

## Rheology of cement pastes with superplasticizer and nanocellulose crystals

*Reologia de pastas de cimento com superplastificante e nanocristais de celulose*

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### Abstract

**T**he addition of nanocellulose to cement-based materials has gained significant attention in recent years due to its potential to improve concrete performance. However, the effects of nanocellulose on the rheological properties of cementitious systems are not fully understood. This work investigated the use of crystalline nanocellulose (CNC) and its effect on yield stress, plastic viscosity and structuration rate of cement pastes with and without superplasticizer admixtures (SP). Different contents of CNC were added (0.00-0.10 wt%) to pastes without and with SP. Rotational rheometry tests were conducted up to 80 min of hydration. Results showed that CNC addition increased static yield stress and structuration rate both of pastes without SP (only for contents above 0.02 wt% of CNC) and with SP (all CNC contents). These effects were more intense in pastes with SP. Regarding dynamic yield stress, only pastes with SP were affected by CNC addition. In turn, plastic viscosity decreased when CNC was incorporated, both in the presence and absence of SP. These findings suggest that CNC can be useful for applications that require relatively high structuration after the material is cast, while keeping a good pumpability (e.g. 3D printing).

**Keywords:** Crystalline nanocellulose. Rheology. Yield stress. Viscosity. Superplasticizer.

### Resumo

A adição de nanocelulose em materiais cimentícios tem obtido destaque devido à capacidade de melhorar o desempenho do concreto. Todavia, os efeitos da nanocelulose nas propriedades reológicas de sistemas cimentícios não são totalmente compreendidos. Este trabalho investigou o efeito do uso de nanocelulose cristalina (CNC) na tensão de escoamento, viscosidade plástica e taxa de estruturação de pastas de cimento com e sem superplastificantes (SP). Diferentes teores de CNC foram adicionados (0,00 a 0,10% da massa de cimento) em pastas com e sem SP. Testes de reometria rotacional foram conduzidos até 80 minutos de hidratação. A adição de CNC aumentou a tensão de escoamento estática e a taxa de estruturação tanto das pastas sem SP (apenas teores acima de 0,02%) quanto das pastas com SP (todos os teores de CNC), nas quais os efeitos foram mais intensos. Quanto à tensão de escoamento dinâmica, apenas as pastas com SP foram afetadas. A viscosidade plástica diminuiu com a incorporação de CNC, tanto na presença quanto na ausência de SP. Tais achados sugerem que o uso de CNC pode ser útil em aplicações que requerem estruturação relativamente alta após o lançamento do material, enquanto mantêm boa capacidade de bombeamento, como na impressão 3D.

**Palavras-chave:** Nanocelulose cristalina. Reologia. Tensão de escoamento. Viscosidade. Superplastificante.

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## Introduction

The ongoing evolution of technology has allowed the construction industry to improve its methods, generating solutions in terms of cost, speed, reliability and sustainability. As a result of this evolution, processes that involve complex casting of concrete have been widely implemented, requiring specific fresh-state properties from the material (Sikora *et al.*, 2021; Yuan *et al.*, 2017). Examples of such processes include guniting, self-leveling, and 3D printing of concrete. The control of the fresh behavior of concrete is enabled by advancements in the field of rheology, the science that describes the flow and deformation of materials (Yuan; Shi; Jiao, 2023; Ferraris *et al.*, 2017; Bessaies-Bey *et al.*, 2022).

Several techniques have been studied to modify rheological properties of cementitious materials. One possible approach is the utilization of nanoparticles, known for their high specific surface area, which leads to increased interactions with the cement particles when the system is well dispersed. In recent years, nanocrystalline cellulose has emerged as a promising material due to its renewable sources, sustainable characteristics, advanced mechanical properties, and lower cost compared to most nanoparticles (Barnat-Hunek *et al.*, 2019; Kargarzadeh *et al.*, 2018). Several works investigated the effects of nanocellulose in the hardened state of concrete, analyzing properties such as compressive and tensile strength, thermal conductivity, microstructure, and durability, among others (Guo *et al.*, 2020; Balea *et al.*, 2019). Regarding the effects of cellulose nanocrystals (CNC) on the fresh properties of cementitious materials, although there is scientific information available, it is limited and more controversial when compared to information about the hardened properties. Some studies suggested that the yield stress and viscosity can either increase or decrease with the addition of CNC, depending on the content added and the dispersion of CNC particles. According to Cao *et al.* (2015) and Guo *et al.* (2020), low contents of CNC (up to 0.2 wt%) can increase the fluidity of the cement paste, acting like water-reducing admixtures. This effect is due to the steric stabilization caused by adsorption of CNC on cement particles, and can only be observed when CNC particles are well dispersed in the cement matrix. Raghunath, Raghunath and Foster (2023) and Cao *et al.* (2015) have stated that higher contents of CNC, on the other hand, decrease the fluidity of the paste and consequently increase its yield stress. According to the authors, this behavior is a result of a less dispersed state of CNC in higher contents. The agglomeration of CNC can entrap free water and generate more friction and contact between the particles within the cement matrix. It is important to observe that some authors noticed that the yield stress reduction caused by lower contents of CNC varies according to the type/characteristics of the CNC used, and in some cases, this reduction is not significant (Montes *et al.*, 2020). In turn, other works have observed a relevant increase in yield stress even in contents as low as 0.035 wt% of CNC, accompanied by a decrease in plastic viscosity (Nassiri *et al.*, 2021). As for the simultaneous incorporation of CNC and superplasticizer, there is no comprehensive investigation to the best of our knowledge.

The main objective of this work is to investigate the effects of cellulose nanocrystals on the rheological properties (static yield stress, dynamic yield stress, plastic viscosity, and structuration rate) of cement pastes with superplasticizers over time, relating the results with variations in CNC content (0.00; 0.02, 0.05 and 0.10 wt%). For this purpose, cement pastes were produced with Portland cement, water, and CNC to address the effects of nanocellulose on a cementitious system without the interference of admixtures. In addition, pastes with superplasticizer were produced to verify how its presence influences the CNC behavior.

## Methods

### Materials

For the production of cement pastes, the following materials were used: ordinary Portland cement (PC) manufactured by Itambé, marketed as CPV-ARI in Brazil; a polycarboxylate-based superplasticizer admixture (SP) commercially marketed as PowerFlow 4001, manufactured by MC-Bauchemie; an aqueous solution of crystalline nanocellulose (CNC), supplied by the company Nanobiocell (Joinville, Brazil) with a concentration of 15 g of solid cellulose per liter of solution; and water.

The CNC was derived from paper residues and produced by acid hydrolysis. No surfactant was used to stabilize the CNC solution. Cellulose nanocrystals, in general, are needle-shaped particles, with less than 100 nm in diameter and a length below 1000 nm. The average length of the nanoparticles used in this work was measured with an Anton Paar Litesizer 500 dynamic light scattering (DLS) instrument (Figure 1). To confirm the shape and width of the CNC, transmission electron microscope (TEM) images were obtained (Figure 2) with a Jeol JEM-2100 electron microscope. A less concentrated solution of CNC (1 g of CNC per liter of solution) was used in DLS and TEM procedures. The size distribution curve presented in Figure 1 confirms that most of the particles have less than 1000 nm in length, although a small portion of the curve lies above

1000 nm. It is important to note that, while the DLS analysis can approximate the CNC particle length, it is best suited to characterize the size distribution of spherical particles, as the measurement is given in terms of hydrodynamic diameter. Therefore, a combination of DLS with image analysis is useful. Figure 2 exhibits the TEM image that proves the typical needle shape of CNC particles, as well as the nanometric width, clearly below 100 nm. Figure 2 also suggests that some particles formed an agglomeration similar to a fiber network (on the right), which may explain the values above 1000 nm on the DLS size distribution. The CNC solution was submitted to an extra process of sonication. For this process, the equipment called Vibracell (by Sonics & Materials INC.) was used, with an ultrasonic probe of 13mm in diameter, a total power of 700 W and 30% amplitude, in order to attenuate the agglomeration of the nanoparticles in the aqueous suspension. The CNC solution (concentration of 15 g of solid CNC per liter of water) was sonicated during 15 minutes, while immersed in an ice bath to prevent temperature from rising.

### Mix design and sample preparation of cement pastes

Eight mixes of cement pastes with a fixed water/cement ratio of 0.35 were assessed, varying the CNC content and the presence of SP. Table 1 details the mix design of the produced pastes (parameters by weight). The name of the composition is given by REF for the control mixes (without nanocellulose) or CNC (for the samples containing the nanomaterial), followed by the content of CNC and “+SP” for the samples containing the chemical admixture. Right after water was added to the cement, the samples were homogenized for 2 minutes and 30 seconds with a 650 W mixer at 600 rpm. For each sample, a batch of 150 ml of paste was produced.

Figure 1 - Size distribution of CNC determined by DLS

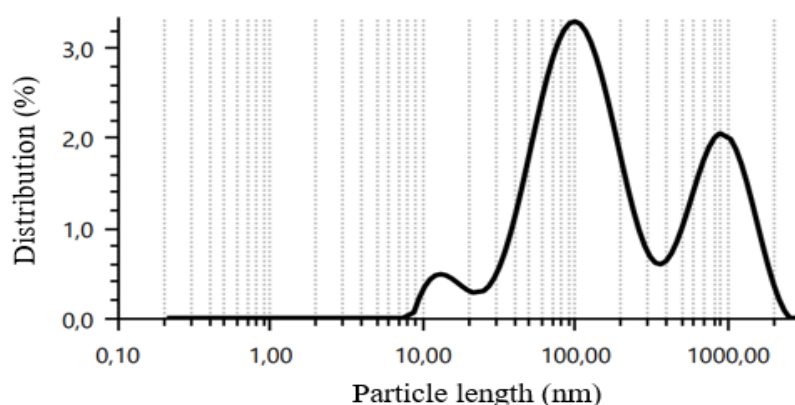


Figure 2 - TEM images of the CNC used

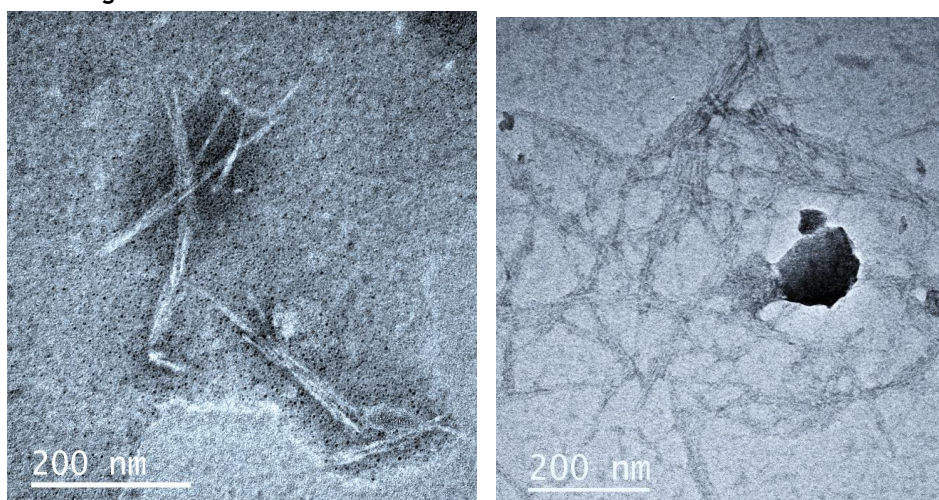


Table 1 - Mix design parameters

| Sample         | Cement (g) | Water (g) | CNC (bcw%)* | SP (bcw%)* |
|----------------|------------|-----------|-------------|------------|
| REF            | 223.00     | 78.00     | -           | -          |
| CNC 0.02%      | 223.00     | 78.00     | 0.02%       | -          |
| CNC 0.05%      | 223.00     | 78.00     | 0.05%       | -          |
| CNC 0.10%      | 223.00     | 78.00     | 0.10%       | -          |
| REF + SP       | 223.00     | 78.00     | -           | 0.15%      |
| CNC 0.02% + SP | 223.00     | 78.00     | 0.02%       | 0.15%      |
| CNC 0.05% + SP | 223.00     | 78.00     | 0.05%       | 0.15%      |
| CNC 0.10% + SP | 223.00     | 78.00     | 0.10%       | 0.15%      |

Note: \*by cement weight.

## Rheological tests for cement pastes

Rheological properties of the cement pastes were obtained by rotational rheometry, conducted using a Viscotester iQ Air (Haake) rheometer with a vane tool (22.0 mm in diameter), in a temperature-controlled environment at 25 °C. The cup was 24.0 mm in diameter and had a serrated surface to prevent wall slip.

### Static yield stress and structuration rate measurement

The static yield stress ( $\tau_{0,s}$ ) corresponds to the applied stress that causes the start of the material's flow. Testing this property generally requires the sample to be sheared at a very low rate during a certain period (a few seconds/minutes) until a peak stress value is found (Yuan *et al.*, 2017). In this work, the shear rate linearly increased from 0.00 to 0.05 s<sup>-1</sup> during 90 s. The  $\tau_{0,s}$  was determined as the maximum stress registered during this step.

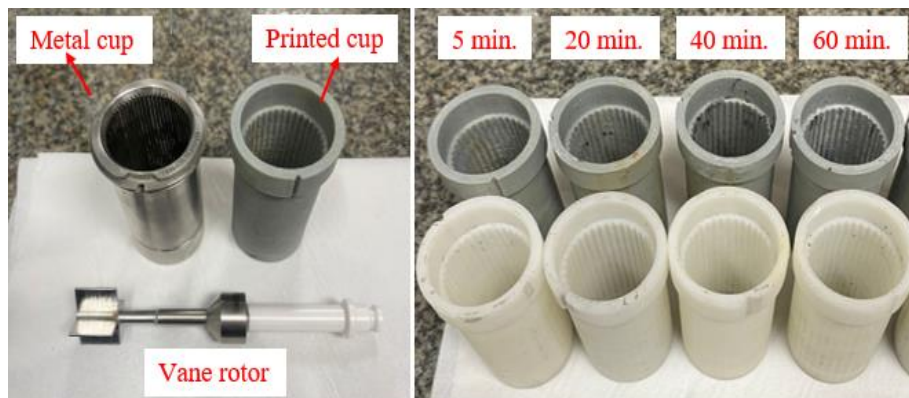
When a series of  $\tau_{0,s}$  measurements are conducted over determined intervals, the development of the yield stress with time can be used to characterize the structural build-up of cement-based materials with time at an early age (Feys *et al.*, 2017). This property is called structuration rate (Athix). In this work, Athix was determined by the linear fit of the  $\tau_{0,s}$  values recorded over the first 60 min of the cement paste age, at 5, 20, 40 and 60 minutes after water was added to cement. The Athix value was defined as the slope of the linear fit. Since yield stress measurements usually disturb the samples and are very sensitive to the shear history of the sample, the rheological properties were measured with different containers (beckers) for each interval of time (4 beakers for each sample). Additional beakers were 3D printed with the same geometry as the original containers. Samples were poured into the rheometer cups (25 ml capacity) and covered with wet paper until the testing time, in order to attenuate water loss by evaporation. This approach is illustrated in Figure 3.

### Flow curves: dynamic yield stress and plastic viscosity measurement

As the flocculation state of cement pastes affects the yield stress, there are differences between values measured in a more structured state ( $\tau_{0,s}$ ) in comparison to a state where flocculation has been broken down (dynamic yield stress –  $\tau_{0,d}$ ) (Qian e Kawashima, 2018). Both  $\tau_{0,d}$  and plastic viscosity are generally measured in a low-flocculated state through a flow curve (Feys *et al.*, 2017). The following routine was elaborated to the generation of a flow curve:

- a linear increasing shear rate varying from 0 to 100 s<sup>-1</sup> was applied over 90 s to the resting sample (ascending portion of the flow curve);
- the shear rate was kept at 100 s<sup>-1</sup> for 30 s to break down flocculation; and
- the shear rate was linearly decreased from 100 s<sup>-1</sup> to 0 s<sup>-1</sup> over 90 s (descending portion of the flow curve).

Figura 3 - Rheometer cups for measurements over the time



The values for  $\tau_{0,d}$  and plastic viscosity were obtained from the descending portion of the flow curve. The curve was adjusted by the Herschel-Bulkley model, which takes into consideration the non-linear behavior of the material, and is often applied when linear models (e.g., the Bingham model) are not suitable to represent the material flow (Wallewik *et al.*, 2015). The model is presented in Equation 1; where  $\tau$  is the shear stress (Pa);  $\tau_{0,d}$  is the dynamic yield stress (Pa);  $K$  is the consistency factor and  $n$  is the flow behavior index.

$$\tau = \tau_0 + K(\dot{\gamma})^n \quad \text{Eq. 1}$$

Also considering a non-linear behavior, an “equivalent” plastic viscosity ( $\mu_{eq}$ ) was determined by Equation 2, which is a curve linearization derived from the Herschel-Bulkley model, where  $\dot{\gamma}$  is the shear rate ( $s^{-1}$ ).

$$\mu_{eq} = \frac{3K}{n+2} \cdot (\dot{\gamma}_{max})^{n-1} \quad \text{Eq. 2}$$

## Results and discussion

### Initial static yield stress

Table 2 presents the initial values of  $\tau_{0,s}$  for all the cement pastes tested. The tests were conducted 5 minutes after the water was added to cement. For pastes with SP,  $\tau_{0,s}$  increased proportionally with the addition of cellulose nanocrystals. A significant increase was observed even with the lowest content added (0.02 wt%). For the pastes without SP, the same 0.02 wt% content did not cause a significant difference when compared to the reference sample. Due to variability of  $\tau_{0,s}$  test observed in previous works (De Matos *et al.*, 2021), only variations above 10 % were considered relevant in this work. The other contents of CNC (0.05 and 0.10 wt%) also caused a relevant raise in the  $\tau_{0,s}$  of the pastes without SP. These results suggest that the addition of crystalline nanocellulose increases the yield stress of cementitious materials in a general way, although this effect may not be observed in some cases where very low contents are added. This conclusion is coherent with the work from Nassiri *et al.* (2021), where a content of 0.035% CNC was enough to increase yield stress of cement pastes. It differs, however, from the observations made by Cao *et al.* (2015) and Montes *et al.* (2020), who state that contents of CNC up to 0.20% wt. generally decrease yield stress because CNC, when well dispersed, can adsorb the cement particles and generate a steric stabilization effect. This effect is responsible for a behavior similar to water-reducing agents. The authors attribute the increase in yield stress at contents above 0.20 wt% of CNC to the agglomeration of particles, which prevail when compared to the steric stabilization effect. Even though sonication was conducted in this work to attenuate particle agglomeration, the full dispersion of nanomaterials is a challenge. In fact, Figure 1 suggests the moderate presence of agglomerates even after sonication was conducted in the CNC used in this work. Investigation of CNC dispersion in the cement matrix is necessary in order to confirm whether the agglomeration is the main reason for the increased yield stress or not. It is important to note that Montes *et al.* (2020) have tested several types of CNC, and in some cases the reductions in yield stress were very shorter than the others. According to the authors, different methods of extraction of cellulose can generate functional groups that affect the adsorption of CNC to cement particles, reducing its capabilities to decrease yield stress. Therefore, it should be helpful to further look into relations between distinct types of cellulose nanocrystals (different degrees of crystallinity, size, shape, production methods, and sources) and eventual changes in its effects on cement pastes.

It can also be concluded that the addition of CNC was more influential in the presence of SP. The percentage variance observed in the pastes with SP is much higher when compared to the samples without SP. As stated



by Cao *et al.* (2015) and Montes *et al.* (2020), the addition of a low content of CNC does not increase (and even decreases) the yield stress of the paste. However, it only happens when CNC particles are well dispersed and adsorbed on the cement grains, causing a steric stabilization effect. On the other hand, when CNC is not adsorbed on cement particles, its physical effects and agglomeration in the matrix cause an intense yield stress gain. Based on these observations, one reason for the more pronounced increase in yield stress by CNC addition in the presence of SP could be a competitive adsorption between CNC and SP. As SP has a strong tendency to adsorb on cement, it could prevent CNC adsorption, attenuating the steric stabilization effect of CNC, thus increasing yield stress. It could even be noticed that, as a possible result of these interactions, the paste with SP and 0.10% CNC presented a higher value of initial yield stress when compared to the sample with 0.10% CNC without SP.

## Structuration rate

Figure 4 shows the evolution of  $\tau_{0,s}$  over the first hour of hydration for the pastes without SP (Figure 4a) and with SP (Figure 4b). The respective  $A_{thix}$  values are presented in Table 3. These results indicate that pastes with CNC have a greater structuration rate when compared to the reference samples. This effect was observed for pastes with and without SP, except when 0.02 wt% of CNC was added to the paste with SP, where the difference in  $A_{thix}$  was not significant. Both for initial  $\tau_{0,s}$  and  $A_{thix}$  values, the 0.02 wt% content of CNC only influenced the superplasticized pastes, which seem to be more sensitive to very low additions of CNC. Higher values of  $A_{thix}$  suggest that the gain of yield stress occurs in a faster way. While it may be positive for applications that require structuration right after the concrete is cast, it also indicates that the material loses its workability in a shorter period. According to Bai *et al.* (2023) and Cao *et al.* (2015), the addition of CNC delays cement hydration in the first hours, which indicates that the gain of  $A_{thix}$  is not related to the faster formation of hydration products. Additionally, Deze *et al.* (2022) have shown that CNC solutions are highly thixotropic. If this thixotropic behavior of CNC also occurs when it is added to the cement paste, a greater  $A_{thix}$  is expected, proportional to the material's resting time.

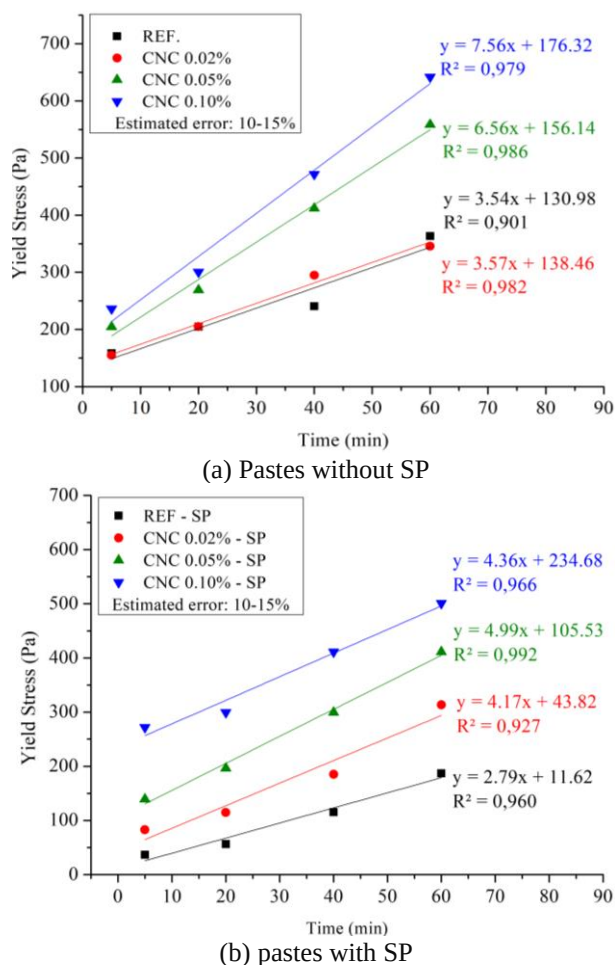
## Dynamic yield stress

The  $\tau_{0,d}$  of the cement pastes measured at the age of 5 and 80 minutes are shown in Figure 5 for the pastes without SP (Figure 5a) and with SP (Figure 5b). While  $\tau_{0,d}$  may seem to slightly decrease with the addition of 0.02% and 0.05% of CNC, the variations observed are not significant enough (considering the testing variability) to conclude that there is a decreasing tendency. Furthermore, the addition of 0.10% of CNC did not significantly change  $\tau_{0,d}$  when compared to the reference. It was expected that a significant increment on  $\tau_{0,d}$  would occur when CNC contents of 0.05 and 0.10 wt% were added, considering these same contents increased the  $\tau_{0,s}$ . One possible reason it did not happen could be that, as the  $\tau_{0,d}$  is measured at the end of the flow curve, there is a significant period of paste shearing before the measurement. Increments in yield stress generated by CNC are attributed to particle agglomeration (Cao *et al.*, 2015) and to water retention by cellulose particles (Souza *et al.*, 2023). This additional shearing could break CNC particle interactions (aggregates, networks) and also cause the releasing of entrapped water into the cement matrix. This would reintroduce the steric stabilization effect of CNC and also make the paste less structured during the  $\tau_{0,d}$  measurement, when compared to the  $\tau_{0,s}$  measurement.

Table 2 - Static yield stress of cement pastes at 5 minutes

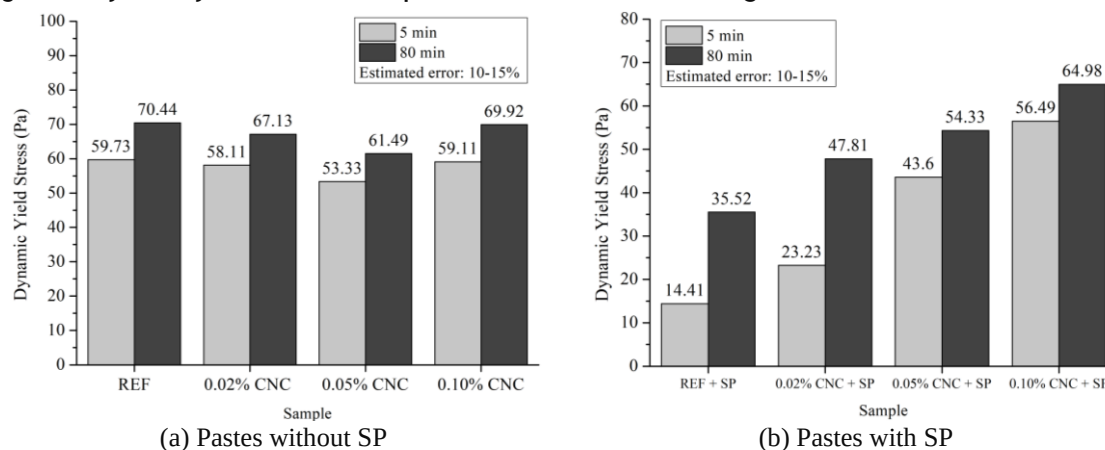
| CNC contente<br>(%wt.) | Initial $\tau_{0,s}$ (Pa)* |                  |
|------------------------|----------------------------|------------------|
|                        | Without SP                 | With SP          |
| 0.00%                  | 158.3                      | 36.48            |
| 0.02%                  | 155.1 (-2.02%)             | 82.97 (+127.44%) |
| 0.05%                  | 204.4 (+ 29.12%)           | 139.3 (+281.85%) |
| 0.10%                  | 236.3 (+ 49.27%)           | 271.9 (+625.34%) |

Note: \*estimated error: 10%.

Figura 4 - Static yield stress of the pastes over time and linear fit for  $A_{thix}$  determinationTable 3 -  $A_{thix}$  of cement pastes

| CNC contente<br>(%wt.) | $A_{thix}$ (Pa/s) |         |
|------------------------|-------------------|---------|
|                        | Without SP        | With SP |
| 0.00%                  | 3.54              | 2.79    |
| 0.02%                  | 3.57              | 4.17    |
| 0.05%                  | 6.56              | 4.99    |
| 0.10%                  | 7.56              | 4.36    |

Figura 5 - Dynamic yield stress of the paste at 5 and 80 minutes of age

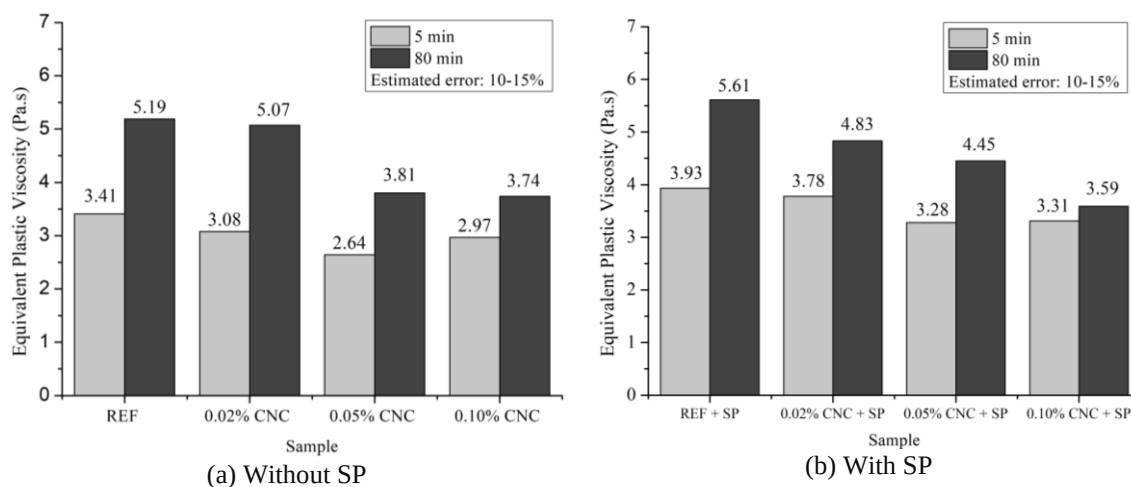


For pastes with SP, on the other hand, an increasing tendency was observed (Figure 4b). The  $\tau_{0,d}$  was raised proportionally to the addition of CNC, both for the initial and final values (5 and 80 minutes). Considering that CNC only seems to increase the  $\tau_{0,d}$  in the presence of SP, the idea of a strong influence of SP in the CNC behavior is highlighted. This conclusion can also be corroborated by the fact that CNC caused a greater influence on  $\tau_{0,s}$  of pastes with SP (Table 2). According to Björnström and Chandra (2003), the main action mechanism of superplasticizers in the cement matrix involves its adsorption on cement particles. Therefore, regarding the pastes with SP, it is possible that, even with the additional shearing of the material before the  $\tau_{0,d}$  measurement, the CNC particles could not adhere to cement grains as they were already adsorbed by SP.

## Equivalent plastic viscosity

Figure 6 shows the equivalent plastic viscosity of the pastes at 5 and 80 minutes of age. The effect of nanocellulose on plastic viscosity was similar in both pastes with and without SP (Figures 6a and 6b, respectively). There was a slight decrease in plastic viscosity when CNC was added, especially in the contents of 0.05% (lowest values at 5 minutes) and 0.10% wt. (lowest values at 80 minutes). The capability of CNC to increase yield stress while reducing the plastic viscosity was also stated by Nassiri *et al.* (2021). It can be useful for applications that require a good structuration of the fresh material (high yield stress) while maintaining its pumping capabilities (low viscosity/dynamic yield stress); 3D printing of concrete is an example of an application that benefits from such characteristics (Zhang *et al.*, 2021; Rehman; Kim, 2021). Deze *et al.* (2022) have stated that CNC solutions exhibit a shear-thinning behavior, as their viscosity is significantly reduced as the shear rate is increased. This happens due to weak interactions between CNC particles that are easily broken by shear stress. Considering the hypothesis that CNC also behaves in a shear-thinning manner inside the cement matrix, it could explain the decrease in plastic viscosity. It is important to note that, in this work, plastic viscosity was measured considering shear rates from 0 to 100  $s^{-1}$ . Also, the hydrophilic characteristics of CNC (Souza *et al.*, 2023) could help decrease plastic viscosity. If CNC aggregates entrap free water, mixing the paste (increasing shear rate) could help the liberation of this water into the cement matrix, making it less viscous.

Figura 6 - Equivalent plastic viscosity of the pastes at 5 and 80 minutes of age



## Conclusions

Rotational rheometry was applied to cement pastes with different contents of added CNC and the presence or absence of SP. The results indicated that the addition of CNC can significantly influence the rheological properties of cementitious systems. This influence depends on the amount of CNC added and its interaction with the SP admixture. The main conclusions are summarized below:

- the addition of nanocellulose significantly increased the static and dynamic yield stress of superplasticized cement pastes. As for pastes without SP, static yield stress increased after a certain amount of CNC was added (in this case, 0.05 wt%) and no clear tendency was observed for dynamic yield stress. Aggregation of CNC particles and entrapping of free water by CNC are possible causes for yield stress increase;



- (b) the influence of CNC on yield stress (both static and dynamic) was greater in pastes with SP, which means that there may be an interaction of CNC and SP that impacts the cement matrix. The main hypothesis consists in a competitive adsorption between SP and CNC on the cement grains. The structuration rate (Athix) of pastes with CNC was higher when compared to pastes without CNC. Furthermore, the plastic viscosity of cement pastes (both with and without SP) was slightly decreased when CNC was added. These observations could be related to thixotropic and shear-thinning behavior of CNC suspensions; and
- (c) some results presented in this work (mainly related to yield stress) differ from existing literature which suggest that contents of CNC up to 0.20 wt% generally act as water-reducing agents. The differences between the type and the dispersion of cellulose nanocrystals used in different works could be a reason for distinct findings.

Overall, it was observed that CNC directly affected the fresh properties of cement paste without and with superplasticizer, and those can be used to tailor the rheological properties of concrete. In general, CNC influence in the presence of SP was often different in intensity, and sometimes a different behavior was observed, when compared to CNC in pastes without SP. Further investigations on chemical and physical interactions between CNC and SP would be useful to confirm the suggested hypotheses. While chemical interactions could be verified with a spectroscopic (e.g. infrared) analysis of a solution with CNC and SP, physical interactions would require adsorption measurements (e.g. through total organic carbon – TOC) of SP and CNC particles on cement grains, as well as dispersion measurements of these same particles in the cementitious matrix. Also, the efficiency of CNC dispersion before its introduction in the paste can be further analyzed by comparing different sonication times and energies.

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