Research Article

Salim Safiddine*, Kamel Amokrane, Farid Debieb, Hamza Soualhi, Benchaa Benabed, and El-Hadj Kadri

How quarry waste limestone filler affects the rheological behavior of cement-based materials

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Abstract: The main objective of this work is to highlight experimentally the effect of quarry waste limestone fillers (LF) on the rheological properties of concrete. All mixtures were made with water-to-cement ratio (W/C) of 0.50. The method adopted based on the rheological behavior of concrete incorporating LF as a partial substitute for sand in multi-scale systems (concrete, mortar, grout and LF suspension). The replacement ratio of sand by LF ranged from 0% to 20% (by weight of sand) in concrete and mortar. Whereas, the addition of LF varied from 0% to 60% (by weight of cement) in cement grout. The rheological behavior of concrete and mortar was modeled by the Bingham model with sufficient precision. However, the cement grout shows a nonlinear behavior, which was modeled by the Herschel-Bulkley model. Results indicate that the use of LF has a significant effect on the superplasticizer (Sp) demand of concrete and mortar, for a given slump on one hand. On the other hand, an increase in yield stress, apparent viscosity and flow time of cement grout was obtained when LF is incorporated, which results in a loss of fluidity of the suspensions of LF.

Keywords: Limestone filler, Yield stress, Viscosity, Grout, Mortar, Concrete

1 Introduction

Quarry waste limestone filler (LF) also called quarry dust is fine powder obtained during the process of crushing rock into crushed aggregates. In quarries, great amounts of dust are generated as by-products of rock crushers. Therefore, environmental impact has become a major problem for all quarries, especially with the increase in the use of crushed sand as replacement to depleted natural sand in the last few decades [1, 2].

Compared to ordinary Portland cement (OPC) and other supplementary cementitious materials (such as slag, fly ash, silica fume), LF only requires energy for quarrying and grinding. It is a key element of sustainable cement-based materials and less CO₂ emissions are associated with their production. In fact, this addition is now largely approved for improving the properties of concrete used for several applications and has been successfully used in a variety of civil engineering applications [3–5].

Usually, the LF complements the deficiency in the cement's particle size distribution, which can enhance both the workability and the stability of fresh concrete [6]. In reality, several factors can lead to different or even contradictory results, especially the mode of addition and the type of LF. Some authors [7–9] have found that the addition of LF improves the rheological properties. Whereas, the authors in references [10–15] found that the LF exhibit undesirable effects on the rheological properties of concrete. So, it is not easy to select the right mineral addition in practice.

Moreover, it has also been shown by several research studies that its effects could be advantageous for certain applications such as increasing viscosity for self-compacting concrete (SCC) in order to preserve its stability [5, 15]. In this case, LF can be considered as a valuable mineral addition from a technical, environmental and economic point of view. In particular, the use of fly ash in concrete in some countries is likely to be greatly affected in the future due to recent limits on coal combustion in power plants to reduce air pollution [3, 16, 17]. In this light, the addition of LF has an inherent advantage throughout the world due to its availability in large deposits.

***Corresponding Author: Salim Safiddine:** Materials and Environmental Laboratory (LME), University of Yahia Fares, 26000, Medea, Algeria; Email: safiddine.salim@gmail.com

Kamel Amokrane, Farid Debieb: Materials and Environmental Laboratory (LME), University of Yahia Fares, 26000, Medea, Algeria

Hamza Soualhi, Benchaa Benabed: Civil Engineering Research Laboratory (LRGC), University of Amar Telidji, P.O. Box 37G, Laghouat, Algeria

El-Hadj Kadri: Laboratory L2MGC, University of Cergy Pontoise, F9500, Cergy Pontoise, France

In the same way, the main causes of these different results are mainly related to the physical properties (morphology, fineness and specific surface), activity (mainly pozzolanic), filling effect (causing release of free water from the paste) and chemical composition of these mineral additions [6, 18–21]. According to Hawkins *et al.* [22], it is important to note that even a chemically inert material can have significant physical effects, such as the effects of improved particle packing and increased surface area on water demand. Also, small particle size distributions generally result in high water demand, while large particle size distributions lead to reduced water demand. Moreover, the experimental protocol adopted (as partial replacement of cement or sand, substitution by weight or volume), the time that passes after the first contact water-binder, mixer type and the mixture tested (grout, mortar or concrete) also have a very significant influence on the different results obtained. Furthermore, Felekoglu [15] found that there is a difference effect between the use of quarry waste limestone filler, with an extremely porous morphology and no smooth surface, or the use of limestone filler produced by direct grinding of limestone, with more smooth and flat surfaces. Lastly, the LF can be utilized as a replacement of cement clinker to reduce energy consumption and reduce CO₂ emission associated with cement production [23–29]. Thus, it may be part of the crushed aggregates, especially sand, used in concrete and mortar to reduce its environmental impact [10, 11, 13, 30, 31].

The main objective of this study is to explain how LF negatively affects the workability and rheology of concrete through the measurement of the rheological properties of its matrix as a function of LF content by different scales (mortar, grout and LF suspension). Since the LF particles bind to the aqueous phase of the concrete matrix.

The idea is inspired from the multi-scale theory [32]. The basic principle of the multi-scale theory is to admit that the material properties can be predicted from knowledge of both the properties of the matrix and its inclusions [33]. Applied to concrete, the multi-scale theory leads to identifying the cement mortar (sand, cement, LF and water) with the matrix and the gravel with the inclusion. As a result, the rheology of the concrete can be predicted from the rheological properties of cement mortar and the shape, size and volume fraction of gravel. However, cement grout is considered as matrix and sand as inclusion, for cement mortar.

Moreover, the rheology of concrete has become more and more important nowadays because of new placement technologies, especially new developed concretes with the use of various mineral and chemical admixtures. Their optimum combination to achieve desired rheological prop-

erties is challenging [18, 34]. In fact, the rheology is an efficient indicator for characterizing the properties of fresh concrete [35]. Actually, the performance of concrete in the hardened state depends on the rheological properties of the fresh state during placement into the formwork. Easy placement must be guaranteed while keeping the product in a homogeneous condition. According to [36], rheology can give precious and useful information about the properties of fresh concrete. It can lead us to achieve an optimization of the composition and how to attain it. In fact, the rheology models have been proven to be an efficient tool for predicting the fresh properties of cementitious composites [35]. Otherwise, the optimization is largely based on feeling.

A flow curve is defined as a plot of shear stress (τ) versus shear rate ($\dot{\gamma}$). In general, there are two main families of rheological behavior for fluids: viscous fluid and viscoplastic fluid (Figure 1). Newtonian fluids present the simplest case of the first family material. Furthermore, the rheologi-

Figure 1: (a) Flow curve for viscous and viscoplastic fluids, (b) graphic representation of differential viscosity and apparent viscosity determined at a certain shear rate.

cal properties of fresh cement-based materials have been studied in literature, showing they have a yield stress (τ_0) [24, 37], i.e., a viscoplastic fluids. The crucial property of this family is that it is able to deform indefinitely (i.e., flow) only if it is submitted to a stress above τ_0 [38, 39]. In other words, we must apply a stress greater than the yield stress before significant deformation can occur and the flow may begin. The simplest case of this family material is the Bingham fluid. For this fluid, after the start of flow (when $\tau > \tau_0$), there is a linear relationship between the shear stress and the shear rate with a constant of proportionality called plastic viscosity (μ). The flow behavior of Bingham fluid [40, 41] is described with the Equation (1):

$$\tau = \tau_0 + \mu \cdot \dot{\gamma} \quad (1)$$

Where:

τ : is the shear stress applied to the material; τ_0 : the yield stress (Pa) (Intersection of the linear curve with the shear stress axis); μ : the plastic viscosity (Pa s) (slope of the linear curve given by the plot of shear stress versus shear rate) and $\dot{\gamma}$: is the shear rate (s^{-1}).

It is usually agreed that fresh concrete and cement mortar can be considered as a Bingham fluid with good precision [40–44]. On the other hand, this model does not succeed for a high workability mix, such as SCC, or for cement grout. In this case, the decrease in yield stress can be so great that it approaches zero, and the extrapolated Bingham yield stress appears to be less than zero (negative), which is physically impossible [36]. Two models have been proposed to describe this relationship: the Herschel-Bulkley model (Equation 2) [39, 42–45] and the modified Bingham model [43–47] (Equation 3):

$$\tau = \tau_0 + K \cdot \dot{\gamma}^n \quad (2)$$

$$\tau = \tau_0 + \mu \dot{\gamma} + c \cdot \dot{\gamma}^2 \quad (3)$$

Where:

K : consistency factor ($Pa \cdot s^n$); n : flow index (-) and c : second order parameter ($Pa \cdot s^2$)

For the Herschel-Bulkley model [48–50], when $n < 1$, a shear thinning behavior and when $n > 1$, the equation signifies a shear thickening behavior, while for $n = 1$ a Bingham behavior. These behaviors are shown in Figure 2.

To explain these two behaviors, we must first define the following two terms: apparent viscosity and differential viscosity (Figure 1b) shows how to graphically calculate these two parameters at a certain shear rate. The differential viscosity is the inclination of the flow curve at a particular shear rate. It can be seen that for the case of a Bingham fluid, the plastic viscosity presents the differential viscosity.

Figure 2: Apparent viscosity versus shear rate.

Whereas, the apparent viscosity at a certain shear rate is the inclination of a straight line in a flow curve, connecting the origin with the point on the flow curve at the corresponding shear rate. It is simply determined by dividing the shear stress by the shear rate at that particular shear rate [51].

On the other hand, the evolution of the apparent viscosity as a function of the shear rate indicates the corresponding type of behavior (Figure 2). The apparent viscosity increases with increasing shear rate, which is called shear thickening. On the contrary, as the apparent viscosity decreases with increasing shear rate, it is called shear thinning [43–45].

2 Experimental program

2.1 Materials

All concrete, mortar and cement grout mixtures were prepared using an ordinary Portland cement (CEM I 52.5), conforming to EN 197-1 Standard. The limestone filler (LF), particles passing 80 μm (according to standard NF P 18-540), was recuperated from crushed limestone sand produced by a local quarry. Table 1 presents the chemical composition and physical properties of the cement and LF used. Whereas, washed crushed sand (CS) of siliceous nature with a maximum size of 5 mm was used, with density of 2.63 according to Standard EN 1097-6. It was chosen to avoid any additional LF content. Its fineness modulus is 3.3, according to Standard EN 933.2, and has an absorption coefficient equal to 4.5% according to Standard EN 1097-6. Whereas, the gravel used is silico-limestone petrography, class 6/20 mm, having an absorption coefficient equal to 1.8% and its density is 2.51.

Figure 3 presents the scanning electron microscopy (SEM) images of OPC and LF obtained with a microscope at 15.00 kV. OPC presents more regular particles with smooth

Figure 3: SEM of (a) cement and (b) limestone filler.

Figure 4: Particle size distribution of LF.

surface (Figure 3a). Likewise, LF grains are containing a significant fraction of particles of the order of 10 μm and below with angular shape and rough surface (Figure 3b). The particle size distribution of LF is given in Figure 4, which was measured using a Malvern laser diffraction instrument (ASTM E2834-12). It shows a bimodal distribution of the LF grains, they have a population of fines around 1.8 μm . It is strongly possible that it corresponds to the size of the crystals of the parent rock. Then, we can see a second pop-

Table 1: Chemical analysis and physical properties of cement and LF used.

Chemical analysis		
Element (%)	OPC	LF
SiO_2	20.23	02.50
Al_2O_3	04.29	02.63
Fe_2O_3	02.35	00.58
TiO_2	00.25	—
MnO	00.02	—
CaO	63.67	70.69
MgO	03.88	—
SO_3	02.80	—
K_2O	00.69	—
Na_2O	00.14	—
P_2O_5	00.31	—
Cl	00.02	—
LOI	01.63	24.77
Physical properties		
Specific density	03.13	02.60
Fineness Blaine (m^2/kg)	359	—
Fineness BET (m^2/kg)	—	1470
D_{10} (μm)	01.50	01.30
D_{50} (μm)	15.00	18.00
D_{90} (μm)	48.00	60.00

ulation appear around 40 μm which, this time would seem to correspond to the crush mode.

The superplasticizer (Sp) used in concrete is modified ether polycarboxylic, with 20% solid content, named SKY 537 by Glenium. Though, for the cement grout, the Sp used is based on sulfonated naphthalene formaldehyde copolymer, with 40% solid content and a density of 1.21, named Medaplast SP 40 by Granitex-NP. All dosages of Sp refer to the aqueous solutions and not to the solid fraction. Thus, the Sp was measured as percentage by weight of cement.

2.2 Mix proportion

The concrete composition was varied to gain insight into the effect of LF as CS replacement on rheological properties. In this investigation, the water/cement (W/C) ratio and the cement content were kept constant for all mixes (W/C=0.5). But, the dosage in Sp varies to keep a constant slump (10.0 ± 1.0 cm). This slump was targeted to avoid blocking the rheometer during the test. To study the effect of LF in concrete, samples were made by incorporating LF as a partial substitute for CS (5; 10; 15 and 20%, by weight). This choice covers the range of different categories of fines con-

Table 2: Concrete composition for 1 m³.

Concrete	LF (%)	Cement (kg/m ³)	CS (kg/m ³)	LF (kg/m ³)	Gravel (kg/m ³)	Water (kg/m ³)	W/C	Sp (%)
C1	0		850.70	0				0.8
C2	5		808.17	42.53				1.1
C3	10	350	765.63	85.07	953,1	175	0.5	1.6
C4	15		723.10	127.60				2.2
C5	20		680.56	170.14				2.6

Table 3: Concrete equivalent mortar composition.

Mortar	LF (%)	Cement (g)	CS (g)	LF (g)	Water (g)	W/C	Sp (%)	Slump (cm)	Spread (cm)
M1	0		1339.24	0			0.8	12.0	25.0
M2	5		1272.28	66.96			1.1	11.5	22.0
M3	10	525	1205.32	133.92	262.5	0.5	1.6	12.0	23.0
M4	15		1138.36	200.89			2.2	12.5	24.0
M5	20		1071.39	267.85			2.6	12.5	24.0

Table 4: Composition and designation of the cement grout.

LF/Cement (%)	0	15	30	45	60
Grout without Sp	G1	G2	G3	G4	G5
Grout with 1% of Sp	G1°	G2°	G3°	G4°	G5°
LF (g)	0	300	600	900	1200
Cement (g)			2000		
Water (g)			1000		
Flow time of grout with 1% of Sp (s)	53	122	—	—	—

Table 5: Composition and designation of the suspension of limestone filler.

LF suspension	S1	S2	S3	S4	S5
LF (g)	0	450	900	1350	1800
Cement (g)			0		
Water (g)			1500		
Solid volume concentration (%)	0	10.34	18.75	25.71	31.69
Flow time of suspension (s)	26.00	29.33	31.42	33.80	37.81

tent of the sands associated with normative specifications of aggregates for mortar and concrete (EN-12620, 2004 “Aggregates for concrete”). The composition of the concrete is summarized in Table 2.

Whereas, the compositions of concrete equivalent mortar (CEM) is summarized in Table 3. The CEM is derived from their corresponding concrete mixtures by eliminating the coarse aggregate fraction and replacing it by a certain mass of sand of the same surface area, according to the authors in references [52, 53].

In order to understand the influence of the LF contents in the CS on the rheological behavior of concrete matrix, an evaluation of their effect on the fluidity of a suspension containing only the LF, as inclusions, and the aqueous phase (water) as a matrix would be of interest. Then, in the cement grout, we consider cement and water as a matrix and the LF as inclusions, with a constant W/C ratio for all suspensions.

So, cement grout is a suspension of cement and LF in water, with a W/C ratio equal 0.5. Four LF contents compared to the weight of the cement (15, 30, 45 and 60%) were

added, and not substituted, to the control composition (0% LF) to study their effect on the rheological characteristics. The objective is to study the influence of LF on the rheological properties of the grout, due to the contribution caused by the LF content in the CS in the case of concrete, while keeping a cement dosage and a ratio W/C constant. In other words, we make a concrete equivalent grout. Table 4 illustrates the composition and designation of the cement grout studied.

The last scale studied is the LF suspension, contains only LF (as solid particles) and water (as matrix). The composition of the LF suspension is given in Table 5.

2.3 Mixing procedure

As concrete rheology can be affected by the mixing procedure, the same mixer and the same mixing sequence were used for all concrete compositions. A concrete mixer was used according to the following procedure: introduce gravel, sand including fines and cement; then, dry mixing for 1 min; lastly, introduce the water and the Sp, for 1 min during mixing, and continue mixing for 2 min and 30 sec.

With regard to the CEM, a mixer according to European Standard EN 196-1 at stage I (62.5 rpm) was used. The dry mortar constituents (sand, cement and LF) were mixed for 1 minute. The water with Sp was added progressively during the next 30 seconds. After continuing the mixing for 1 minute it was stopped for 30 seconds in order to scrape off the material that had adhered to the sides of the mixing bowl. After that, it was followed a second mixing for 1 minute. The same method was adopted for the cement grout and the LF suspension, without adding the sand for the first and again without the cement for the second.

2.4 Test methods

2.4.1 Concrete and mortar cement

For the concrete, a slump test and rheological test were performed in order to characterize the rheological behavior of the different concretes.

In the first test, a cone with an upper diameter of 100 mm, a lower diameter of 200 mm and a height of 300 mm, according to Standard EN 12350-2, was used. The cone was gently lifted and the slump was measured. For the mortar, a cone with a half of the values given in this standard was used to measure the slump and the flow spread.

In the second test, the rheological properties were measured. Fresh concrete is considered here as a fluid. There-

Figure 5: Rheometer and the steel vane for (a) concrete and (b) cement mortar.

Figure 6: The profiles of the imposed rotational speed used in (a) concrete and (b) mortar rheology measurements.

fore, fluid rheology methods are used to describe the concrete flow.

The rheological properties of the concrete were measured on the same mixtures as concrete slump test. They were determined by a new rheometer apparatus developed and validated by Soualhi *et al.* [54]. The used rheometer is composed of three main parts (Figure 5a): an agitator with a torque recorder; a cylindrical container of 30 cm diameter and 25 cm height and a steel vane (4 solid blades in double U shape) with a height of 15 cm and a diameter of 10.5 cm. The agitator is driven by a computer using software (watch & control). In order to optimize the adhesion of the concrete to the inside wall of the container, 18 square steel sticks of dimension $9 \times 9 \text{ mm}^2$ have been welded. The measurements are performed using the rotational speed profile defined in Figure 6a.

The objective of the measurements is to determine the rheological parameters τ_0 and μ from the obtained measured torques ($M_i - \Omega_i$), considering concrete as Bingham fluid (Equation (1)) [34, 42, 55].

The calculation of the Bingham model parameters (yield stress and plastic viscosity) is based on the Reiner-Riwlin equation (Equation (4)) [56]:

$$\Omega = \frac{M}{4\pi h\mu} \left(\frac{1}{R_1^2} - \frac{2\pi h\tau_0}{M} \right) - \frac{\tau_0}{2\mu} \ln \frac{M}{2\pi h\tau_0 R_1^2} \quad (4)$$

Where:

Ω : rotational speed (rad/s); M : torque (N m); h : vane height (m); R_1 : vane radius (m); R_2 : outer container radius (m); μ : plastic viscosity (Pa s); τ_0 : yield stress (Pa).

However, the rheological properties of the mortar were measured by the same rheometer with the following characteristics: Rotational speed from 4 to 540 rpm (± 1 rpm) and maximum torque of 740 Ncm (± 0.1 Ncm); the vane dimensions are 10 cm in height and 5 cm in diameter; the container is a cylinder of 10 cm in diameter and 13 cm in height (Figure 6b and 5b). The calculation of the Bingham model parameters is based on the method used in [57].

2.4.2 Cement grout and LF suspension

For the cement grout and the LF suspension, a Marsh cone test and rheological test were performed in order to characterize their fluidity.

In the first test, according to European standard EN 445, the flow time was measured. The cone used has an upper diameter of 150 mm and total height of 355 mm, with length and internal diameter nozzle 50.8×4.76 mm (Figure 7). To be more rigorous in the measurements, we followed the recommended use of a volume of grout of 1500 ml and

taking the flow time corresponding to the time of 946 ml flow.

Figure 7: The Marsh cone test.

In the second test, the rheological properties of cement grout were measured using a rotary rheometer, AR2000 from TA Instruments, with vane geometry (Figure 8). It is controlled by TA Data Analysis software, which enables the execution of the imposed speed profile (Figure 9) and the processing of the data supplied by the measuring equipment. The software also allows the calculation of the various rheological parameters according to the modeling chosen for the flow as well as an instant visualization of the

Figure 8: Rheometer AR 2000 with a geometry vane.

Figure 9: Profile of the imposed rotational speed used in grout rheology measurements.

flow curve. In our case, the Herschel Bulkley model is the most suitable with the rheological behavior of the cement grout studied.

A 30 ml sample for the vane geometry was used, according to the manufacturer's recommendations. Then, pre-shearing at 50 s^{-1} of the sample for 60 seconds, in order to deconstruct the material and obtain an identical reference state. After a 30 second rest, an increasing ramp of imposed shear rate up to 430 s^{-1} is applied gradually over 5 minutes. The rheometer is equipped with a bath used to condition and maintain a constant temperature at 20°C during the tests.

3 Results and discussions

3.1 Effect of LF on rheological properties of concrete

Figure 10 shows the results obtained by rheological test of concrete with different contents of LF in the CS. As it can be seen, the evolution of the torque with rotational speed is linear for all the compositions tested. To this end, the conversation towards the fundamental unit is done perfectly with the Bingham model. The rheological properties (the yield stress and the plastic viscosity) thus obtained from this model are given in Figure 11. The composition adopted aims at a constant slump ($10.0 \pm 1.0 \text{ cm}$) for the five concretes tested by varying the content of Sp.

Figure 10: Evolution of the torque with rotational speed of concrete with different percentage of LF contents in the CS.

On the other hand and as it is shown in Figure 12, the increase in the amount of LF in the CS means a greater Sp admixture demand. So, the demand for Sp admixture clearly shows that the increase in the content of LF in the CS

Figure 11: Effect of LF content in the CS on the evolution of the rheological properties of concrete with different LF contents in the CS.

negatively affects the workability of the concrete. Whereas, the addition of Sp reduces the yield stress and the viscosity, i.e. improves the workability, by favoring the dispersion of cement particles due to the electrostatic repulsion and the steric effect, i.e. repulsive particle-particle interactions [58, 59]. These results are in good agreement with those already published [10–14, 60, 61].

Figure 12: Dosage of superplasticizer required for concrete to achieve the target slump flow.

Moreover, the decrease in workability may be attributed to the angular and irregular shape of LF grains, as shown in Figure 3, and their high fineness, which can enhance the friction between the solid grains of the mixture [13]. In addition, according to [60], as the LF has a large specific area ($1470 \text{ m}^2/\text{kg}$) also an average size comparable to cement (Table 1), the decreased of workability can also be attributed to the LF's porosity which leads to the increase in water absorption and therefore reducing the effective W/C ratio.

Furthermore, the improvement in the rheological properties of concrete with 20% of LF is due to the high dosage of Sp in its composition (Table 2), which provides good dispersion even in the presence of a high content of LF particles in the sand.

3.2 Comparison of the rheological behavior of concrete and its concrete-equivalent mortar

The measurement of the rheological properties of CEMs shows a linear behavior with a sufficiently acceptable precision by calculating the rheological parameters of Bingham. Figure 13 shows that the evolution of the yield stress and the plastic viscosity of concrete and its MBE, as a function of the LF content in the CS, is almost identical. For this purpose, we can say that the use of the MBE method is very promising. On the other hand, this result encourages us to continue our research towards smaller scales to better explain the mechanism of the influence of LF in the concrete matrix on the rheological behavior of concrete.

Figure 13: Correlation between (a) the yield stress and (b) the plastic viscosity of concrete and its CEM.

3.3 Effect of LF on rheological properties of cement grout and LF suspension

Figure 14 shows the evolution of the LF suspension flow time, of 950 ml through the Marsh cone, as a function of the solid volume concentration of the LF grains in the suspension. The figure shows that the flow time increases with the increase in the solid volume, i.e., the flow velocity decreases with increasing LF content in the water. So, the presence of the LF grains in the water makes the suspension more viscous. Given that the water (with 0% volume solids) is initially considered a Newtonian liquid with a flow time of 26 s, this value increases by almost 50% when the solid volume concentration reaches 34%. This can be explained by the increased interaction between the LF particles in the suspension. It should be noted that this increase was recorded without the presence of cement (active binder).

Figure 14: Influence of solid volume concentration on the flow time of LF suspension.

On the other hand, to fully explain the effect of the LF content in CS on concrete, we will study the rheological behavior of the cement grout. The fine fraction content in this suspension shows the amount of LF that is found in the CS used in the concrete. In this way, we will study a matrix similar to that of concrete based on CS.

In the same way as with the LF suspensions, we measured the flow time of the cement grout through the Marsh cone. Five LF suspensions were used to study the effect of LF on the rheological properties: 0, 15, 30, 45 and 60% in dry LF relative to the weight of the cement, without changing the W/C ratio. In this way, we approximate as close as possible the actual composition of the concrete matrix. Indeed, the use of a CS containing a certain percentage of LF leads to adding this quantity to the cement, constructing what is called a binder. For this reason, we added the LF to the control cement grout (G1) in order to assimilate the

actual composition of the concrete matrix according to the LF content.

In the case of grout without Sp, a blockage at the outlet of the Marsh cone was found. According to [62], the pressure gradient created by the grout weight above the nozzle may not be sufficient for the shear stress to overcome the yield stress in the nozzle. On the other hand, the addition of Sp made it possible to take the measurement of the flow time for the two grouts G1° and G2° (Table 4). Whereas, the flow of G3°, G4° and G5° gradually stops before the container is completely filled, so the cone becomes useless. The increase of the flow time can also be explained by the increase of inter-particle friction due to the increase in solid-solid contact [61], because we will be increasing the solid volume concentration of the suspension by adding LF. So, we can say that LF affects the workability of suspension of LF and cement grout even in the presence of Sp.

Unlike the Marsh cone test, we were able to perform rheological tests on grout without Sp for four different levels of LF. However, there was a blockage of the rheometer vane for the grout G5, containing 60% of LF. In addition,

Figure 15: Effect of LF content on the evolution of the shear stress with shear rate (a) without Sp and (b) with Sp of the cement grout.

we were able to carry out the tests on the five Sp grouts. However, the model of Bingham leads to the worst fitting for the cement grout. These results are in good agreement with those already published [38, 39, 45].

The flow curves in Figure 15 and 16 show that cement grout is non-Newtonian fluid, as for mortar and concrete. They are viscoplastic fluids with a non-linear appearance and a yield stress. The most popular model of these flow curves is that of Herschel-Bulkley. It can also be seen that all the grouts show shear thinning behavior, with a decrease in the apparent viscosity (Figure 16). On the other hand, the grout without LF and Sp (G1) can be likened to a Bingham fluid. However, the grout with Sp and without LF (G1°) can be likened to a Newtonian fluid where the yield stress tends towards zero.

Regarding the effect of increasing of LF content, it is clear that the rheological properties of the grout are affected. The volume concentration of LF has an influence on the

Figure 16: Effect of LF content on the evolution of the apparent viscosity with shear rate of the cement grout (a) without Sp and (b) with Sp.

curvature of the flow curves. However, the yield stress, intersection of the curve with the shear stress axis (Figure 15), increases with increasing LF content in the grout.

The identification of the parameters of the Herschel-Bulkley model (Table 6) is carried out by optimizing using the conjugate gradient method, which consists of minimizing (for a given shear rate) the quadratic difference between the value of the measured stress and that calculated by the model. Figure 17 shows an example of modeling the flow curve of G4°. The figure illustrates the experimental appearance and its modeling under the Herschel-Bulkley model. In the same way, using the results obtained, we were able to extrapolate the values of the yield stress for a zero shear rate according to the Herschel-Bulkley model, which are summarized in Table 6.

Table 6: Parameter values of the Herschel-Bulkley model.

Grout	ϕ (%)	τ_0 (Pa)	η (Pa s)	n
G1	39.2	19.40	0.14	0.36
G2	43.2	45.90	0.31	0.32
G3	46.7	133.40	0.70	0.17
G4	49.8	236.80	0.96	0.08
G5	52.5	-	-	-
G1°	39.2	0.18	0.04	1.03
G2°	43.1	1.55	0.09	0.75
G3°	46.6	32.19	0.33	0.37
G4°	49.6	59.03	0.55	0.29
G5°	52.4	222.80	0.65	0.02

Figure 17: Evolution of the shear stress and the apparent viscosity with shear rate of the cement grout G4°.

The “n” parameter of the model is the flow index. This parameter is less than 1.0 (Table 6) for all cement grouts studied, except for the case of the grout G1° (with Sp and without LF) which shows a behavior very close to Newton’s fluid with a coefficient $n = 1$ and an almost zero yield stress. While, the behavior of the grout G2° is practically linear viscoplastic and could be modeled by model of Bingham. These results correspond to a shear thinning behavior of a viscoplastic fluid (with a yield stress). The evolution of the rheological parameters of the grout shows that their behavior deviates more and more from viscous behavior by increasing the LF content (Figure 1 and 15). In parallel, the curve of the evolution of the apparent viscosity moves further and further away from the Newtonian plateau (Figure 2 and 16). In addition, the apparent viscosity and the yield stress increase with increasing LF (Table 6) over the entire range of shear rate tested. This shows that the addition of LF in the grout composition negatively affects the rheological properties of the cement grout without and with Sp. So the LF particles also bind to the aqueous phase of cement grout matrix, which resulted in loss of workability.

4 Conclusion

In this paper, a study on the effect of LF on the rheological behavior of cement-base materials was carried out. Based on the obtained results, the main conclusions can be listed as follows:

- For a given slump, the LF content in the sand has a significant effect on the Sp demand of concrete, due to high surface area of LF particles, their rough texture and their angular shape. This effect is also observed for the other three systems tested (mortar, grout and LF suspension). The workability of the systems was affected when LF is incorporated. This results in an increase in Sp demand, a decrease in the mini-slump and the spread diameter of the mortar, and an increase in the flow time, the apparent viscosity and the yield stress of the cement grout as well as a decrease in the fluidity of the suspensions of LF. The LF particles bind to the aqueous phase of the mortar and the grout matrix, resulting in loss of fluidity.
- The results show that the measurement of the rheological properties of simple scales such as mortar or cement grout, which requires small quantities of materials, can provide information about the behavior of concrete.
- The rheological behavior of concrete and mortar was modeled by the Bingham model with sufficient pre-

cision. However, the cement grout shows a nonlinear behavior, which was modeled by the Herschel-Bulkley model.

- The evolution of the rheological properties of the concrete, mortar and cement grout studied based on LF, in particular the presence of yield stress and the decrease in the apparent viscosity with increasing shear rate, shows viscoplastic fluids with shear thinning behavior.
- The use of Sp improves the workability of concrete with a high LF content without adding water, in order to avoid the undesirable effects of high W/C ratio on concrete, especially at the hardened state. This has an advantage for the wide use of LF in the production of structural concrete.

However, further experimental research is needed to establish a comparative study using filler produced by direct grinding of limestone. Then, repeating this same work by substituting cement with LFs by volume.

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