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Adhesive and rheological properties of fresh fibre-reinforced mortars

A. Kaci [a], R. Bouras [a], V.T. Phan [a], P.A. Andréani [b], M. Chaouche [a,*], H. Brossas [b]

[a] Laboratoire de Mécanique et Technologie, Cachan (ENS Cachan/CNRS UMR8535/UPMC/PRES UniverSud Paris), 61, av. du Président Wilson, F-94230 Cachan, France
[b] Centre d'Innovation de ParexLanko, BP 5 – 38, rue du Montmurier, ZI Parc des Chesnes 38291 Saint Quentin Fallavier, Lyon, France

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ABSTRACT

Adhesive properties of fibre-reinforced joint mortars in fresh state are investigated using the probe tack test. This test consists of measuring the force required to separate at a given velocity two plates between which a thin layer of the tested material is inserted. The adhesive properties of the mortars, including cohesion and adherence to the plate surface, are inferred from the curves representing the evolution of the tack force versus instantaneous plate separation for different pulling velocities. The adhesive properties are qualitatively related to the rheological behaviour of the mortars. The latter are shown to behave as Herschel–Bulkley shear-thinning fluids. The influence of fibre content on both adhesive and rheological properties is investigated.

1. Introduction

The probe tack test is widely used to characterize debonding properties of different types of soft materials. This includes for instance pressure-sensitive adhesives (PSA) [1,2]. It has been shown that the tackiness of these copolymer-based materials arises from a complex combination of cavitation and visco-elastic dissipation. In order to be effective, an adhesive must possess both liquid properties, to wet the surface when the bond is formed, and solid properties, to sustain a certain level of stress during the process of debonding [3]. Tack tests have also been used to consider rupture dynamics of liquids, involving in particular the phenomenon of Saffman–Taylor instability arising when separating two plates between which a Newtonian or a Non-Newtonian fluid is confined [3–6].

Adhesive properties of mineral or granular based materials (pastes) have been much less investigated [7]. In the present study we consider the case of adhesive mortars. These materials are used in practice as thin joints to bind construction blocks (bricks, stones, etc.) together or to fix tiles on horizontal or vertical surfaces. Adhesive mortars are mainly composed of sand and different mineral fillers, a binder (cement and/or lime) and organic additives. The latter are included to improve in particular the adhesive and rheological properties of the joints in the fresh state. The polymer additives are generally redispersible resin powders or water-soluble polymers. In a recent study [8] we have considered the influence of a high molecular weight water-soluble polymer additive (modified cellulosic ether) on the adhesive properties of mortars. In the present investigation we consider the influence of organic millimetric fibres on the adhesive properties of mortars using the probe tack test. The inferred adhesive parameters are qualitatively related to the rheological properties of the mortars.

In general fibres are added in cementitious materials in order to improve their mechanical properties in hardened state, and this issue has been the subject of numerous studies [9–11]. On the other hand the effect of fibre addition on the fresh properties, including the rheological behaviour has been much less investigated [12–14]. Moreover, to the best of our knowledge, there are no reported studies concerning the effect of fibres on the adhesive properties of cementitious materials in the fresh state.

2. Materials and experimental set-up

2.1. Materials

In general mortar joints include cement, fine sand, different fillers and organic additives. The composition of the simple formulation used here is reported in Table 1. Overall the sand particles are highly polydisperse with irregular forms. The maximum grain size is about 0.5 mm. In practise different polymer additives are included in the formulation to enhance the adhesive properties of the joints in the fresh state and to improve their placement properties. The polymer additives are generally redispersible resin powders and/or water-soluble polymers. In our case we used a

* Corresponding author.
E-mail address: chaouche@lmt.ens-cachan.fr (M. Chaouche).

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Table 1
Mix proportioning of constituents of the joint mortar.

Constituent	Portland cement	Siliceous sand	Fibres	Cellulose ether	Water
% (by weight)	30	70	Varied (0 → 0.82)	0.22	30

cellulose ether based water-soluble polymer (METHOCEL® from Dow Chemical).

In order to avoid creeping in the fresh state and improve the mortar properties in the hardened state, in particular to limit cracking, fibres are often included in the mix-design. In the present study, we consider the influence of cellulosic fibres on the adhesive and rheological properties of the mortars during the induction period. During this period of time (about 2 h in our case) the hydration kinetics is slow and may have then negligible effects on the adhesive and rheological properties. The average length of the fibres is about 1 mm and their average diameter on the order of 10 μm. The fibre concentration by weight is varied between 0% and 0.82%. In practice the used fibre content is about 0.5%.

The water dosage rate is fixed to 30% by weight for all the mortar pastes considered.

2.2. Experiments

2.2.1. Determination of the adhesive properties

The experimental set-up is sketched in Fig. 1. A rheometer (AR2000ex from TA Instruments) is used in the probe tack test mode. The mortar pastes are inserted between two parallel plates with rough surfaces (to minimize wall-slippage) and squeezed out at a given velocity (V_i = 500 μm/s) to reach an initial gap thickness of h_0 = 3 mm (see Fig. 1a) before starting the tack test. The initial diameter of the mortar sample (the same than the upper plate) is 40 mm. Since the initial gap thickness is much smaller than the diameter of the sample, one can assume that (at least in the beginning of the stretching test) the flow is *a priori* dominated by the shear component. The lubrication-type approach may then apply. In this approximation flow gradients in the radial direction and are ignored and the tack force can be calculated analytically even for Non-Newtonian fluids (see Section 3.3).

In order to erase eventual memory effects, the material is let to relax for 3 min before starting the tack test. By recording the evolution of the normal force, it is checked that a steady state is actually reached within this period of time. Then, the sample is stretched at a constant velocity by moving the upper plate and the temporal evolution of the normal force is recorded. The stretching velocity V is varied by two orders of magnitude (between 10 and 1000 μm/s).

To check the reproducibility of the experimental results, at least three different runs are performed for each freshly prepared sample.

2.2.2. Determination of the rheological properties

The rheological properties of the mortars are determined using the same rheometer equipped with a 4-blade vane geometry. Vane geometry is recognized to be suitable for granular pastes like mortars since with this system wall-slippage is minimized (the material is sheared in volume) [15,16]. The gap thickness (distance between the periphery of the vane tool and the outer cylinder) is 8.3 mm, which is more than an order of magnitude higher than the maximum size of the grains (0.5 mm). Then, the measurements may not be sensitive to the discrete aspect of the mortar composition. On the other hand, since the gap thickness is not sufficiently smaller than the vane tool diameter, the variation of the shear-rate and shear-stress throughout the gap space cannot be neglected. Therefore the fundamental rheological quantities cannot be determined straightforwardly from the measured torque and the rotational velocity of the vane tool. A calibration method, which is described in details in Ref. [17], is then used.

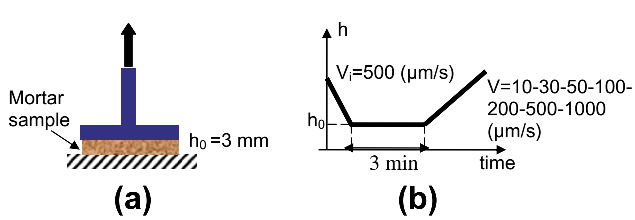

Fig. 1. Tack test set-up (a) and test procedure (b).

2.3. Results analysis

Fig. 2 represents the typical shape of the force curves (stretching force versus instantaneous gap thickness) obtained in the tack tests. The force first increases (zone 1), passes through a maximum F_{max} and then decreases (zone 2) reaching finally a plateau (zone 3). In zone 1 the mortar displays mainly elastic and then visco-elastic behaviours. The force peak is related to the adhesive strength of the material. In zone 2 one has irreversible rupture and inward flow of the material towards the plates centre. Analysis of the force decay in this zone allows characterizing rupture dynamics of the mortar. Zone 3 starts as soon as the rupture process is completed. The average value of the force plateau is related to the amount of material remained stuck onto the mobile plate. This gives the adherence strength of the mortar relative to the surface of this plate.

The value of the force peak F_{max} is related to both viscous dissipation (dynamic property) and cohesion strength (static property) whose origin includes in particular intermolecular and capillary forces. To infer the cohesion component from the adhesion strength, the force peak is represented as a function of the stretching velocity. The cohesion force is then taken to be the value of the force peak when the velocity tends to zero (Fig. 2b).

3. Results and discussion

3.1. Adhesive properties

Fig. 3a–e represents in a semi-logarithmic scale the evolution of the measured normal force versus time (or instantaneous gap thickness) for different applied pulling velocities. Each figure corresponds to a given dosage rate of fibres. The force curves have roughly the general shape represented schematically in Fig. 2a and consisting of three different zones. However, at high velocities the 'visco-elastic' zone (increasing part of the force curve) is difficult to be distinguished. In particular at 1000 μm/s it was not possible to

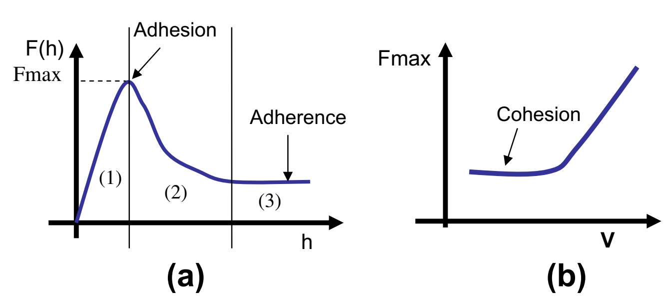

Fig. 2. Analysis of the tack force curves. (a) General shape of the tack force curves; (b) evolution of the force peak versus stretching velocity.

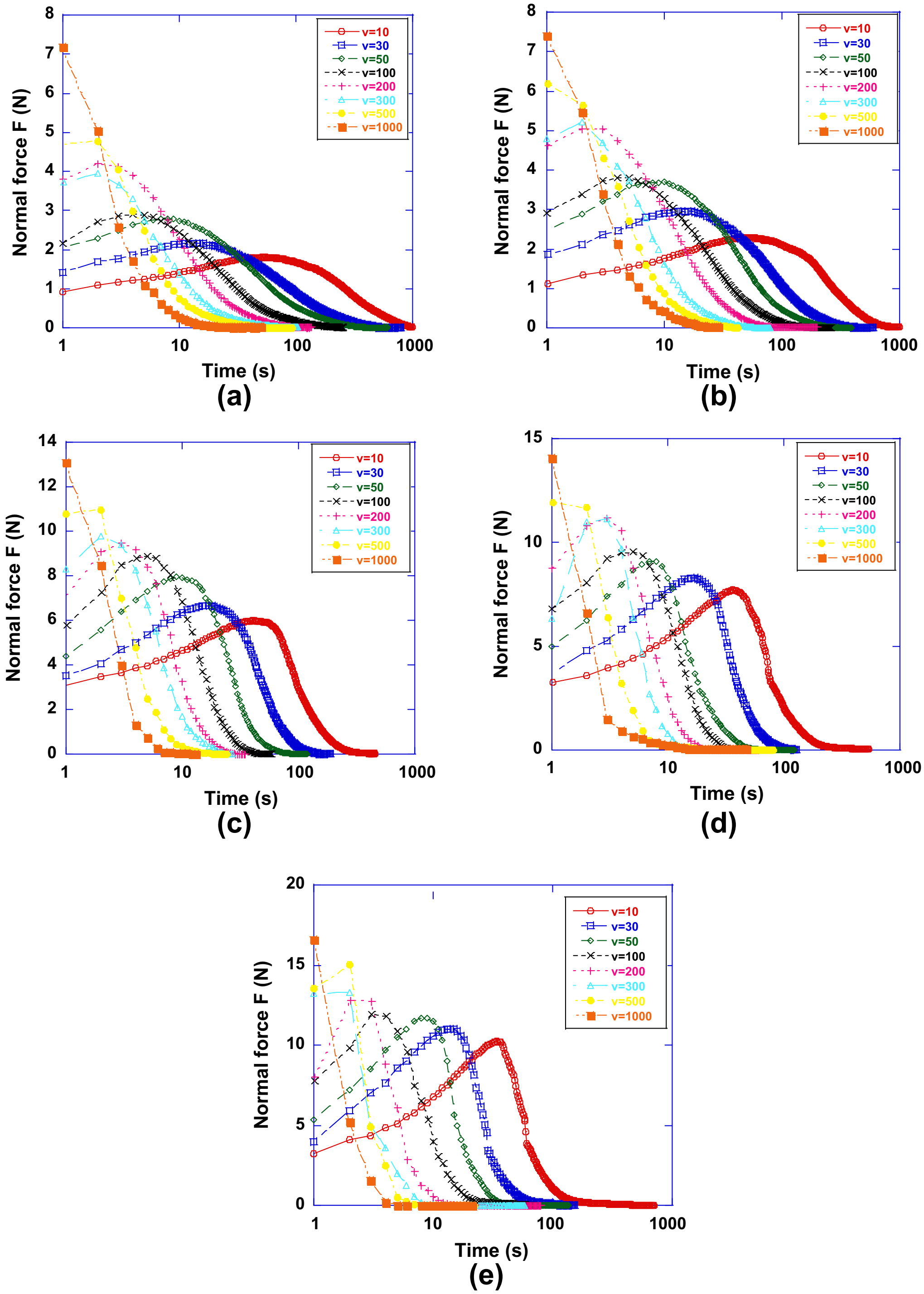

Fig. 3. Evolution of the stretching force versus time for different velocities (in μm/s). (a) Formulation with 0.13% by weight of fibres; (b) 0.27%; (c) 0.55%; (d) 0.68%; (e) 0.82%.

observe this part of the force curve. This is due to the limited rate of data acquisition of our experimental set-up (1 measure per second).

Before going into more detailed analysis of the tack results, it can be readily seen that the characteristic duration of the debonding process (duration of zone 2) significantly decreases when increasing fibre dosage rate. For instance, at 10 μm/s the duration of the debonding process is divided by an order of magnitude when increasing the fibre content from 0.13% to 0.82%. This indicates that we have probably a *qualitative* change in the rupture dynamics, passing from a liquid-like inward viscous flow to a solid-like rupture. The adhesive strength (peak of the force curve) also significantly increases with fibre concentration as it can be seen more quantitatively in the analysis reported hereafter.

3.1.1. Adhesive strength

From the measurements represented in Fig. 3, the evolution of the maximum stretching force (also referred to as the adhesive force) as a function of the velocity can be determined for each mortar formulation corresponding to a given fibre content. The results are represented in Fig. 4. For each given fibre content, the adhesive force increases with the pulling velocity. This is expected and can

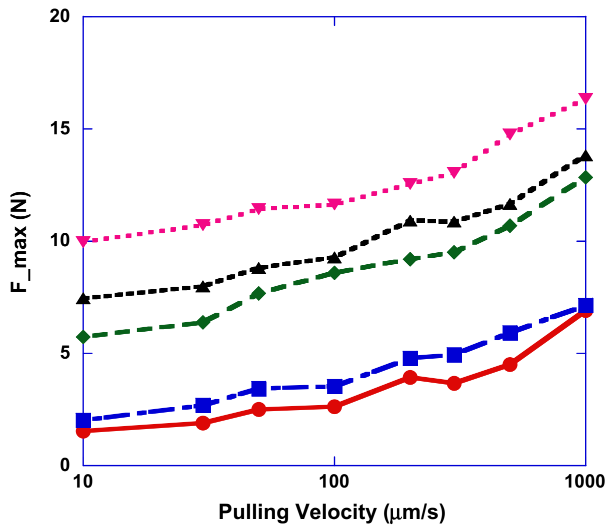

Fig. 4. Evolution of the adhesive force as a function of the pulling velocity for different fibre contents. (●) 0.13%; (■) 0.27%; (◆) 0.55%; (▲) 0.68; (▼) 0.82%.

be attributed to the increase of the viscous contribution to the adhesive force when the pulling velocity increases.

The curves are approximately parallel, which indicates that there is no significant change of the dependence of the adhesive force upon the velocity when varying the fibre content. As it can be seen further this result is in apparent contradiction with the behaviour in shear flow (rheological behaviour).

3.1.2. Cohesion

As it has been discussed above, the adhesive force comprises both viscous effects, which are velocity dependent, and cohesion, which is related to the strength of the interactions between the material constituents at rest (Fig. 2b). The later can be then determined from the adhesive force at zero-velocity. The evolution of the cohesion force versus fibre dosage rate is represented in Fig. 5. We can observe a significant increase of the cohesion when increasing fibre content. Moreover, there seems to be a critical fibre content (located between 0.27% and 0.55%) above which we obtain a large increase of the cohesion strength.

3.1.3. Adherence

The adherence force is assumed be equal to the weight of the mortar that remains stuck on the moving plate at the end of the tack test. This is determined from the force curve plateau. The evolution of the adhesion force versus fibre content for three different pulling velocities is represented in Fig. 6. Although the adherence force values are quite small (the accuracy of the force measurement is 1 mN), one can observe a dramatic decrease of adherence when adding fibres in the formulation. At high fibre content (0.82%) there is almost no-adherence at the plate surface. This may have important practical implications. The decrease of adherence with fibre content can be related to the evolution of the rheological properties when adding fibres as it is discussed below.

3.2. Influence of fibre concentration on the rheological properties of the mortars

The influence of short-fibre addition on the rheological behaviour of different types of liquids has been largely considered in the literature. This has been considered mainly in the framework of the issue of polymer composite processing [18,19]. In this case the composite melt is generally approximated as a fibre suspension in a Newtonian fluid, for which a number of studies have been devoted in the literature (see for instance the review [20] and the references therein). On the other hand the influence of fibre addition on the rheological behaviour of cementitious materials such a mortar has been much less considered. A mortar cannot be approximated as a Newtonian fluid (see below), and then approaches developed for fibre suspensions cannot be rigorously applied in our case.

3.2.1. Flow curves

The flow curves of the mortars, determined at controlled stresses, for different fibre contents are reported in Fig. 7. Fig. 7a represents the flow curves in a linear scale to display the overall form of the curves while Fig. 7b represents the corresponding Log–Log plot in order to highlight the rheological behaviour at low shear-rates.

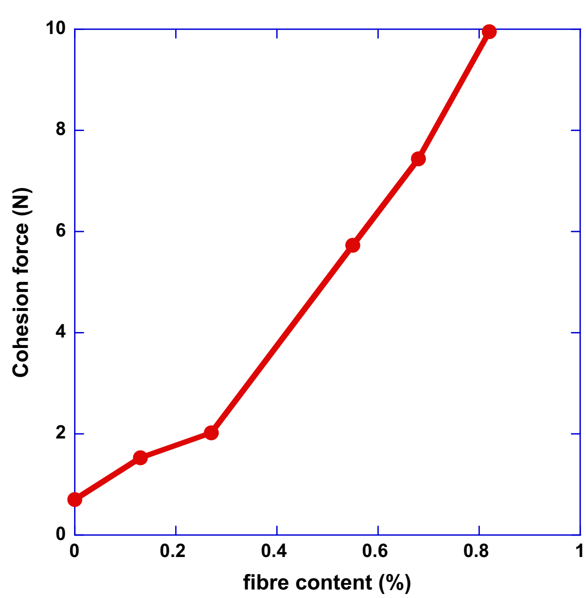

Fig. 5. Evolution of the cohesion force versus fibre content.

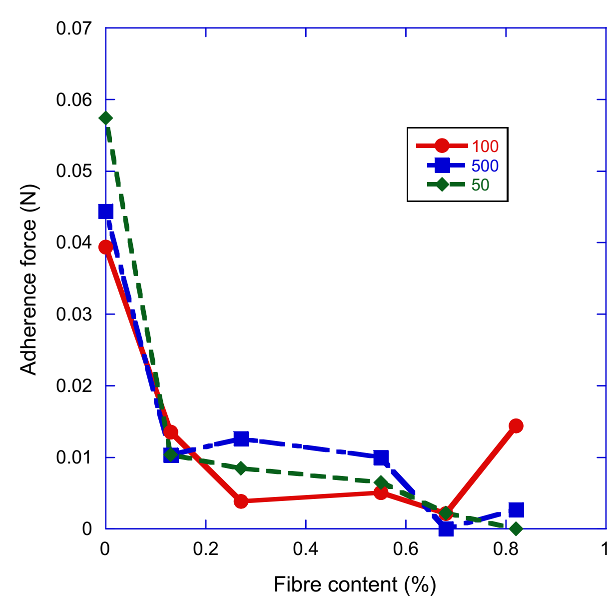

Fig. 6. Evolution of the adherence force versus fibre content for three different pulling velocities. (♦) 50 µm/s; (•) 100 µm/s; (■) 500 µm/s.

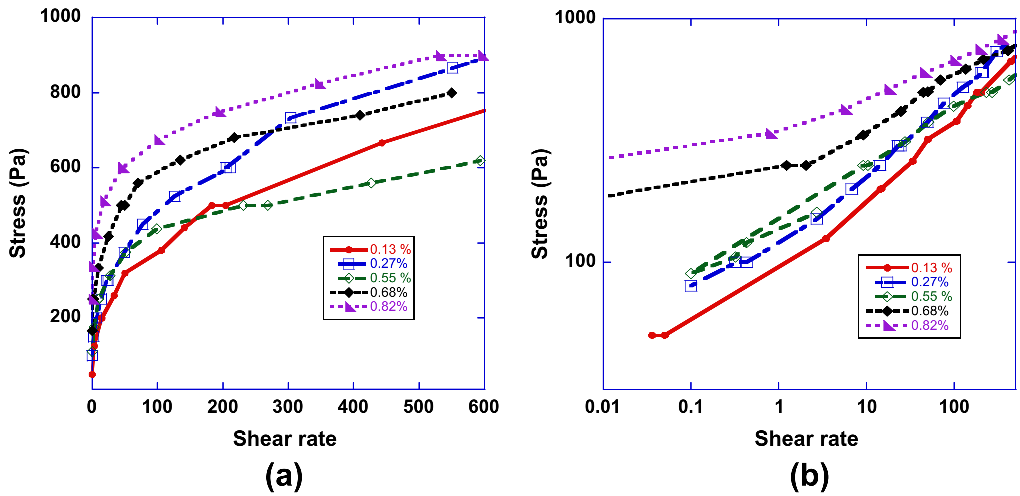

Fig. 7. Flow curves of the mortars for different fibre contents. (a) Linear plot; (b) logarithmic representation.

The general form of the flow curves indicates that the mortars behave as Herschel–Bulkley shear-thinning fluids. The corresponding rheological parameters are determined further. Examining the flow curves, we can observe an unexpected phenomenon: the flow curves cross over. This means that for some shear-rate and fibre concentration intervals, the apparent viscosity (stress divided by shear-rate) may decrease with fibre content. To the best of our knowledge, this phenomenon has never been reported in the literature. Fig. 8 shows the evolution of the apparent viscosity versus fibre content for three different shear-rates (low, intermediate and high).

At high shear-rates we can observe a minimum for the apparent viscosity for a fibre concentration of 0.55%. This minimum disappears at low shear-rates. A possible physical origin of the presence of these extrema in the evolution of the apparent viscosity versus fibre content may be the following. The presence of the fibres in the mortar may lead to two different antagonistic effects: on one hand they will increase the viscous dissipation since they resist flow gradients experienced by the liquid phase, but on the other hand they will locally increase the flow gradient and then decrease the viscosity of the mortar since it is shear-thinning. Then depending upon the value of shear-rate the presence of the fibre may lead to either increase or decrease of the global viscous dissipation (apparent viscosity).

3.2.2. Rheological parameters

The yield stress is measured directly by determining the applied stress for which we have a finite shear-rate. The evolution of the yield stress versus polymer dosage rate is represented in Fig. 9. As expected the yield stress increases with fibre content. However this increase is highly non-linear. Below a certain value of fibre content (around 0.55%) the yield stress has only a moderate dependence upon this additive. Beyond this critical content we obtain a huge increase of the yield stress. The existence of this critical value of fibre concentration may be related the appearance of a significant entanglement of the fibres leading to an interlocking and then a resistance to an initiation of the flow. If this is actually the case the critical concentration will then depend upon the geometry of the fibres (in particular their aspect ratio). A rheological investigation with different fibre sizes is needed in order to check this hypothesis.

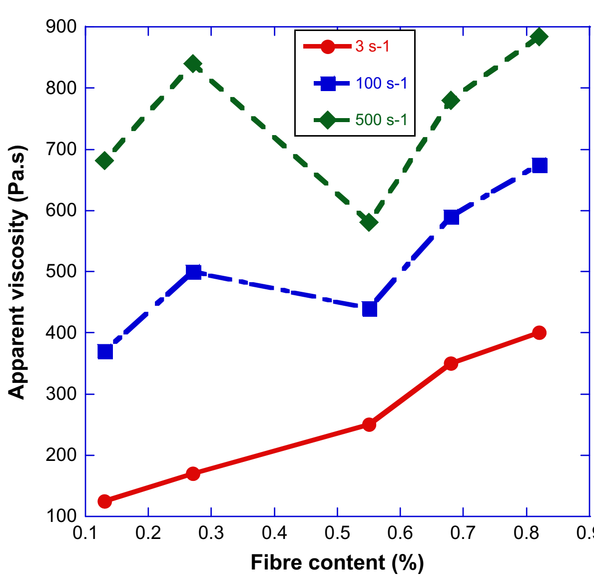

Fig. 8. Evolution of the apparent viscosity versus fibre content for different shear-rates.

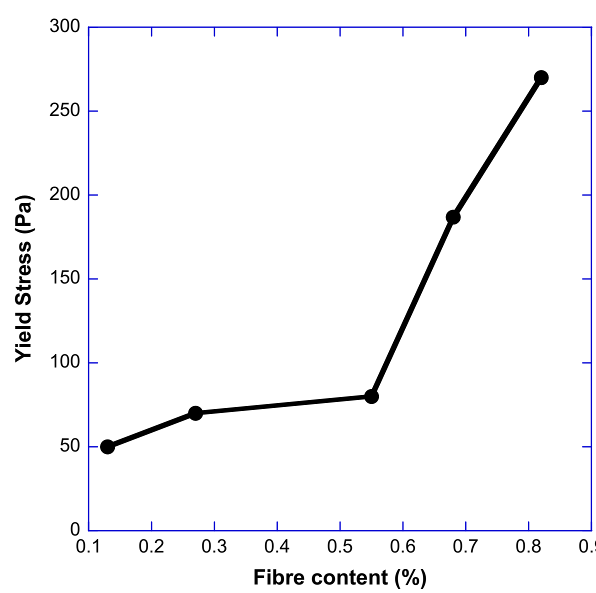

Fig. 9. Evolution of the yield stress versus fibre content.

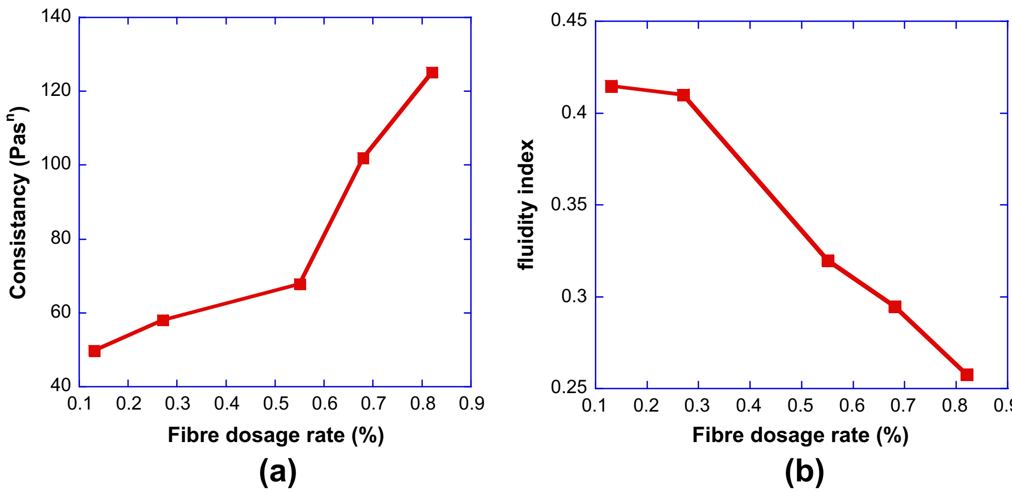

Fig. 10. Influence of the fibre dosage rate on the rheological parameters of the mortar, including the consistency (a) and the fluidity index (b).

Two other rheological parameters, including the consistency and the fluidity index, are determined by performing the best fit of the experimental results with the Herschel–Bulkley model. That is: $\tau = \tau_0 + k\dot{\gamma}^n$, where τ is the shear-stress and $\dot{\gamma}$ the shear-rate. The rheological parameters involved in this model include the yield stress τ_0, the consistency k, and the fluidity index n.

The evolution of the consistency and fluidity index versus fibre content is reported in Fig. 10. The behaviour of the consistency is very similar to that of the yield stress. A similar physical interpretation may be then put forward. The fluidity index decreases with fibre content, indicating that the material becomes more and more shear-thinning when adding fibres. The increase of the sensitivity of the stress to the shear-rate may be due to the flow induced deflocculation of fibre aggregates [21].

3.3. Linking the adhesive properties to the rheological behaviour

In tack tests the instantaneous distance between the plates is small compared to the sample diameter, in particular in the first zone (see Fig. 2) of the tack curves. We can then use the lubrication approach, in which one assumes that the flow is dominated by the shear component, to determine the adhesive force (F_{max}). For Herschel–Bulkley fluids this calculation has already been performed in the literature [22].

$$F_{\max} = \frac{2\pi R^3 \tau_0}{3h_m} + \frac{2\pi k}{n+3}\left(\frac{2n+1}{n}\right)^n \frac{R^3}{h_m}\left(\frac{RV}{h_m^2}\right)^n \tag{1}$$

R is the mortar sample radius, h_m the instantaneous distance between the plates corresponding to F_{max} and V the pulling velocity. The relationship (1) can be used to link the adhesive properties as determined with a tack test to the rheological parameters.

From expression (1) we can infer the cohesion force by setting the pulling velocity to zero, which gives:

$$F_{coh} = \frac{2\pi R^3 \tau_0}{3h_m} \tag{2}$$

In Fig. 11 the cohesive stress (as inferred from (2)) is compared to the yield stress for different dosage rates of fibres. These results indicate that the resistance of mortars with fibres is significantly higher in extension than in shear. This can be understood as the following: in shear flow the fibre tend to be orientated in the flow direction, perpendicular to the flow gradient. Consequently they exert quite low resistance to flow. On the other hand in an extensional flow (tack test) the fibres tend to be orientated in the direction of the extensional-gradient and may then contribute significantly to flow resistance.

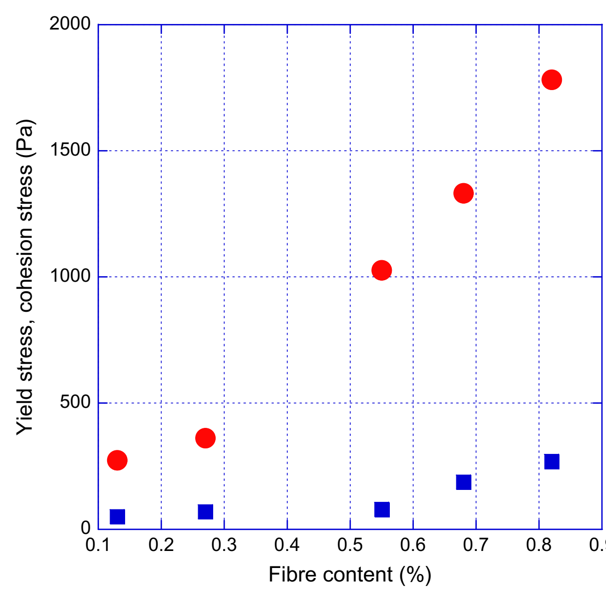

Fig. 11. Comparison between the yield stress (■) and the cohesion stress (•) for different fibre contents.

We cannot go further and make a comparison between the dynamic rheological properties, including the consistency and the fluidity index, as determined in shear flows and those corresponding to the tack tests. Indeed, in the tack tests the flow gradients involved are actually very low. The highest shear-gradient can be estimated as V_{max}/h_{min} = (1 mm/s)/(3 mm) = 0.33 s^{-1}, where V_{max} is the highest pulling velocity considered in the tack tests and h_{min} the minimum value of the gap (initial value).

4. Conclusion

Adhesive properties of joint mortars containing different dosage rates of cellulosic fibres were investigated using the probe tack test. From the measured tack force curves three different adhesive quantities were determined, including adhesion, cohesion and adherence. It was found that the evolution of the cohesion strength versus fibre content was highly non-linear: below a certain value of the fibre dosage the effect of the fibres is quite small; beyond that value we obtain a high increase of cohesion. Such behaviour was attributed to a probable transition to fibre entanglement and interlocking when increasing fibre content. In contrast, addition of fibres leads to the decrease of the mortar adherence to the support. From the practical point of view this implies the existence of an optimal dosage rate to use in order to sufficiently increase the cohesion (to avoid creeping) without diminishing the adherence to an undesirable level. To improve adherence one has to use other type of additives like redispersable resins or water-soluble polymers.

Finally, a qualitative comparison between adhesive and rheological properties was presented. It was found that the cohesion stress was correlated with the yield stress, but it was significantly larger. This was attributed to the difference between the extension-induced and shear-induced orientations of the fibres. Tack tests and rheological measurements can be considered to be then complementary to characterise placement properties of adhesives mortars. Further investigation, in particular by taking into account the fibre geometry, is needed to achieve quantitative interpretations of the tack test and rheological results.

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