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Investigation on the influence of granular packing on the flow properties of cementitious suspensions

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ABSTRACT

Fresh concrete can be considered as a suspension of grains of various sizes in a continuous fluid phase. The rheological properties of fresh concrete greatly depend on the physical factors, chemical and mineralogical characteristics of the fine components. Interparticle interactions occur during flow and modify the apparent rheological behaviour. Therefore, a thorough understanding of the rheological behaviour of cementitious suspensions with respect to particle packing is required. The objective of this study is to characterise the interrelationship between the flow properties and the particle packing density of the cementitious suspensions. The experimental investigation included Puntke tests for determining the packing density, and rheological tests that were performed in a rheometer for the characterisation of cement and silica fume (C + SF) as well as cement and fly ash (C + FA) mixtures. The effect of the water to powder ratio (*w*/*p* ratio) and the packing density on the flow properties of the cementitious suspensions was studied. From the study, it was observed that a good correlation exists between the *w*/*p* ratio and the yield value (*g*) for both C + SF and C + FA mixtures. The packing density shows a marked influence on the value of g for both mixtures, but has less influence on the value of plastic viscosity (*h*) for C + FA mixture.

1. Introduction

A number of studies have been performed on measuring rheological properties in the fresh state of cement paste and mortar; these range from conventional measurements – flow test and slump test – to more quantitative fundamental measurements using rheometers [1]. Rheology is the science of flow and deformation of matter and therefore is the appropriate tool to describe the workability and mobility of fresh cement-based materials like cement paste, mortar or concrete [2]. Established rheological methods permit the determination of characteristic scientific parameters in contrast to the workability numbers obtained by empirical tests. Using rheological methods, the elastic and viscous properties can be detected separately [3]. The quality of concrete structures not only depends on the ingredients of the concrete, but also on the rheological behaviour of the fresh concrete during placement into the formwork. With good rheological properties, a concrete should fill the formwork without excessive effort [4]. The rheological behaviour of the concrete basically depends on a complex interplay of a number of factors including those related to the particle – liquid interaction (water to cement ratio, particle shape and size distribution of the suspended phase), cement chemistry and the test conditions [5].

The rheology of cement-based materials can be described by the Bingham model which involves two fundamental parameters, the yield stress and the plastic viscosity. The rheological properties of cement pastes have been the subject of study over the years due to the influence of their characterisation on concrete technology. Moreover, formulation of models which satisfactorily describe the general behaviour of concrete has proved difficult [6–8]. As a result, most research efforts reported in the literature [9] are focussed on studying the rheology of pastes.

The concept of particle packing has always been a key element in the mixture proportioning of concrete. Particle packing involves the selection of appropriate sizes and proportions of particulate materials to get suitable combination for optimal packing. Experiments show that the packing density of concrete mixtures and the flow properties of the corresponding fresh concrete are related. The optimal flow properties are obtained for mixture compositions close to maximum packing density. In this way, the packing density has an important effect on fresh concrete properties [10]. In 1907, Fuller and Thompson [11] investigated the importance of the size distribution of the aggregates and the properties of the concrete on the basis of packing of constituent materials. Most of the literature published on this subject described the optimisation of packing for filtration of slurries containing fine solids and

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Table 1
Physical characteristics of the cementitious materials

Material	Colour	Type	Density (g/cm^3)	Mean particle size d_{50} (μm)
Cement	Pale grey	Cem I 42.5 R, HeidelbergCement AG, plant Weisenburg	3.18	15
Fly ash (class F)	Dark grey	Safament fly ash, plant Voelklingen	2.26	10
Silica fume	White	BASF Elkem Grade 983	2.36	0.15

sintering of pressed solids. A comprehensive overview of various particle packing theories can be found in [12]. Research has also shown that good flow properties and strength characteristics are obtained for mixtures designed with the particle packing approach. No standard method exists for the determination of the packing of aggregates. The densest packing is achieved based on experience. This is not by vibration, but by a combined shaking and tapping process [13].

While particle packing has a significant influence on the properties of concrete, which contains different sizes of particulate inclusions, the paste properties are also affected by the interaction between the cementitious particles. Moreover, as the paste rheology has a major influence on the concrete rheology, it is important to understand the effect of packing of cementitious particles in paste.

The present study focuses on characterising the interrelationship between the particle packing density and the rheological properties of cementitious suspensions without superplasticisers.

2. Characteristics of the cementitious materials

The physical characteristics of cement, fly ash and silica fume used in the present study are provided in Table 1. No superplasticisers were used in the present investigation.

3. Experimental methodology for the determination of particle packing

3.1. Introduction

As stated earlier, there is no standard automated method for measuring the packing density of fine materials. The determination of the packing density of a granular material is afflicted by the influence of van der Waals and electrochemical surface forces. For this reason, with decreasing medium particle size (d_{50}), increased attention has to be given to the compaction method and the duration of compaction. In the present experimental programme, the Puntke test [14] was selected for determination of particle packing. The basic principle of the test is that the water which is added to the dry materials fills the voids in between the particles and acts as a lubricant to make the materials compact efficiently. The water, which is in excess after completely filling the voids, appears at the surface of the mix, indicating the saturation limit.

3.2. Puntke test – procedure

The mass equivalent to 20 cm^3 of the cementitious materials was placed in a beaker. The cementitious materials were mixed thoroughly for homogenisation before water was added. De-ionized water was added gradually, working the mixture with a stirrer until it acquired a closed structure after repeated tapping of the beaker. In the next step, water was added drop by drop with a pipette, mixing carefully, until the saturation point was reached. At this point, the surface smoothes itself after repeated tapping of the beaker and appears glossy. The total time taken for each experiment was approximately 10 min. The experiment was repeated 3 times to get the least water required to achieve saturation. From the volume of water used, the packing density Φ_p was determined by using:

$$\Phi_p = 1 - \frac{V_w}{V_p + V_w} \tag{1}$$

where V_w = volume of water (cm^3); V_p = volume of solid particles = 20 cm^3.

4. Investigated material compositions

Partial replacement was done on a volume basis instead of mass basis due to different particle densities of cement, silica fume and fly ash. The packing density was determined for different cement + silica fume and cement + fly ash combinations by volume, using the Puntke test. The packing densities of 100% cement, 100% silica fume and 100% fly ash by volume were found to be 53.50 vol.%, 52.08 vol.% and 68.14 vol.%, respectively. Theoretically speaking, the increase in percentage replacement of silica fume in cement will increase the packing density. This is due to the high fineness of silica fume which fills the voids in between the cement

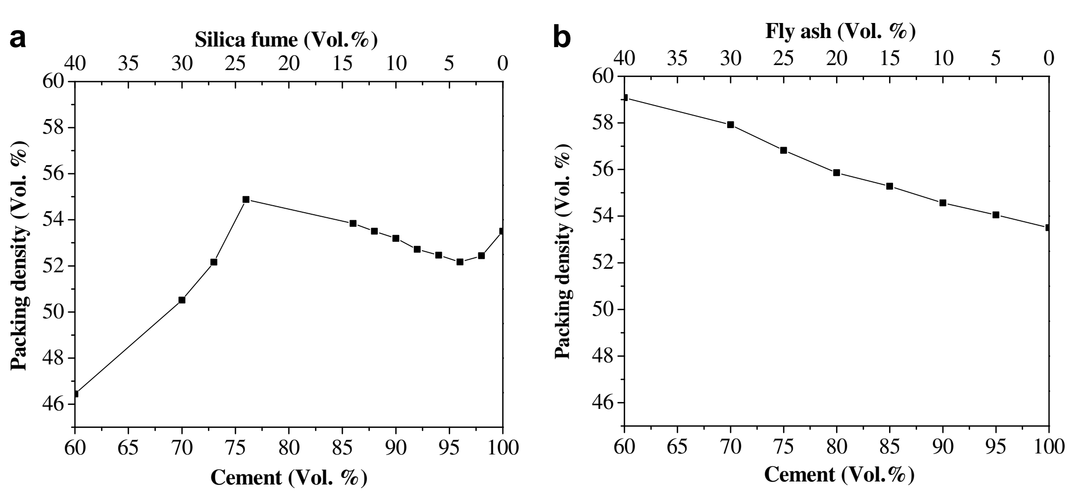

Fig. 1. Relationship between packing density and binder composition for (a) C + SF and (b) C + FA.

particles. But in this case (see Fig. 1), initially the packing density decreases by 1.33% at 4% replacement of silica fume. This negligible decrease in packing density is due to the agglomeration of silica fume due to its high fineness which obstructs the water in filling the voids when mixed. This is evident from the Fig. 1. In case of cement and fly ash mixtures, the packing density increases with increase in percentage replacement of fly ash, owing to the spherical nature and the smaller size of fly ash particles, which results in better packing (see Fig. 1).

4.1. Investigations on cement and silica fume mixtures

The percentage of silica fume was varied between 0% and 40% by volume of cement and the particle packing density was determined (see Fig. 1a). No uniform tendency with regard to the packing density was observed; however, three different compositions (C100, C88S12 and C74S26) have the same packing density of 53.50%. An attempt was made to evaluate the rheological behaviour of these three different compositions. For each composition, the water powder ratio (by volume) was varied at 1.86, 1.50, 1.22 and 1.00 (these correspond to a solid phase volume of 35%, 40%, 45% and 50%).

4.2. Investigations on cement and fly ash mixtures

The fly ash percentage was varied between 0% and 40% by volume of cement and the particle packing density was determined (see Fig. 1b). According to the results of the Puntke tests, the packing density increased with increase in percentage replacement of fly ash. As per German Standard (DIN 1045-2 [15] and EN 206-1 [16]), the maximum percentage of fly ash to be used is 30% by mass equivalent (41% by volume). Therefore, three compositions between 0 to 30% by volume were considered for further investigations. For each composition, the water to powder ratio (by volume) was varied at 1.86, 1.50, 1.22 and 1.00. The investigated compositions of pastes are shown in Table 2.

Table 2
Water powder ratio and composition of pastes

Designation	w/p Ratio (vol.)	Cement (vol.%)	Silica fume (vol.%)	Fly ash (vol.%)	Packing density (vol.%)
C100	1.86 1.50 1.22 1.00	100	–	–	53.50
C96S04	1.86 1.50 1.22 1.00	96	04	–	52.16
C88S12	1.86 1.50 1.22 1.00	88	12	–	53.50
C74S26	1.86 1.50 1.22 1.00	74	26	–	53.50
C90F10	1.86 1.50 1.22 1.00	90	–	10	54.57
C80F20	1.86 1.50 1.22 1.00	80	–	20	55.86
C70F30	1.86 1.50 1.22 1.00	70	–	30	57.92

Nomenclature: C88S12 = cement 88% by volume and Silica fume 12% by volume; C80F20 = cement 80% by volume and fly ash 20% by volume.

5. Studies on flow properties

5.1. Introduction

The rheological characterisation of the selected pastes was performed using a HAAKE Rheostress 600 rheometer in combination with the software Rheowin 3.23 [17]. Based on the required rheological parameters, a job (shear profile) was set up. The rheometer was operated in two modes, i.e. controlled stress (CS) mode (a defined shear stress is applied and the resulting shear rate is measured) and in controlled rate (CR) mode (a defined shear rate is set and shear stress is measured). The measurements in CS mode reflect real life conditions e.g. the behaviour of fresh paste under the influence of gravity. The measurements of torque, speed and time were recorded. The measured torque was not converted to shear stress, as there was no well-defined shear plane (this is clear from the schematic arrangement of the rheometer paddle shown in Fig. 2).

The flow curve, plotted in the form of torque T against speed N, conforms to the equation:

$$T = g + hN \tag{2}$$

where g (N mm) and h (N mm s) are characteristic constants of the paste.

The Bingham equation is written as

$$\tau = \tau_0 + \mu\dot{\gamma} \tag{3}$$

where τ_0 = yield stress (Pa), μ = plastic viscosity (Pa s) and $\dot{\gamma}$ = shear rate (1/s).

Comparing Eqs. (2) and (3), it is apparent that g is related in principle to the yield stress and h to the plastic viscosity. For most practical purposes it is sufficient to use the values of g and h. It should be noted that the rate of change of rheology with time in chemically reacting cement is affected by the temperature [18]. Therefore, all the tests reported here were done at 20 ± 1 °C. Additionally, in order to get comparable results, the materials were stored at constant temperature of 20 °C. Furthermore, a constant mixing procedure and constant time was maintained between adding water to the starting of the rheological measurements.

5.2. Experimental procedure

5.2.1. Cementitious suspension preparation

Rheological tests were performed for the selected compositions (see Table 2). The cementitious suspensions were prepared for a volume of 1.2 l. A variable speed mixer was used for mixing. The mixing procedure is shown in Table 3.

5.2.2. Test setup

The rheometer consists of a paddle shaped rotor and a cylindrical beaker which are concentric with each other. The dimensions of the cylinder are shown in Table 4. The schematic diagram of the cylindrical beaker and the paddle is shown in Fig. 2. To avoid slippage at the inner surface of the cylinder, protruding ribs were fitted. The paddle shaped rotor was designed in such a way as to avoid slippage and guarantee the homogeneity of the mixture.

The slump flow was measured using mini-slump test (DIN EN 1015-3 [19]). In addition, density of the cementitious suspensions was also measured. The mass equivalent of a volume of 416 cm^3 was taken for rheological measurements in the cylinder of the

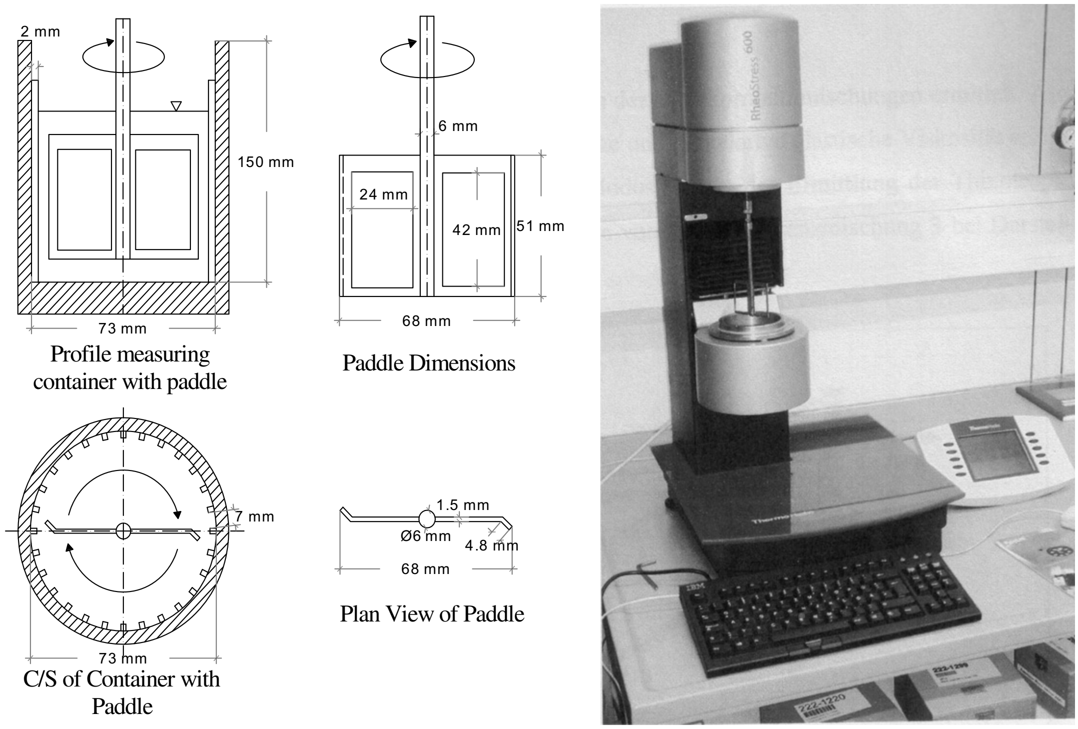

Fig. 2. Schematic diagram of cylinder and the paddle (left) and the Haake Rheostress 600 rheometer (right).

Table 3
Mixing procedure for the cementitious suspensions

Step no.	Activity	Speed	Time (s)
1	Dry mixing	1	120
2	Mixing dry materials and water	1	60
3	Hand mixing to break the clumped cement particles (if any) + cleaning paste on the walls		90
4	Mixing	2	60
5	Cleaning the paste on the walls		30
6	Mixing	2	120

Speed 1 – Agitator = 140 ± 5 rpm and attachment = 62 ± 5 rpm.
Speed 2 – Agitator = 285 ± 10 rpm and attachment = 125 ± 10 rpm.

Table 4
Dimensions of the rheometer specimen container

Dimensions	mm
Inner diameter	73
Inner height	150
Gap between ribs	7
Thickness of ribs	2

rheometer. All rheological tests were started 15 ± 1 min after the addition of water to the cementitious materials.

5.3. Results and discussion

5.3.1. Influence of the w/p ratio (by volume) on rheology of cementitious suspensions

The yield value, *g* (N mm) decreased with an increase in the *w*/*p* ratio, due to decrease in interparticle interaction (as a result of increased water volume), for all cement and silica fume (C + SF) (see Fig. 3) as well as cement and fly ash (C + FA) compositions (see Fig. 4). Correspondingly, this is evident from the graph between slump flow and *w*/*p* ratio, in which the slump flow increases with increase in *w*/*p* ratio for both the cases (see Fig. 3 and 4). For C + SF mixture, the torque was overloaded for *w*/*p* ratio of 1.00 and the data was not recorded.

At the same level of silica fume replacement or same packing density, by varying the *w*/*p* ratio, the yield value *g* of C + SF mixture varied between 3 and 10 times that of the yield value of pure cement paste (see Fig. 3).

The value of *h* (N mm s) increased with decrease in the *w*/*p* ratio for both C + SF and C + FA mixtures due to the increase in solids content (see Fig. 5 and Fig. 6). Moreover, silica fume and fly ash complement the deficiency in fine particle size in the particle size distribution of cement, which enhances viscosity. Relatively the change in *h* for C + FA is negligible in comparison with C + SF.

5.3.2. Influence of silica fume and fly ash on rheology of cementitious suspensions

As stated earlier, three compositions (C100, C88S12 and C74S26), which had the same packing density (53.50 vol.%), were taken into consideration for comparison. At the same *w*/*p* ratio, with increasing levels of replacement by silica fume, both g and h were increased (see Fig. 7). This may be attributed to the fact that the silica fume particles increase the water demand due to high surface area. In case of C + FA mixture, for constant *w*/*p* ratio, the yield value g decreased with increase in percentage replacement of fly ash (see Fig. 8). In other words, the yield value g decreases as the packing density of the C + FA mixture increases. The fly ash in the C + FA mixture acts as a lubricant in reducing the interparticle friction, thereby decreasing the yield value, which shows that good packing leads to good flow properties.

For constant *w*/*p* ratio, the increase in replacement of silica fume increases the value of *h* (see Fig. 7). On the other hand, for constant *w*/*p* ratio, the increase in fly ash percentage decreases the value of *h* (see Fig. 8). Relatively, the value of *h* for C + FA mixtures is negligible in comparison with the C + SF mixtures.

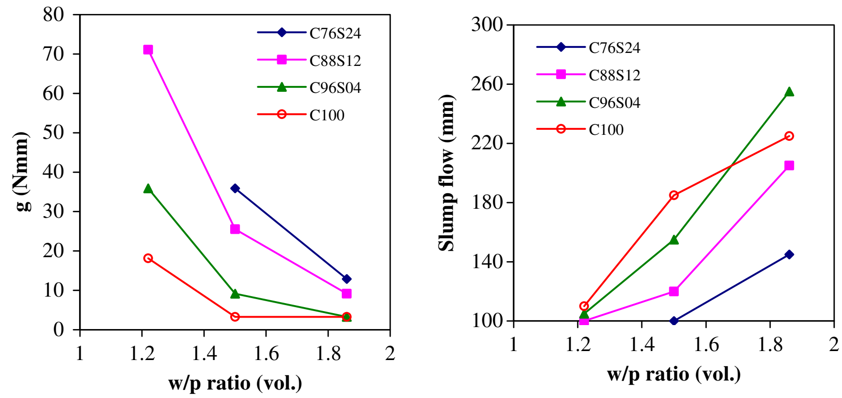

Fig. 3. Relationship between *g*, slump flow and *w*/*p* ratio of C + SF.

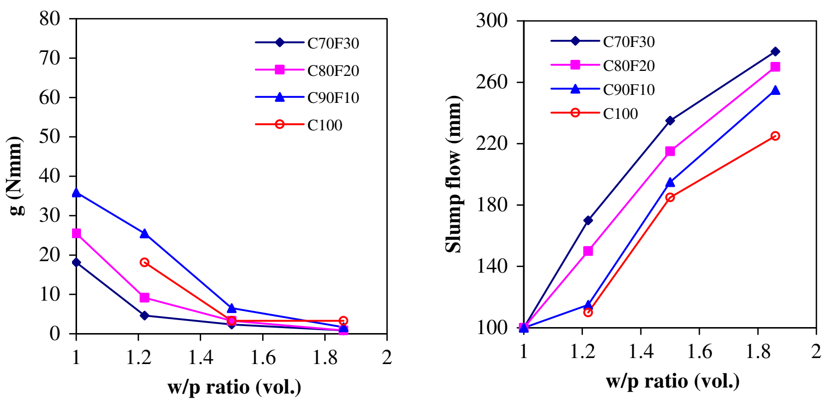

Fig. 4. Relationship between *g*, slump flow and *w*/*p* ratio of C + FA.

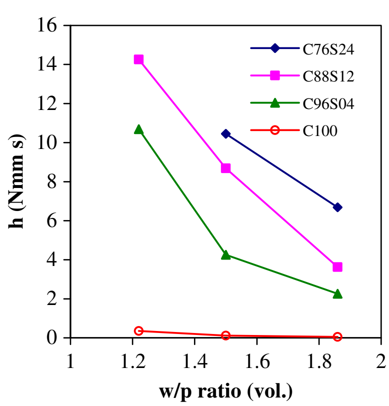

Fig. 5. Effect of *w*/*p* ratio of C + SF on *h*.

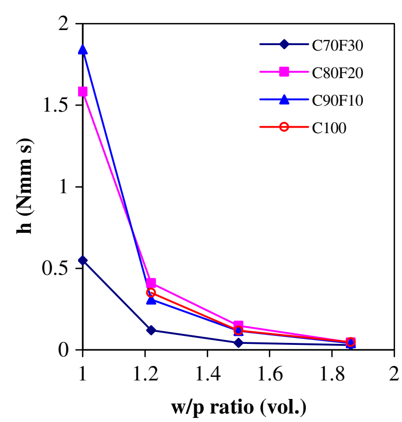

Fig. 6. Effect of *w*/*p* ratio of C + FA on *h*.

6. Conclusions

- The packing density of the C + SF mixture and C + FA mixture was calculated by Puntke test and based on the results, three compositions from each mixture were selected for studies of the flow properties.
- At the same level of silica fume replacement, by varying the *w*/*p* ratio, the yield value g of C + SF mixture varied between 3 and 10 times that of the yield value of pure cement paste. This increase in yield value may be attributed to increase in interparticle interaction, primarily the friction between the moving particles. The slump flow of C + SF mixtures was also lower compared to the pure cement paste.
- For the three compositions (C100, C88S12 and C74S26), which had the same packing density (53.50 vol.%), both *g* and *h* were increased with increasing level of replacement by silica fume for all *w*/*p* ratio. This effect is attributed to the higher specific surface of the silica fume particles, resulting in a higher water demand and an increase in yield value.

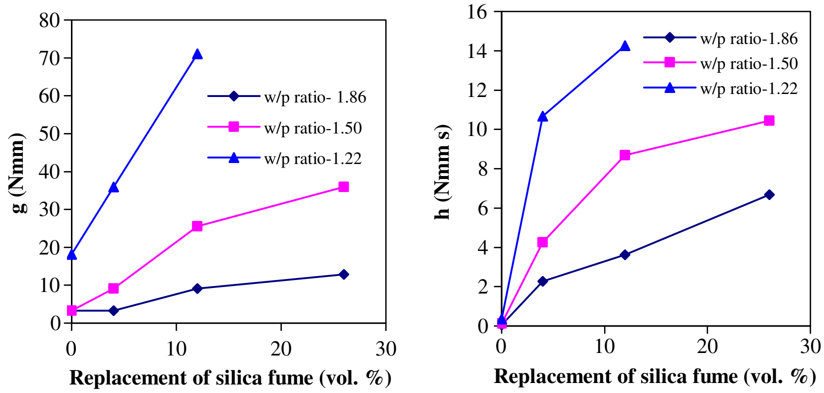

Fig. 7. Effect of replacement of silica fume on *g* and *h*.

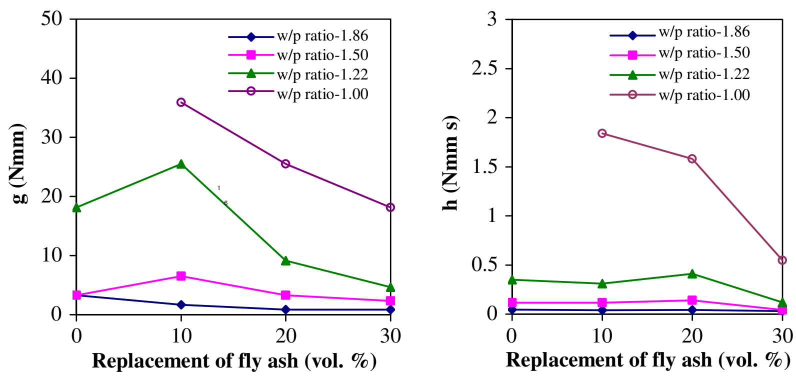

Fig. 8. Effect of replacement of fly ash on *g* and *h*.

- In case of C + FA mixtures, for constant *w*/*p* ratio, the yield value g decreased with increase in replacement of fly ash. In other words, the yield value *g* decreases as the packing density of the C + FA mixture increases proving that good packing density leads to enhanced flow properties.
- The value of *h* (N mm s) increased with decrease in the *w*/*p* ratio for C + SF and C + FA mixtures due to the corresponding increase in solids content. The silica fume and fly ash in the mixtures lead to an increased viscosity by increasing the cohesion of suspensions.
- For a constant *w*/*p* ratio, the increase in replacement of silica fume increases the value of the plastic viscosity (*h*). On the other hand, for a constant *w*/*p* ratio, the increase in fly ash content decreases the value of plastic viscosity (*h*).

In general, the packing density has a marked influence on the value of *g*, for both mixtures and has less influence on the value of *h* for C + FA mixtures.

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