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Why is fresh self-compacting concrete shear thickening?

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ABSTRACT

The rheological properties of fresh concrete are mostly described by means of the Bingham model. For self-compacting concrete, the Bingham model is applicable in a lot of cases, but some authors report that the rheological behaviour is non-linear. The apparent viscosity increases with increasing shear rate and the SCC shows shear thickening behaviour. Shear thickening becomes important in operations occurring at high shear rates, like mixing and pumping. In these cases, shear thickening should not be forgotten in order to avoid breaking of the mixer, pump or pipes.

This paper will describe two possible theories for shear thickening behaviour of SCC, based on results published in the rheology literature. The first theory consists of the formation of so-called (hydro-)clusters, which are temporary assemblies of small particles. These clusters start being formed from a certain shear stress on: the critical shear stress. They cause the viscosity to increase with increasing shear rate. A second theory is based on grain inertia, where a part of the shearing force is transmitted through direct momentum transfer between solid particles. Results on cement pastes prove that the grain inertia theory is not the main cause of shear thickening in self-compacting concrete. The influence of several parameters on the shear thickening behaviour of SCC can be well explained by means of the cluster theory.

1. Introduction

Concrete is described by rheologists as one of the most difficult materials to study. Nevertheless, the rheological properties of fresh concrete have been studied extensively in literature, showing that concrete is a thixotropic material [1–3], having a yield stress and a plastic viscosity [1,2,4], and that time influences these properties due to hydration [2]. Until now, the most applied rheological model for the steady-state rheological properties, in which thixotropy and loss of workability are not incorporated, is the Bingham model (Eq. (1)) [1,4,5]:

$$\tau = \tau_0 + \mu \cdot \partial\gamma / \partial t \qquad (1)$$

where:

τ = shear stress (Pa)
τ_0 = yield stress (Pa)
μ = plastic viscosity (Pa s)
$\partial\gamma/\partial t$ = shear rate (1/s)

The influence of changes in mix proportions of concrete on the rheological properties, especially yield stress and viscosity, has been evaluated in great detail [5,6]. Superplasticizers, which are necessary to produce self-compacting concrete, cause the yield stress to decrease [5–7]. In order to maintain the stability of the SCC, extra materials can be added to increase the viscosity [7–9]. In Western-Europe, fine powders are mostly applied, like limestone fillers, fly ashes, silica fume, … as a consequence, the amount of fine materials in this type of SCC is quite high, while the total water amount is still low.

When focusing on rheology again, the decrease in yield stress, caused by the addition of the superplasticizers, can be so large that it approaches zero. It has been reported that in some cases, the extrapolated Bingham yield stress appears to be negative, which is physically impossible [10–12]. Detailed investigations of the flow curves, after elimination of thixotropy and loss of workability show a non-linear relationship between the shear stress and the shear rate: an increase in apparent viscosity with increasing shear rate [11–14]. Two models have been proposed to describe this relationship: the standard Herschel–Bulkley model (Eq. (2)) and the more alternative modified Bingham model [11,14,15] (Eq. (3)):

$$\tau = \tau_0 + K \cdot (\partial\gamma / \partial t)^n \qquad (2)$$

$$\tau = \tau_0 + \mu \cdot \partial\gamma / \partial t + c \cdot (\partial\gamma / \partial t)^2 \qquad (3)$$

where:

K = consistency factor (Pa s^n)
n = flow index (–)
c = second order parameter (Pa s^2)

A discussion about the advantages or disadvantages of these two models has been given in [14] and [16], in which one of the arguments was to find the physical meaning of all terms in the equations. Based

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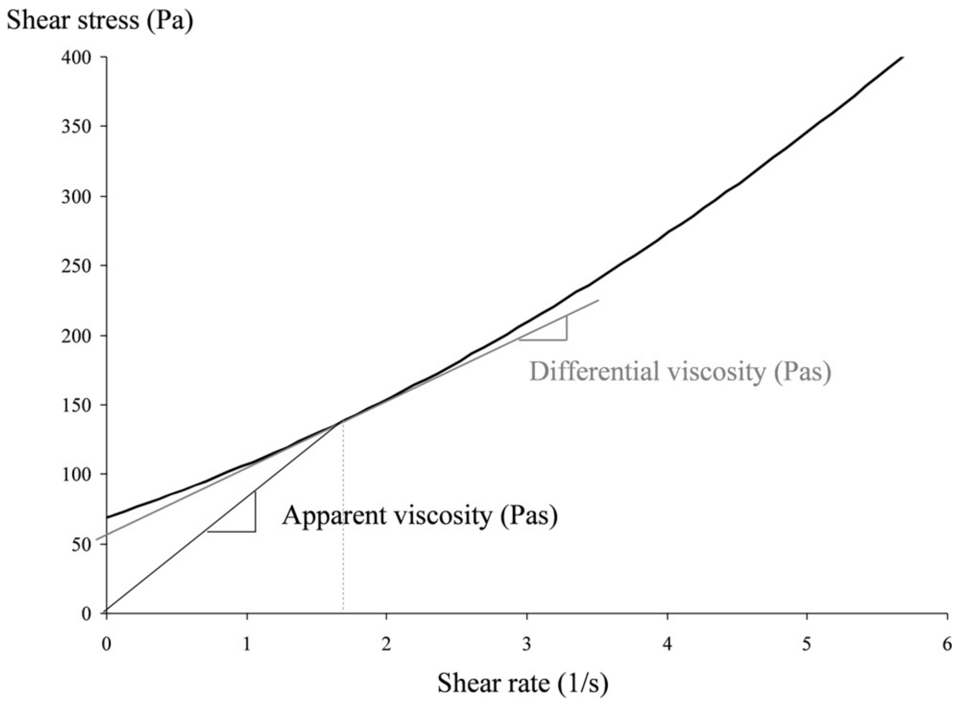

Fig. 1. Difference between differential viscosity and apparent viscosity, determined at a certain shear rate.

on the findings in this paper, the discussion on the physical meaning of the terms in Eqs. (2) and (3) will be continued at the end.

This paper will describe the physical background of shear thickening behaviour in general, based on rheological measurements and simulations described in literature. The derived theories will be tested qualitatively to the results of a large series of rheometer tests performed on different concrete compositions [11], mortars and cement pastes. The authors hope to provide a clear theory, explaining why fresh SCC behaves as a shear thickening material. It is not in the author's intentions to give a quantitative description and prediction of shear thickening, but more an insight into the physical causes and influencing parameters.

One might wonder why shear thickening behaviour is so important. To be honest, shear thickening is something causing more disadvantages than advantages [17]. As the (apparent) viscosity increases with increasing shear rate, a larger increment in energy will be needed to further accelerate the flow of the material. Especially in processes where high shear rates are applied (mixing, pumping, extrusion, …) shear thickening can become significant and even dominant. Controlling shear thickening behaviour is consequently necessary in order to avoid breaking of any system.

In order to be consistent in this paper, two different definitions of viscosity will be applied. The differential viscosity at a certain shear rate is defined as the parameter μ in the Bingham and $\mu + 2 \cdot c \cdot \partial\gamma/\partial t$ in the modified Bingham model. It is the inclination of the flow curve at that particular shear rate [18]. 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 [18]. It is easily determined by dividing the shear stress by the shear rate. The nomenclature "apparent" viscosity will be used in this paper in order to make a clear distinction between the real (apparent) viscosity used in rheology and the best known definition of viscosity in concrete rheology: the plastic (Bingham) viscosity. The difference between differential and apparent viscosity is illustrated in Fig. 1.

2. Physical background of shear thickening

Shear thickening is described in literature as "an increase in (apparent) viscosity with increasing shear rate" [17]. Two different theories are considered to be applicable on self-compacting concrete. One is based on the formation of clusters and the other is based on grain inertia. These theories are both qualitatively described in the following sections, without going in detail into formulae and calculations. In case of concrete, quantification of shear thickening is a very hard task due to the large polydispersity of the material (particle size varies in the range of 0.1 μm–1 cm).

2.1. Cluster formation

2.1.1. Theory

The apparent viscosity curve as a function of the shear stress for concentrated monodisperse "near-hard-sphere" silica ($d = 600$ nm) suspensions is shown in Fig. 2 [19]. It can be seen that for low volume

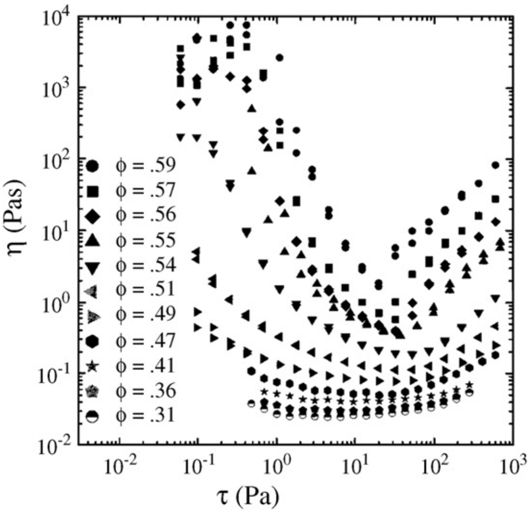

Fig. 2. The apparent viscosity as a function of shear stress shows, for high volume fractions, a decrease at small shear stresses (shear thinning) and an increase at large shear stresses (shear thickening), having an increasing importance with increasing volume fraction. Figure from [19].

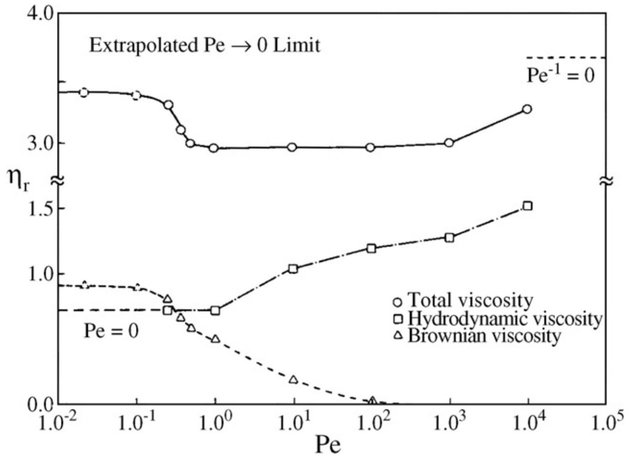

Fig. 3. The importance of the Brownian viscosity decreases with increasing Péclet number (dimensionless shear rate), which proves that shear thickening, starting in this figure at $Pe = 10^3$, is a purely hydrodynamic phenomenon. Figure from [26].

fractions (low φ) of particles, the apparent viscosity does not change significantly with the shear stress. For higher concentrations on the other hand, a clear decrease in apparent viscosity is observed at low shear stresses (shear thinning) and an increase in apparent viscosity at high shear stresses (shear thickening). During a long time, several authors described shear thinning due to shear induced ordering of the particles, where the Brownian forces acting on the particles are dominated by the hydrodynamic forces and as a result, the particles cannot reorganise into a random packing [20,21]. The flow is caused by shearing several layers of particles with only interstitial fluid in between the layers. At the onset of shear thickening, the layers start to disappear and shear induced disordering was thought to be the cause of shear thickening [22–25].

Recently, several authors have proven that this effect is not necessary to obtain shear thickening behaviour. Even when no shear induced ordering has been measured, shear thickening has occurred [26,27]. The shear thickening is caused by the high hydrodynamic (lubrication) forces between the monodisperse, spherical particles, overcoming the particle repulsion forces and forming temporary assemblies of particles, named (hydro-)clusters [19,26–28]. In time, particles can join and leave the cluster, causing this phenomenon to be transient. If a cluster is formed through the entire sheared zone, jamming can occur, which stops the flow [29–31]. Jamming leads to a sudden increase in apparent viscosity with a few orders of magnitude, blocking the flow and possibly destroying the measurement system.

Shear thickening starts at the critical shear stress (which appears to be a more fundamental unit than a critical shear rate) at which the ratio of the hydrodynamic forces to the repulsive forces (which can be Brownian, electrostatic or steric) becomes larger than one [19,26,28]. From this shear stress on, the relaxation time of the repulsive forces is too large compared to the hydrodynamic forces, causing the particle to remain in the cluster [32,33]. If the applied stress is decreased under the critical shear stress, the repulsive forces dominate the hydrodynamic forces, causing the clusters to disappear. This shear thickening behaviour is fully reversible.

In [26], it is proven that shear thickening caused by the formation of clusters is a purely hydrodynamic effect. In Fig. 3 [26], the total apparent relative viscosity is divided into a Brownian part and a hydrodynamic part in a range of several orders of magnitude of the Péclet number (Pe), which is a dimensionless shear rate (Eq. (4)).

$$Pe = \frac{\eta_s \cdot (\partial\gamma / \partial t) \cdot a^3}{k \cdot T} \tag{4}$$

where:

η_s = apparent viscosity of suspending medium (Pa s)
$d\gamma/dt$ = shear rate (1/s)
a = particle radius (m)
k = Boltzmann's constant = 1.38 10^{-23} (J/K)
T = temperature (K)

It can be seen that at low Pe-numbers (lower than 1), the Brownian viscosity dominates. For higher Pe-numbers, the importance of the Brownian viscosity decreases, in favour of the hydrodynamic viscosity. In Fig. 3 [26], it can be seen that shear thickening starts around a Péclet number of 10^3, where the Brownian viscosity has no importance anymore. Consequently, shear thickening (due to cluster formation) is a purely hydrodynamic phenomenon. Still, Bossis and Brady mention that at these high Péclet numbers where shear thickening occurs, the Brownian forces still show large enough peaks to provide the particles with sufficient energy to leave the cluster and causing these to be transient in time [26].

2.1.2. Influence of particle size

The repulsive forces between Brownian particles become larger as the particles become smaller. Depending on the type of repulsive force, the critical shear stress can scale on the particle radius to a power between −1 and −3 [17,19,32]. In each case, as long as the particle size remains in the colloidal range, increasing particle size in a monodisperse concentrated suspension causes shear thickening to occur earlier, which can be clearly seen in Fig. 4 [28]. On the other hand, particle size does not significantly influence the intensity of shear thickening, which can be defined as the increase in apparent viscosity for a certain increase in shear stress. The larger this increase, the more severe the shear thickening (larger ‘n’ in Herschel–Bulkley and larger c/μ in modified Bingham [14]).

2.1.3. Influence of volume fraction

As it has been shown in Fig. 2 [19], the intensity of shear thickening increases with increasing volume fraction of suspended Brownian particles [17,19,28,31]. In Figs. 2 and 4, it can be seen that the volume fraction does not influence the critical shear stress significantly [19,28].

2.1.4. Influence of bi- or polydispersion

As mentioned in the previous sections, the results have been mainly obtained for monodisperse suspensions of spherical particles. Bender and Wagner [27] mention that mixing two monodisperse suspensions increases the critical shear stress and decreases the intensity of the shear thickening. Similar results have been mentioned by Barnes [17] for polydisperse suspensions. This effect is attributed to

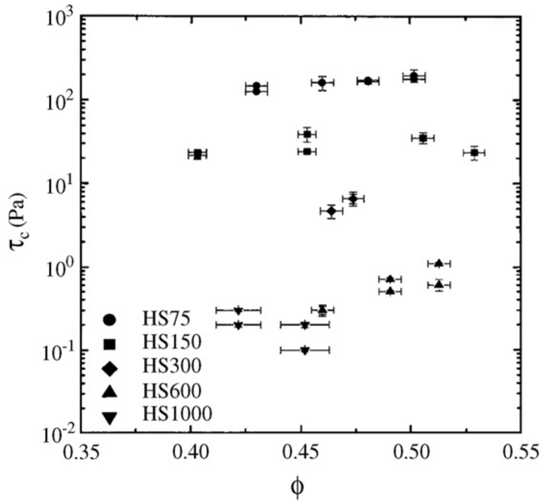

Fig. 4. Critical shear stress (τ_c) shows no significant dependency on the volume fraction (φ), but a large dependency on the particle size (varying in this graph from 75 to 1000 nm). Figure from [28].

the increase in maximal volume fraction due to the presence of other particle sizes. At equal (absolute) volume fraction, there is more space for flow in case of a polydisperse suspension compared to a monodisperse suspension, resulting in lower hydrodynamic forces between the particles [27].

2.1.5. Influence of particle shape

The results in previous sections have only been obtained for spherical particles. In [31,34], the influence on non-spherical, ellipsoidal particles on the critical shear stress is studied. The particles had aspect ratios from 1/2 to 1/7. At very low shear stresses, where the Brownian forces are dominant, the particle alignment is quite anisotropic, except for the suspensions with high volume fractions. Increasing the shear stress causes particle alignment in the flow direction and the suspension exhibits shear thinning behaviour. Further increase of the shear stress, above the critical shear stress, causes only a slight loss of alignment of the particles. As a result, the particles do not lose significantly their orientation and do not tumble in the flow direction [31,34]. Instead, the shear thickening is caused by the same phenomenon as for spheres: cluster formation. The parameter influencing the critical shear stress is the small radius of the particle, similar to the radius for spheres. On the other hand, shear thickening is more severe at lower volume fractions with increasing aspect ratio. This is due to a decrease in maximal volume fraction, creating less space available for the flow.

2.1.6. Influence of polymer coatings

Polymer coatings change the nature of the repulsive forces, which has directly an influence on the critical shear stress [32]. The solvent permeability of the polymer coating influences in addition the hydrodynamic forces. Brownian particles, coated with polymer brushes (similar to the addition of superplasticizers in cement based mixtures) exhibit a higher critical shear stress at equal particle size [35].

2.1.7. Influence of flocculated particles

In [17, and references herein], Barnes mentions, based on a large series of results, that flocculated systems do not have a tendency to show shear thickening behaviour. Concrete in general, can be regarded as a flocculated (or coagulated) system [1–3], but due to the addition of the superplasticizers, the degree of flocculation in SCC is much lower than in traditional concrete. This will be further discussed in Section 4.3 when dealing with the influence of slump flow and SP-content on the shear thickening behaviour. Furthermore, special attention will be paid to the estimated interaction between thixotropy, causing flocculation or coagulation, and shear thickening in Section 5.3.

2.1.8. Conclusion

According to this theory, shear thickening is caused by the formation of clusters. Shear thickening starts at a certain critical shear stress at which the hydrodynamic forces between the particles dominate the repulsive forces, causing particles to stick together temporarily. This shear thickening effect is a fully reversible, purely hydrodynamic phenomenon. The critical shear stress increases with decreasing particle size, increasing polydispersity and due to polymer coatings. The intensity of shear thickening increases with an increase in volume fraction (relative to the maximal volume fraction) and is dependent on the kind of repulsive forces. Particle shape does not have a significant influence because the particles remain aligned in the flow direction, but it does influence the intensity of shear thickening due to a decrease in maximal volume fraction.

2.2. Grain inertia

Shear thickening can also be caused by the transfer of momentum between suspended particles [36]. This is caused by grain inertia, where (a part of) the momentum is transferred directly between the particles. In order to estimate whether inertia is dominant, the particle Reynolds number (Re_p) (Eq. (5)), which is the ratio of the inertia to the viscous forces, can be calculated. If Re_p is much smaller than 0.1, inertia is fully dominated by the viscous forces and can be neglected [37–39].

$$Re_p = \frac{\rho_s \cdot (\partial\gamma/\partial t) \cdot a^2}{\eta_s} \tag{5}$$

Where:

Re_p = particle Reynolds number (–)
ρ_s = suspending medium (fluid) density (kg/m^3)
$\partial\gamma/\partial t$ = shear rate (1/s)
a = particle radius (m)
η_s = apparent viscosity of the suspending medium (fluid) (Pa s)

If spheres with a radius of 1 mm are suspended in water ($\rho_s = 1000$ kg/m^3, $\eta_s = 0.001$ Pa s), the particle Reynolds number equals the shear rate. Even at low shear rates in the order of 1 s^{-1}, the particle Reynolds number indicates that inertia is not negligible and that the shearing force will be partly transferred by direct momentum transfer. In case inertia dominates the viscous forces, it has been reported that the apparent viscosity increases linearly with increasing shear rate and consequently, the shear stress scales with the shear rate to a power of 2 [36,40,41]. As a result, the second order term in the shear rate in the modified Bingham model can be an expression of particle momentum transfer, if this theory is applicable on self-compacting concrete.

Recently, authors have proven that for suspensions of particles in a liquid carrier fluid, the perfect second order relationship between shear stress and shear rate has not been obtained experimentally, even at high particle Reynolds numbers [42]. For the results shown in [42], when applying a power law function on the rheological data, the exponent did not exceed the value of 1.58. This means that, even at high particle Reynolds numbers, the viscous forces still have an influence on the rheological behaviour of the material. On the other hand, suspensions of solid particles in a gassy carrier fluid (like air), do show the perfect quadratic relationship between shear stress and shear rate [41], but this is of no practical interest in this paper.

2.3. Applicability on concrete

The two shear thickening theories mentioned in previous sections are physically completely different. The cluster theory scales with the relative importance of hydrodynamic forces to the inter-particle repulsive forces. This theory only applies on small, Brownian particles. In concrete, small, Brownian particles are present, which is proven by the fact that concrete is a thixotropic material. At rest, the small particles coagulate/flocculate, which means that they must have a kind of motion, even at rest, which must be caused by the Brownian forces. Consequently, an attempt to apply the cluster theory on concrete is justified.

The nature of grain inertia shear thickening is the dominance of inertia to the viscous forces in the liquid. As can be seen in Eq. (4) for the particle Reynolds number, inertia gains importance with increasing particle radius. As a result, the larger particles are responsible for shear thickening. As concrete contains large particles, in the order of a few centimeters, this theory is also suitable for application.

The order–disorder transition theory, shortly introduced when discussing the cluster formation, has recently been pointed as the cause of shear thickening in a suspension of monodisperse, non-Brownian particles [43]. Rastogi and Wagner [20,21] on the other hand, state that the layered ordering of particles disappears gradually with increasing polydispersity. As a result, the order–disorder transition theory is not found suitable for application on concrete.

3. Materials and methods

3.1. Materials tested

3.1.1. Concrete

In this research project, more than 50 different SCC mixes have been produced and the main part of the results has been reported in [11]. In Table 1, the composition of the reference mixes can be found. The two superplasticizers used (SP 1 and 2) are both poly-carboxyl-ethers, of which SP 1 is very efficient, but has a short workability retention, while SP 2 is less efficient, but has a longer workability retention. Table 2 shows for all mixes tested and for both SP, the maximal and minimal values of the different constituent elements in the concrete compositions. All concretes have been prepared with CEM I 52.5 N. Four different kinds of fillers have been applied: two limestone fillers (LS 1 and LS 2) from different manufacturers, fly ash and silica fume. The reference filler applied was LS 1.

All concretes have been mixed according to the following procedure: mixing all dry components during 15 s, adding water and mixing 2 min, adding SP and mixing 3 more minutes. After the mixing, the SCC was evaluated visually or by means of a slump flow. If the fluidity was not sufficient, an extra amount of SP was added and the SCC was mixed for 2 additional minutes.

A special test series has been carried out to investigate the influence of demoulding oil in SCC [44]. Six different SCC mixes have been produced, in a similar way to the above mentioned mixing procedure. After taking a slump flow, the batch of SCC has been split in two parts. The rheological properties from the first part, being the reference part, have been determined immediately, while the second part has been put back inside the mixer, a small amount of demoulding oil has been added and the SCC has been mixed for two more minutes. After mixing, the rheological properties of the second part have been determined. A detailed description and all test results of this specific test series can be found in [44].

3.1.2. Mortar and concrete

One special test series has been carried out on mortar and concrete mixes. Three SCC-compositions have been mixed, without adding the coarse aggregates, thus containing only cement, limestone filler, sand, (all) water and (all) SP. The coarse aggregates have been added in three steps: first the 2/8-fraction, afterwards the 8/12-fraction and finally the 12/16-fraction. The rheological properties of each of the four mixes have been determined.

3.1.3. Cement paste

The rheological properties of traditional cement pastes with W/C-ratio of 0.4 and 0.5, only containing cement and water, and of five self-compacting cement pastes, based on the self-compacting concrete compositions, have been determined separately. The mixing procedure of the cement paste was similar to that of the concrete: dry mixing (if necessary) during 15 s and adding water and mixing for 2 min, followed by homogenising the paste by hand during 1 min. Finally, the cement paste was mixed for 3 more minutes, with the addition of SP, when applicable. An attempt has been made to include a self-compacting cement paste without SP in the testing program, but the measurements failed due to excessive slippage inside the rheometer.

Table 1
Composition of reference mixes with SP 1 and SP 2 (units in kg/m^3).

	SP 1	SP 2
Gravel 8/16	434	434
Gravel 2/8	263	263
Sand 0/4	853	853
CEM I 52.5 N	360	360
Limestone filler 1	240	240
Water	165	165
SP (l/m^3)	3	14.55

Table 2
Variation of the different elements in the composition, for each SP.

		SP 1		SP 2	
		Min	Max	Min	Max
C	(kg/m^3)	250	450	300	400
P	(kg/m^3)	400	700	500	700
C/P	(–)	0.417	0.75	0.5	0.67
W	(kg/m^3)	165	192.5	133.3	186.5
W/C	(–)	0.37	0.66	0.4	0.55
W/P	(–)	0.24	0.41	0.23	0.32
SP	(l/m^3)	1.8	4.7	7	18
SP/C	(–)	0.0063	0.0131	0.0192	0.0547

C = cement, P = powder (cement + filler), W = water, SP = superplasticizer.

3.2. Rheometry

3.2.1. Contec viscometer 5

This rheometer has been used to determine the rheological properties of SCC, as verification for the results of the Tattersall Mk-II rheometer, and also for the mortar–concrete tests (Section 3.1.2). This rheometer has an inner cylinder with a radius of 10 cm, and an outer, rotating cylinder of 14.5 cm. The inner cylinder consists of an upper and lower part, of which the latter does not participate in the measurements. In this way, the influence of the complex 3-D flow at the bottom is eliminated [2]. The upper part of the inner cylinder is submerged for 12 cm in concrete or mortar. Both cylinders are equipped with ribs in order to prevent slippage. A detailed description of the rheometer and the transformation of the results into fundamental rheological units, only by means of the Herschel–Bulkley equation, can be found in [11,12,45].

3.2.2. Tattersall Mk-II rheometer

This rheometer, originally developed by Tattersall [1], consists of an outer cylinder with a radius of 12.5 cm provided with ribs, and an inner cylinder equipped with a helicoidal screw. Contrary to the Contec viscometer 5, the inner cylinder rotates and registers torque. A calibration procedure has been developed [14] in order to transform the results into fundamental rheological units. More information on this rheometer can be found in [1], [11] and [14]. This rheometer has been applied for the main part of the SCC tests and the tests with SCC containing oil. The mortar tests could not be executed on this rheometer because the material was too liquid to be measured accurately.

3.2.3. MCR-100 with plate–plate geometry

In order to test the rheological properties of the cement pastes, the MCR-100 rheometer (Anton Paar) has been used. The rheometer was equipped with a so-called plate–plate configuration, of which the upper plate, with a diameter of 5 cm, rotates and the lower plate remains stationary [38]. Both plates are smooth, but no significant slippage, segregation or bleeding has been observed for the tested materials and the applied shear rate range. At higher shear rates, these assumptions can be doubted for the more fluid cement pastes. When lifting the upper plate after the tests which are reported in this paper, the cement paste appeared homogeneous. The gap between the plates was 2 mm, which is sufficiently large to eliminate the influence of individual particles or flocculated groups [38]. On the other hand, during the pre-shearing period, a considerable amount of cement paste has escaped from between the plates. This has decreased the actual torque value, but as the torque reached equilibrium during pre-shearing, it is assumed that from the start of the decreasing shear rate ramp, no more material has escaped from between the plates. As it is not in the intention to compare the values of cement paste and concrete, but it is only the purpose to reveal whether or not shear thickening is present, this escape of material does not affect the conclusions of this paper. Furthermore, tests with a 1 mm separation distance reveal, qualitatively, very similar results for the shear thickening behaviour.

3.3. Testing procedure

As concrete shows time dependent behaviour due to thixotropy and loss of workability, appropriate actions have been taken to eliminate their influence. Thixotropy has been eliminated by stepwise decreasing the rotational velocity after a pre-shearing period at the maximal rotational velocity. If during the pre-shearing period, not all thixotropy was removed from the sample, the data points which were not in equilibrium have been eliminated from the results [11].

Loss of workability has been eliminated by testing each concrete at an age of 15 min. On the other hand, this procedure could not be applied to the tests of SCC with oil and the mortar–concrete tests, where the samples are needed to be modified between the rheological tests. In these cases, the tests have been carried out as soon as possible, leading to a time difference between the first and the last test of 7 min, in case of the tests with oil, and 1 h between the test with the mortar and the test with the SCC containing all aggregates. Only in the latter case, for the mix with SP 1, some loss of workability can be expected to influence the results. On the other hand, when only studying shear thickening behaviour, it has been found that loss of workability does not significantly influence shear thickening during the first 2 h after the water addition [46].

Measures have been taken in order to eliminate further measurement artefacts [47], which are described in detail in [11], where, by measurements with the two concrete rheometers, it is proven that shear thickening is a physical phenomenon in self-compacting concrete, and not a measurement artefact.

4. Application of theory on experimental results

This section contains six different influencing parameters on shear thickening behaviour, for which an attempt will be made to describe these influences by means of the theories mentioned in Section 2.

4.1. Cement pastes

First, the tests on cement pastes are mentioned in order to investigate the source of shear thickening behaviour. Fig. 5 shows the relation between the apparent viscosity and the shear rate for a traditional cement paste with a W/C-ratio of 0.5 and a self-compacting cement paste, based on the reference composition with SP 1. The results have been determined with the MCR-100 rheometer, decreasing the shear rate from 100 s^{-1} linearly to 1 s^{-1}, during 90 s, after a pre-shearing period of 60 s at a shear rate of 100 s^{-1}. The traditional cement paste shows a clear decrease in apparent viscosity over the full range of shear rates measured. In Fig. 6, showing the flow curve, which is the shear stress–shear rate relationship, it can be seen that the TC-paste behaves Bingham-like at high shear rates and appears to be shear thinning at lower shear rates. No shear thickening is observed.

For the self-compacting paste, a different behaviour is found. The apparent viscosity increases from a shear rate of 12 s^{-1} on, clearly indicating shear thickening behaviour. For lower shear rates, shear thinning occurs. Results of other pastes, both traditional and self-compacting, show very similar behaviour. Other results in literature on plasticized cement pastes indicate similar behaviour [48].

When investigating the source of shear thickening, one can calculate the particle Reynolds number in order to investigate the influence of grain inertia. The values of density and (apparent) viscosity are chosen to be those of the cement paste, because removing 2 particles does not influence the overall density and viscosity. Density has not been measured, but it has been estimated based on the composition. A value of 2200 kg/m^3 is applied, which is an overestimation on the safe side. The (apparent) viscosity has been taken as the minimal viscosity from Fig. 5: 0.5 Pa s. This is an underestimation for the full range of shear rates, but again, it is a safe assumption. The shear rate is the maximal shear rate applied: 100 s^{-1}, and the grain diameter is chosen to be 1 µm and 300 µm. 1 µm corresponds to a small particle, while 300 µm is one of the largest particles in the paste. The particle Reynolds numbers for these two particle diameters is 10^{-7} and 10^{-2} respectively. In each case, the particle Reynolds number is at least one order of magnitude lower than 0.1, indicating that grain inertia is not significant. On the other hand, shear thickening is observed and as a result, grain inertia is not

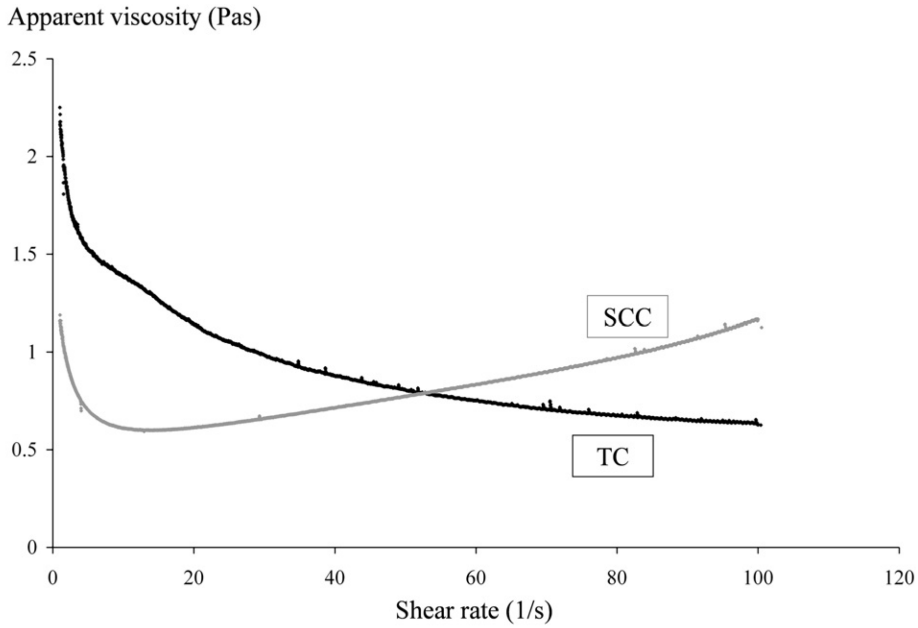

Fig. 5. Apparent viscosity as a function of shear rate showing no shear thickening in case of traditional cement paste (black), and shear thickening from 12 s^{-1} for the self-compacting cement paste (grey).

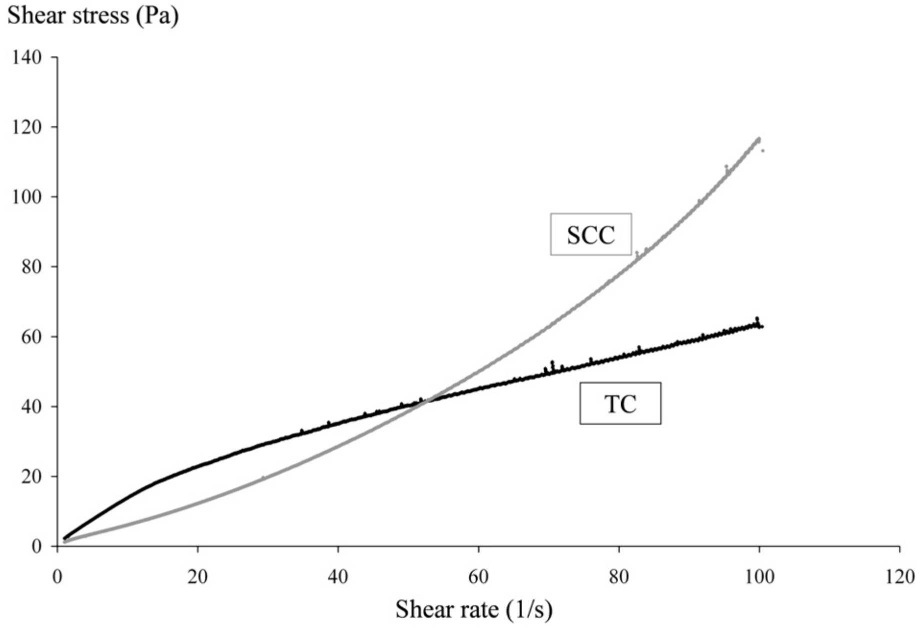

Fig. 6. Flow curve (shear stress as a function of shear rate) for traditional cement paste (black), showing Bingham behaviour at large shear rates and shear thinning at small shear rates. The self-compacting cement paste (grey) is shear thickening.

causing the shear thickening behaviour in this case, although it cannot be fully excluded, for example when applying higher shear rates.

4.2. Water to powder ratio

As mentioned in [11], the water/powder ratio plays a significant role in the shear thickening behaviour, especially when this ratio is becoming low. When eliminating the influence of slump flow (Section 4.3), it can be clearly seen in Fig. 7 that the intensity of shear thickening (c/μ) increases with decreasing W/P-ratio. This is in accordance with the cluster formation theory, saying that increasing the solid volume fraction causes an increase in shear thickening behaviour. On the other hand, the volume fraction, which is directly controlled by the water content, should not influence the onset of shear thickening. This is shown in Fig. 8, where the critical shear stress is shown as a function of W/P-ratio. The influence of W/P-ratio on the critical shear stress is not significant, especially compared with Fig. 10 in the following section.

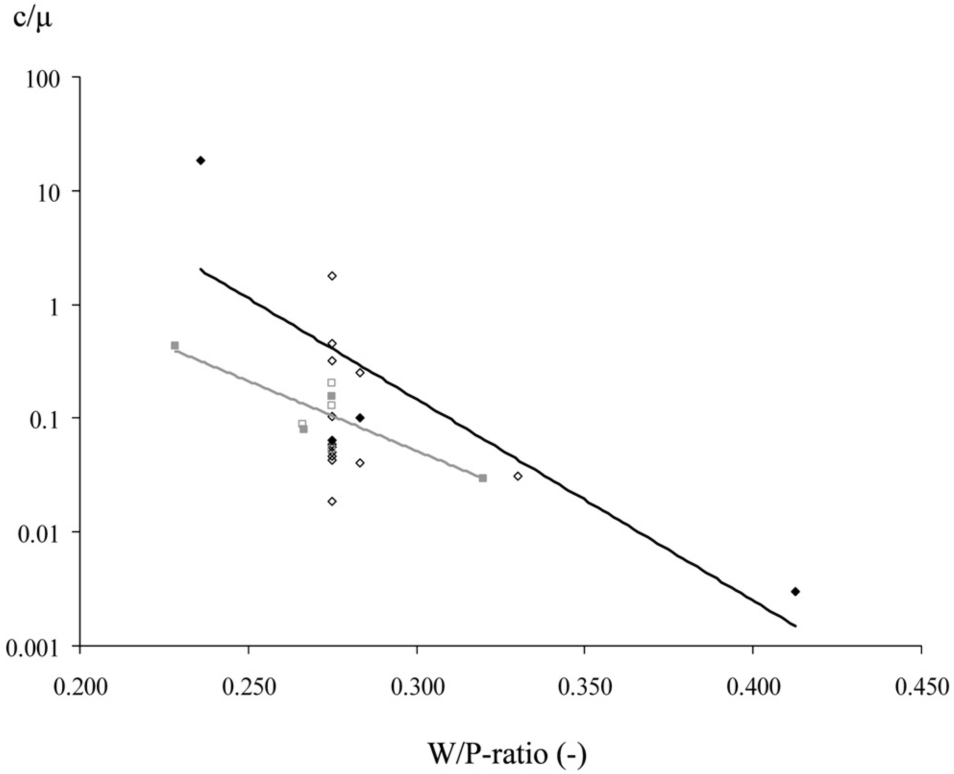

Fig. 7. Intensity of shear thickening (c/μ, from the modified Bingham model), as a function of the W/P-ratio. Data points with equal slump flow (full data points) have been connected showing an increase in c/μ with decreasing W/P-ratio. The black and grey data correspond to SP 1 and 2 respectively.

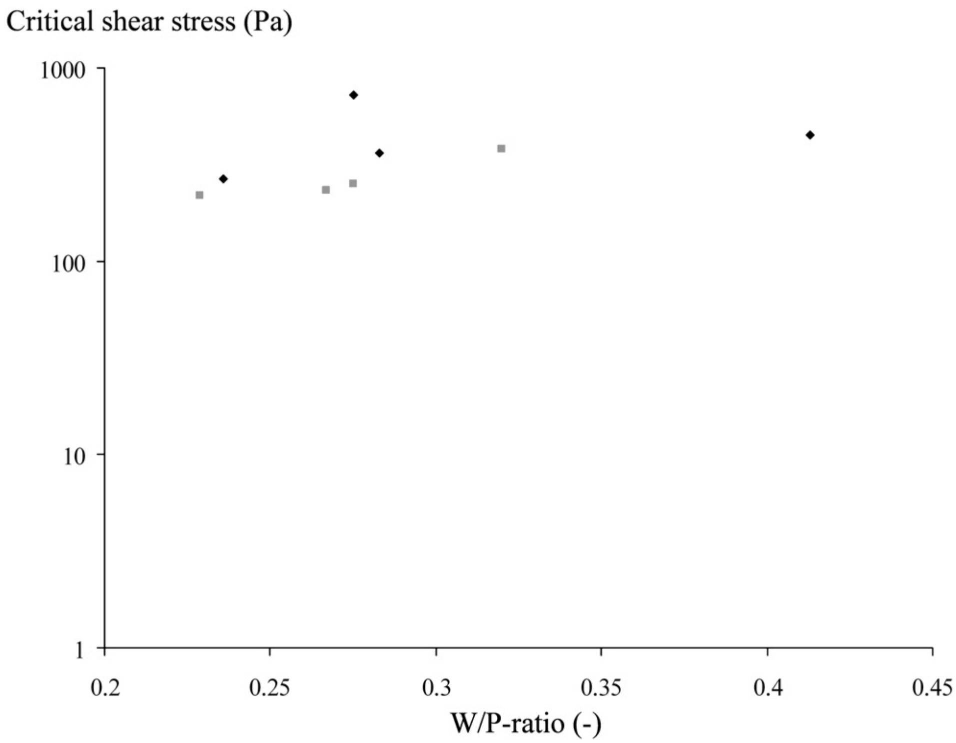

Fig. 8. The critical shear stress for SCC with equal slump flow does not show a significant dependency on the W/P-ratio (volume fraction). The black and grey data correspond to SP 1 and 2 respectively.

4.3. Slump flow and SP-content

In [11], it has been shown that for all mixes with W/P-ratio of 0.275, no clear relationship between the intensity of shear thickening and the superplasticizer content has been found. On the other hand, a relationship between c/μ and the slump flow has been obtained (Fig. 9), showing an increased intensity in shear thickening with increasing slump flow. The relationship is dependent on the type of superplasticizer. Fig. 10 shows the relationship between slump flow and the critical shear stress, indicating that increasing slump flow causes a lower critical shear stress. Fig. 11 shows the plot of apparent viscosity to shear stress for four different mixes with LS 2. The slump flow and SP 2-content of each mix is also shown. It can be seen in this graph that with increasing slump flow/SP-content, the critical shear stress decreases and the intensity, being the inclination of the

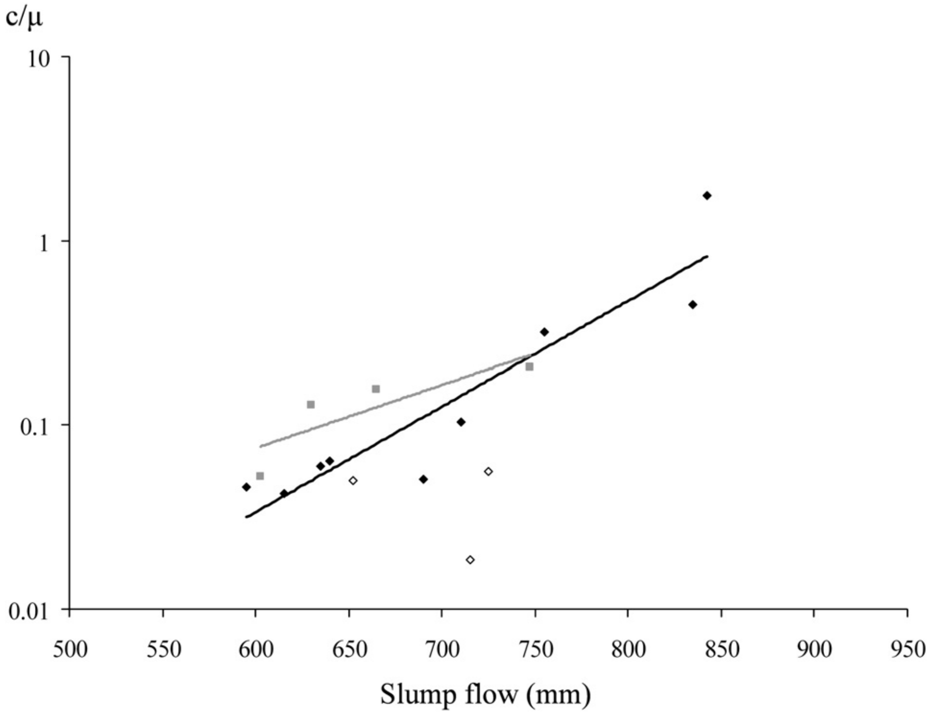

Fig. 9. Intensity of shear thickening (c/μ, from the modified Bingham model) increases with increasing slump flow at a constant W/P-ratio. The black and grey data correspond to SP 1 and SP 2 respectively, and the hollow diamonds represent SCC where all aggregates 2/8 have been replaced by aggregates 8/16.

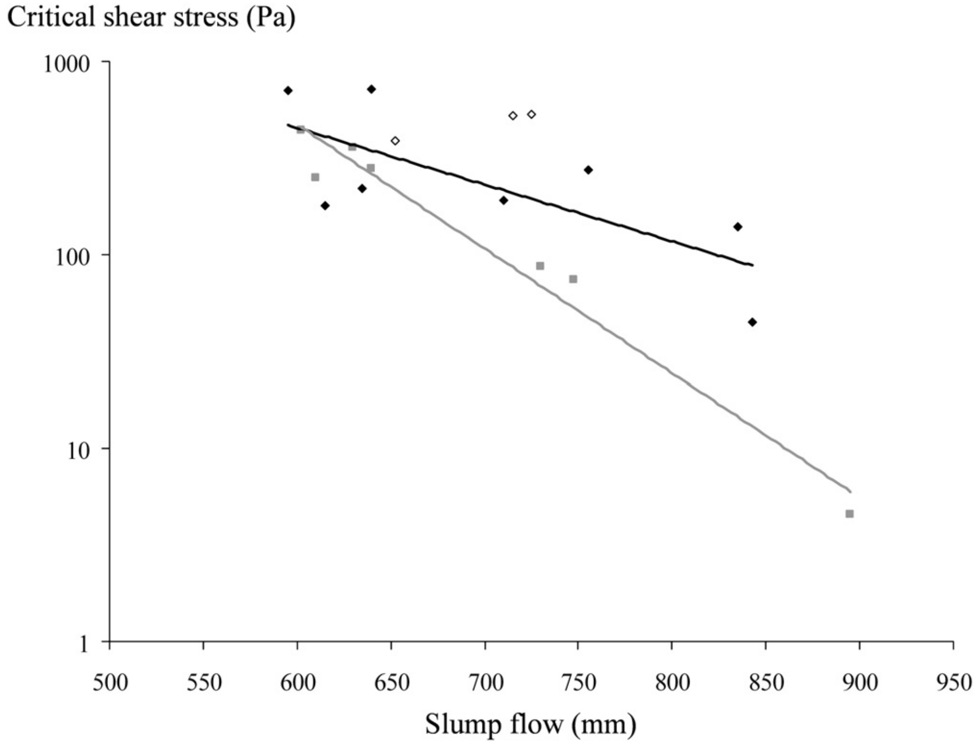

Fig. 10. The critical shear stress decreases with increasing slump flow (at constant W/P-ratio). The black and grey data correspond to SP 1 and SP 2 respectively, and the hollow diamonds represent SCC where all aggregates 2/8 have been replaced by aggregates 8/16.

curve beyond the critical shear stress, increases. Note that the critical shear stress for the SCC with SF = 300 mm, is not determined and can even be higher than the highest shear stress measured.

From these results, as a larger slump flow is obtained due to a higher SP-dosage, it is clear that the critical shear stress decreases and the intensity of shear thickening increases with increasing SP-content. This is apparently in contradiction to the cluster theory [35] and in contradiction to [48], where an increase in critical shear stress has been observed with increasing SP-content. This discrepancy can be due to the amount of particles available for shear thickening. In [48] a sufficient amount of small particles were available in the fly-ash cement paste, where this phenomenon has been observed, while for the LS-mix in [48], no shear

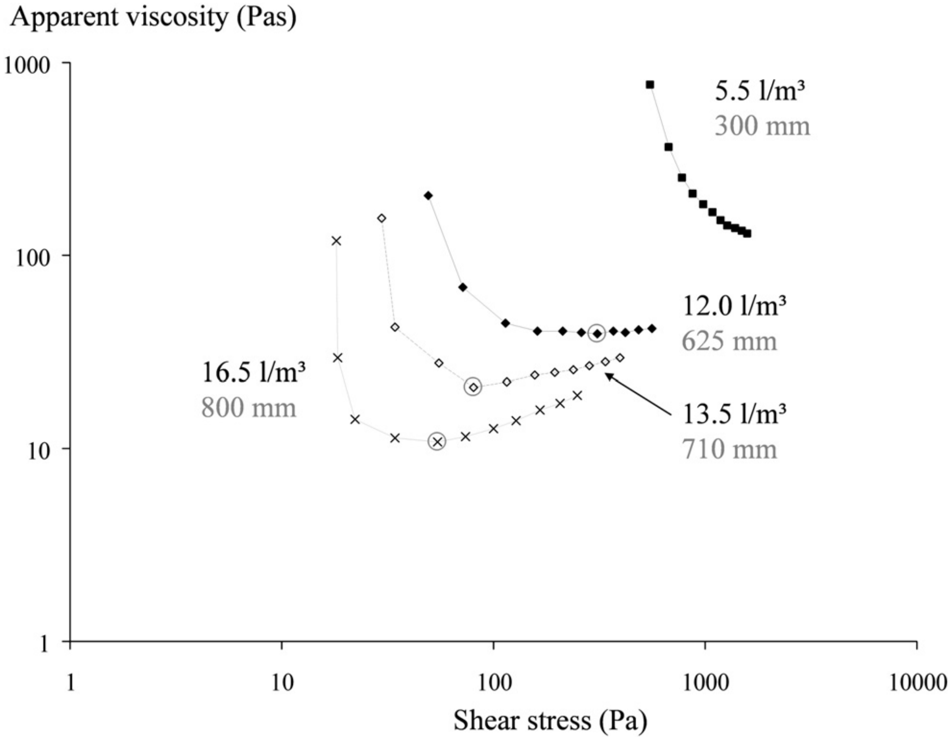

Fig. 11. Apparent viscosity as a function of shear stress for 4 SCC mixes with LS 2. The numbers represent the amount of added SP (black) and the slump flow (grey). The critical shear stress for each mix has been marked with a grey circle. Increasing SP-dosage leads to an increase in intensity of shear thickening and a decrease in critical shear stress.

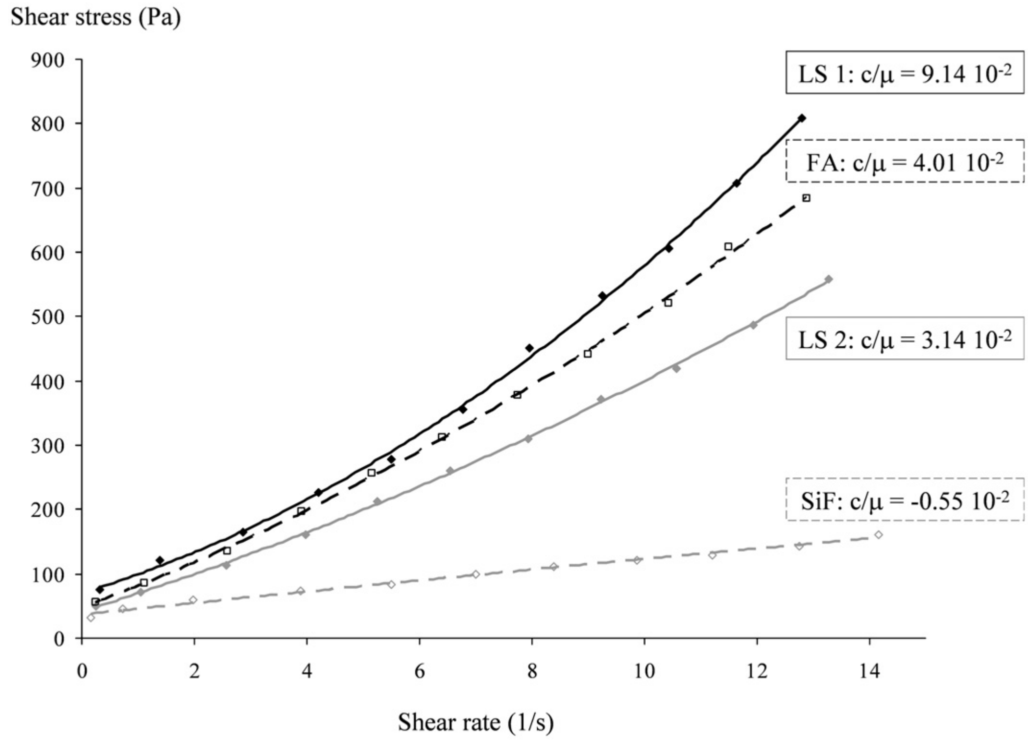

Fig. 12. Flow curves for 4 SCC mixes, having an equal volume fraction, but different kinds of filler. The mix with silica fume does not show shear thickening in the stress range measured.

thickening occurred. The results reported in [11] and in Figs. 5–11 in this paper, are purely based on LS-filler SCC mixes. Due to an increase in SP-content, more (cement) particles are dispersed, increasing the amount of small particles, susceptible for shear thickening. This can explain why in these test results, an increase in intensity of shear thickening and a decrease in critical shear stress is observed. Lowering the amount of SP leads to larger amounts of flocculated (or coagulated) particles, which do not show shear thickening behaviour. As a result, as traditional cement and concrete are not dispersed as well as SCC due to no or low SP-contents, these materials do not shear thicken in the shear stress ranges measured.

4.4. Fillers

Fig. 12 shows the influence of different fillers on the intensity of shear thickening. It is clear that the SCC with silica fume does not show any shear thickening at all, in contrast to the SCC with LS 1, LS 2

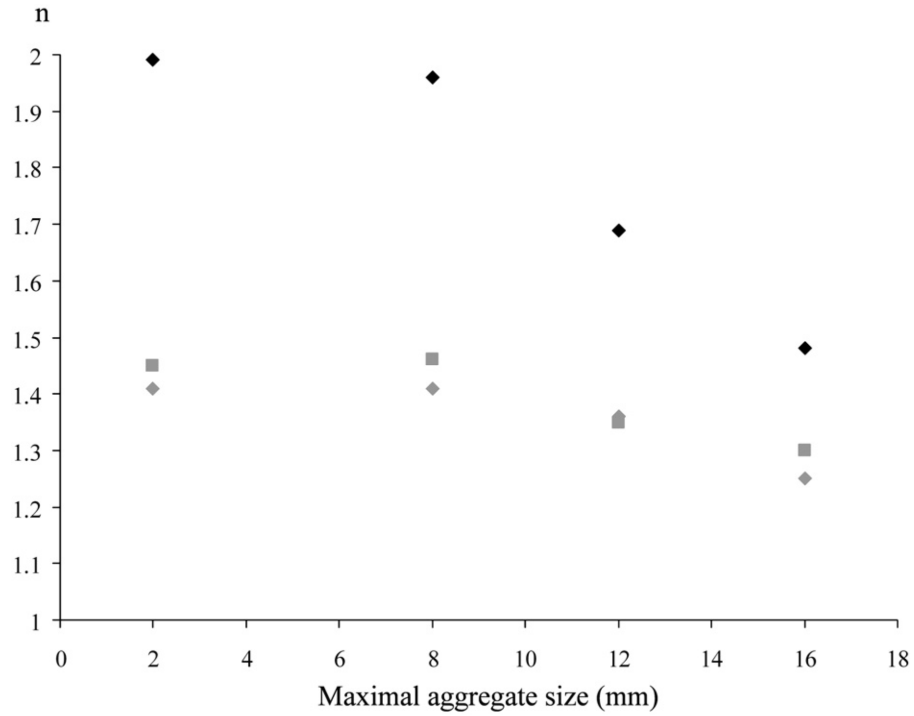

Fig. 13. Intensity of shear thickening (“n” from Herschel–Bulkley) as a function of the maximal aggregate size added for two mixes with SP 2 (SCC 1, 2 – grey) and a severely shear thickening mix with SP 1 (SCC 3 – black). From a maximal aggregate size of 8 mm on, the intensity of shear thickening starts to decrease.

Table 3
Particle Reynolds numbers, indicating the importance of inertia, for different aggregate sizes.

Maximal aggregate size	Re_p
(mm)	(–)
2	5.0 10^{-3}
8	7.7 10^{-2}
12	1.7 10^{-1}
16	3.0 10^{-1}

and fly ash. This is most probably due to the smaller grain size of the silica fume particles. As mentioned in the theory, the critical shear stress increases with decreasing particle size, for small particles. In addition, the critical shear stress for the three other mixes in Fig. 12 is around 300 Pa, while the stress at maximal shear rate for the silica fume SCC is only 160 Pa. As a result, the SCC with silica fume does not show any shear thickening due to the too low stresses obtained in the rheometer in order to see shear thickening behaviour. By means of these measurements, it is not certain whether or when this SCC will show shear thickening behaviour.

The other mixes in Fig. 12 all show shear thickening behaviour. They all have an equal volume fraction and an equal slump flow. Still, differences are noticeable, probably due to the different grain size distributions. LS 2, for example, contains somewhat more large particles than LS 1, resulting in a lower specific surface, increasing the free-water/powder ratio, decreasing effective volume fraction, and also decreasing the amount of particles available for shear thickening.

4.5. Coarse aggregates

In this paragraph, the influence of the amount and size of coarse aggregates will be discussed. Fig. 13 shows the intensity of shear thickening, expressed by the value "n" of Herschel–Bulkley, as a function of the maximal aggregate size for three mixes. SCC 1 and 2 are the reference mixes, with different amounts of SP 2, while SCC 3 is a severely shear thickening mix, with low W/P-ratio and made with SP 1. From Fig. 13, it can be seen that increasing the maximal aggregate size from 8 to 12 and further to 16 mm decreases the intensity of shear thickening. This is apparently in contrast to the expected behaviour. Adding more aggregates increases the volume fraction and increases local shear rates between the aggregates, which should result in a more intense shear thickening behaviour.

On the other hand, if shear thickening is caused by the formation of clusters, inertia forces acting on the larger particles can possibly break the clusters. In Table 3, the particle Reynolds numbers are shown, acting on the largest particles in each mix. All particle Reynolds numbers have been calculated, assuming a density of 2400 kg/m^3, a shear rate of 20^{-1}, and an apparent viscosity equal to 10 Pa s, which is the viscosity of the mortar. For the mixes with larger maximal aggregate size, the viscosity of the suspending medium in the calculations is not increased due to the addition of large aggregates, because the probability of finding another coarse aggregate very close to a certain one is quite low, due to the principle of making SCC [9]. As a result, the mortar viscosity has been applied in each calculation.

As can be seen in Table 3, the inertia forces gain importance with increasing aggregate size, and they become non-negligible from an aggregate size of 8 mm on. It is from this size on that shear thickening starts to decrease with increasing maximal aggregate size. It is suspected that due to inertia forces, the coarsest aggregates partly break down the clusters formed in the nearby cement paste. Due to the large separation distance between two coarse aggregates [9], inertia is not suspected to cause additional shear thickening behaviour.

When replacing all aggregates with size 2/8 by aggregates 8/16 only, creating a total aggregate 8/16 content of 697 kg/m^3, it is suspected from the above mentioned theory that the intensity of shear thickening decreases when this replacement has occurred. In Fig. 9, expressing intensity of shear thickening versus slump flow, three hollow black dots can be seen, representing the mixes with 697 kg/m^3 aggregate 8/16

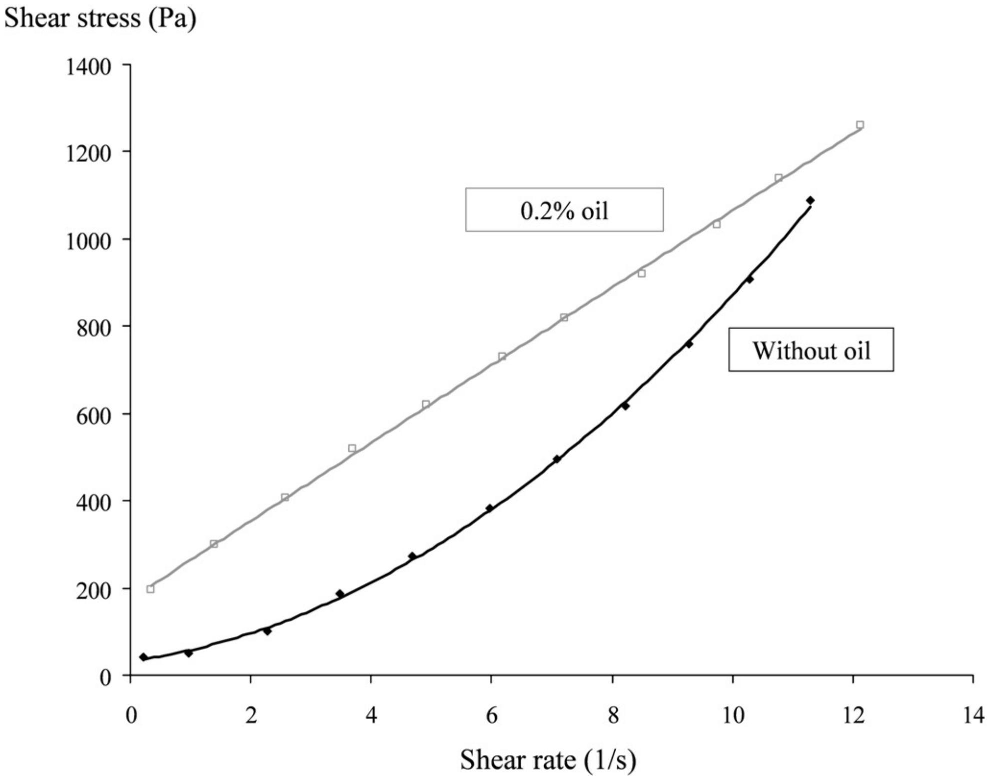

Fig. 14. Adding demoulding oil to SCC causes shear thickening to diminish and even disappear.

only. These mixes show systematically lower shear thickening than the mixes with 2/8 and 8/16, confirming the above mentioned theory.

4.6. Demoulding oil

Adding a small amount of demoulding oil to self-compacting concrete causes shear thickening apparently to decrease and even disappear, at least in the range of shear stresses measured [44]. Fig. 14 shows a severely shear thickening mix, without and with 0.25% vol. of demoulding oil. It can be seen that the mix with oil does not show shear thickening in the stress interval measured. Other results, not shown here, confirm this observation. We cannot provide a fundamental explanation for this phenomenon, but it is possible that the oil decreases the hydrodynamic forces in the suspending medium, or increases the inter-particle repulsion forces, or combines both effects,…

5. Discussion

5.1. Why is shear thickening often not reported?

In order to observe any shear thickening behaviour, small particles must be present in sufficient amounts, and the amount of coarse particles must be relatively low. For example, Danish SCC [5,49], containing low powder amounts and higher amounts of coarse aggregates is more unlikely to show shear thickening than a SCC with high powder content. Also, in countries where silica fume is frequently applied – it is not in Belgium due to the high cost – shear thickening behaviour will not be seen very soon, as has been observed in this paper.

On the other hand, the measurements in the rheometers have been performed at relatively high shear rates, as an attempt to approach pumping conditions [50,51]. In literature, rheological measurements on concrete are performed at lower shear rates and consequently, at lower shear stresses [5,52]. As a result, for a SCC mix susceptible for shear thickening, the critical shear stress is not exceeded and shear thickening is not observed. Or it can be that the critical shear stress is only exceeded for the measurement points at the highest shear rates, which do not confirm the traditionally expected linear (Bingham) relationship, and mostly these data are eliminated by the argument of non-equilibrium. Detailed investigation of the time-plot of the shear stress (only at constant shear rate) will indicate whether the deviation from the linear relationship is due to thixotropy or shear thickening.

5.2. Why is shear thickening important?

As mentioned in the Introduction, some processes in concrete industry occur at (much) higher shear rates than those in rheometer tests. Mixing and pumping are the two best known examples. As the shear rate increases, the resulting shear stress is gradually more dominated by the plastic viscosity and shear thickening behaviour. For pumping processes, for example, the authors have found that, at discharges of around 20 l/s (which is only the half of the capacity of the used pump) pressure losses are dominated by shear thickening and plastic viscosity, not by the yield stress [50,51]. Increasing the discharge even more should finally cause shear thickening to be much more important than the plastic viscosity only. Not incorporating shear thickening in any calculations or predictions will underestimate the pumping pressures significantly at high discharges, endangering the safety of the working staff on the worksite and leading to large economical and ecological losses.

Also for mixing processes, which are assumed to occur at even higher shear rates than pumping, shear thickening is very important. Insufficient control over this parameter can lead to destruction of the mixing equipment, which the authors have experienced personally.

On the other hand, it is mentioned in the first sentence of this paper: “Concrete is described by rheologists as one of the most difficult materials to study.” Due to particle migration, segregation,… very high shear rates cannot be imposed onto the material [47]. Due to the high aggregate dimensions, large geometries must be built, making concrete rheometry extremely difficult. Imposing very high shear rates in order to study shear thickening is consequently not without any risks, and careful observations during the measurements should be executed in order to investigate measurement artefacts. Therefore, obtaining shear rates as in pumping or mixing processes [53] is very unlikely to occur, but applying a reasonably high shear rate, when still no artefacts are present should be a good tool to give some indication of the expected behaviour. Still, one must keep in mind to apply a shear stress or shear rate in the rheometer which corresponds to the expected application of the concrete, mortar or cement paste [5].

5.3. Relationship between thixotropy and shear thickening

Thixotropy and shear thickening act, according to the theory of cluster formation, on the same particles, namely the small particles in concrete, which are influenced by inter-particle forces. As a result, one could expect that both effects are well related to each other. In fact, they are not so well correlated. There is a fundamental difference between shear thickening and thixotropy. Shear thickening is a reversible time-independent phenomenon, but it depends on the applied shear stress (and shear rate). Thixotropy is fully time dependent, being regulated by build-up during rest and breakdown during shear [2,3]. The higher the shear rate, the faster the breakdown.

Does thixotropy influence shear thickening? It would be wrong to say no. As flocculated systems do not shear thicken, more dispersion causes more shear thickening, at least for the particle ranges in the order of 1 to 10 μm (the opposite might be true for the results shown in [48], where the average particle size is smaller). Higher dispersion can be obtained by using larger amounts superplasticizer, but also by applying a higher pre-shear rate [2,3,54–55]. Consequently, the structural state [3] will partly influence shear thickening behaviour.

Does shear thickening influence thixotropy? Not really. During build-up at rest, shear thickening is not present. During breakdown, shear thickening can be observed, but in a shear rate controlled device, which is mostly applied in concrete rheometry, shear thickening only influences the applied stress, and thixotropic breakdown scales only with the shear rate [3,54]. As a result, shear thickening does not really influence thixotropy.

5.4. Physical meaning of different parameters in rheological models

As it has been proven that the grain inertia theory in its present state is not applicable to the results obtained for these self-compacting concretes, the proposed physical meaning for the second order term in the modified Bingham model is no longer valid. Consequently, as we did not find another physical meaning for this second order term, the modified Bingham model is only a Taylor-series development of the Herschel–Bulkley model of the order two. On the other hand, no measurements have shown an “n” value of Herschel–Bulkley larger than 2, but theoretically, it should be possible, causing in that stage the modified Bingham model to be invalid. Luckily, in practice, no extreme shear thickening SCC are being produced and the advantage of the modified Bingham model in describing pipe flow is still valid [16].

6. Conclusions

Two possible theories to explain shear thickening behaviour of self-compacting have been described. Clusters can be formed when the hydrodynamic forces between Brownian particles are larger than the repulsive forces. At a shear stress larger than the critical shear stress, apparent viscosity increases with increasing shear stress. When grain inertia dominates, shear stress scales with the shear rate squared, due to direct momentum transfer. Grain inertia seems to be negligible compared to the viscous forces when the particle Reynolds number is much smaller than 0.1.

Measurements on cement pastes show that self-compacting cement paste shows shear thickening. Calculation of the particle Reynolds number for the largest grains shows that grain inertia does not have a significant influence. Shear thickening is consequently most probably caused by cluster formations, and probably not by grain inertia in this case.

Decreasing the W/P-ratio, resulting in an increase in volume fraction, increases the intensity of shear thickening, but does not affect significantly the critical shear stress.

Increasing slump flow or superplasticizer dosage causes an increase in intensity of shear thickening and a decrease in critical shear stress for the concretes tested. This is most probably caused by the higher dispersion caused by the SP, resulting in a larger amount of particles available for shear thickening.

SCC with silica fume does not shear thicken in the range of stresses measured, probably due to the addition of smaller particles, causing an increase in the critical shear stress. The differences between the SCC with other fillers are probably due to a difference in grain size distribution and free-water/powder ratio.

Coarse aggregates larger than 8 mm have non-negligible inertia forces, which can break down the clusters formed nearby, causing the shear thickening to decrease with increasing maximal aggregate size. An increase in shear thickening due to grain inertia is unlikely to occur due to the large separation distance between the coarse aggregates. Adding demoulding oil in a shear thickening SCC makes (almost) all shear thickening to disappear. The actions behind this phenomenon are still unclear, but probably, the oil influences the hydrodynamic forces or the repulsive forces, or both.

Shear thickening can only be observed if the material is susceptible for shear thickening, meaning that a sufficient amount of small particles must be present, together with a low amount of coarse aggregates. In addition, the shear stress applied must be sufficiently high to exceed the critical shear stress. On the other hand, it should be kept in mind that the shear stress or shear rate cannot be increased infinitely in concrete rheometry due to the problems of particle migration and segregation.

Due to the validity of the cluster formation theory, no physical meaning can be given to the second order term in the modified Bingham model, making this model, theoretically, invalid in cases of very intense shear thickening ($n>2$ in Herschel–Bulkley). On the other hand, the modified Bingham model still keeps its advantages in describing pipe flow.

Thixotropy influences shear thickening partly through the structural state. Shear thickening does not really influence thixotropy. Both phenomena have a different physical background.

Shear thickening in concrete is a very important phenomenon during mixing and pumping and it should be understood and controlled very well. If not, several devices can be broken and dangerous situations can occur on worksites.

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