It indicates the possibility to use NCC as a stabilizer extending the pot-life of UF resin.

## ACKNOWLEDGMENTS

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