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Viscosity prediction of cement-filler suspensions using interference model: A route for binder efficiency enhancement

Bruno L. Damineli [a], Vanderley M. John [a], Björn Lagerblad [b], Rafael G. Pileggi [a,*]

[a] Department of Construction Engineering, Escola Politécnica, University of São Paulo, 05508-900 São Paulo, Brazil
[b] Department of Construction Engineering, Royal Institute of Technology, SE-100 44 Stockholm, Sweden

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ABSTRACT

Producing cementitious materials with low CO_2 emissions is a key challenge for sustainability, considering the increasing demand for cement and the inefficacy of current industrial solutions. Improving the efficiency of binder use is mandatory, so that binder replacement by inert fillers with lower environmental loads is an alternative, which demands careful control of the rheological behaviour to decrease the water demand of pastes. Dispersion and packing models are well known, but other less explored parameters of raw materials (surface area, density, roughness) and paste (water content, distance between particles) determine the interaction among particles, affecting the paste's rheological behaviour.

The aim of this paper is to assess the influence of inert fillers on the rheological behaviour of cementitious pastes. A range of 12 inert fillers with varied aspects was evaluated. The results indicated a good agreement between the Casson viscosity and the interference parameter calculated using the interference model.

1. Introduction

Due to the demands in housing and infrastructure of the developing countries, cement production and related CO_2 emissions are increasing steadily; as a result, the industry is under pressure to reduce CO_2 emissions due to global warming constraints [1,2]. The current strategies used for decreasing CO_2 emissions – kiln efficiency, use of renewable fuels, and clinker replacement by blast-furnace slag and fly ash – have limits and will not prevent emissions to grow [2–4]. Carbon capture and storage could double the cement cost [5–7], which would primarily affect the poorest and neediest countries.

New options based on the improvement of binder use efficiency must be developed [8]. The partial replacement of clinker and reactive supplementary cementitious materials (SCMs) by progressively higher fractions of low-cost, widely available low CO_2 footprint mineral fillers is a better option than any calcined materials. The challenge is to develop low-binder products with performances and durability similar to those of the conventional high binder ones [9]. Limestone filler production releases from 26 to 75g CO_2/kg of material [10–11] mostly during the milling process, reducing CO_2 emissions proportionally to the increase of limestone content [12] and several other mineral fillers can be added to limestone as low footprint SCMs.

Since most of these mineral fine particles do not react with water [13,14], the amount of inert filler must be controlled in order to control porosity ensuring competitive mechanical performance and durability. In practical terms, the amount of water needed to achieve appropriate rheological behavior defines the maximum filler content. This might be one reason the actual average filler content in cement is around 6% despite the 34% limit in the European standard [15].

Efficient clinker–inert filler systems must have lower water content (vol./vol.) than the conventional systems with the adequate rheological behavior during mixing, transportation, and casting. This can be achieved by techniques of packing and dispersion of particles [16], even if packing models do not predict water demand since their unique aim is to calculate the volume of empty space among the particles. They usually assume particles as perfect spheres, ignoring surface area, morphology, and particles interactions. Most packing models also ignore the way the particles interfere on each other's movement during flow in highly concentrated suspensions. Divergences between predicted values of porosity and experimental results are assumed to be related to wall effects and irregularities in particles shapes, so they are considered in more advanced models [17].

In this scenario, models aiming to predict the amount of water to provide suitable rheological behavior are of practical interest. More precisely, both yield stress and viscosity parameters need prediction.

Regarding yield stress, significant progress in predictive models is observed in literature, as reported by Buscall et al., Kapur et al., Scales et al., Zhou et al., and Flatt & Bowen [18–22]. These researchers modeled different aspects of the suspensions, including particles size

* Corresponding author.
E-mail addresses: bruno.damineli@usp.br (B.L. Damineli), vmjohn@lme.pcc.usp.br (V.M. John), bjorn.lagerblad@cbi.se (B. Lagerblad), rafael.pileggi@usp.br (R.G. Pileggi).

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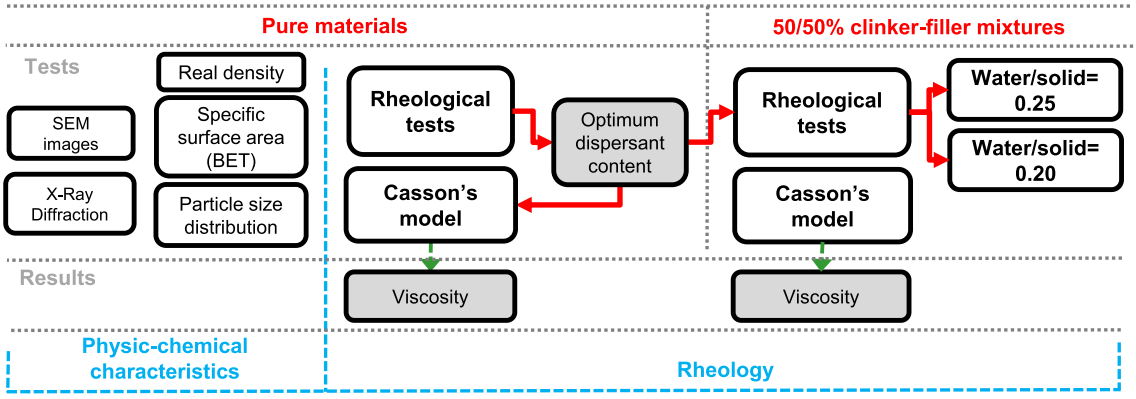

Fig. 1. Experimental planning flowchart.

distributions, volume fraction of solids, inter-particle forces, radius of curvature at particles contacts, etc.

Similar approach for viscosity forecast still need investigation, since most traditional models are based on packing factors [23–27]. A proposal to overcome this lack is the *Particles Interference Model* [28], which succeeded in correlating several physical aspects of particles and the immersing fluid in order to predict the viscosity of alumina ceramic suspension. This model has not been tested in Portland cement mixtures.

For this reason, the present paper aims to explore the Particles Interference Model to predict the viscosity of highly concentrated water suspensions of Portland cement with fillers with different mineralogical composition, when dispersed with admixtures.

2. Experimental setup

A CEM I 42.5 cement and a dozen of different fillers of four distinct mineralogical characteristics and particles smaller than 63 μm were tested, as well as 50/50 wt% cement-filler combined systems. The flowchart in Fig. 1 summarises the experimental plan, highlighting the materials characterization testing methods and the rheological evaluation program.

2.1. Materials

The fillers were named according to their predominant mineralogical characteristics: "D" = dolomite, "C" = calcite, "Q" = quartz, and "G" = granite. "Cem1" is the cement CEM I–42.5. All tested materials were commercially available in the Swedish market.

2.1.1. Chemical and mineralogical and characterization

Table 1 presents the chemical characterization results of the raw materials based on a semi-quantitative X-ray fluorescence analysis performed using a PANalytical Axios Advanced instrument. Table 2 presents mineralogical compositions of the raw materials evaluated by X-ray diffraction analysis using a PANalytical X'Pert PRO instrument equipped with an X'Celerator detector, as quantified by Rietveld method using PANalytical Inorganic Structure Database.

2.1.2. Physical characterisation

The powders were evaluated using the following testing methods:

1) particle-size distribution was determined using a Malvern Mastersizer laser diffraction granulometer, on liquid environment using water, pre-treatment on ultrasound dispersion for 2 min;
2) specific surface area was determined using the BET method with Gemini 2375 equipment, Micromeritics;
3) true density of particles was determined using a He picnometer Quantachrome MVP 5DC;
4) scanning electron microscopy (SEM) images were obtained using a Quanta 600FEG instrument.

The testing conditions for SEM images were as follows: 1) vacuum mode—high vacuum; 2) HV—10.00 kV; 3) det—ETD; 4) spot—2.0. The parameters of Mag and WD were varied.

Fig. 2 shows the particle size distributions of the tested materials. Table 3 presents values of the specific surface area (BET), the particles true density, and the calculated shape factor (ShF), herein defined as the ratio between the BET surface area and the theoretical surface area [29] calculated from the laser particle size distribution (Fig. 2), assuming that all of the particles are perfect spheres [30]. A high value of ShF indicates that particles deviate substantially from a spherical shape as a consequence of their shape and roughness. A ShF value close to 1 corresponds to a spherical, smooth particle. The calculated deviation from a spherical shape was confirmed by SEM micrographic analysis, as shown in Fig. 3.

The results shown in Table 3 and Fig. 3 make evident the need for extending the evaluation of particles beyond only size distribution. As a consequence, it is necessary to consider other characteristics to understand the rheological behaviour of cement-filler suspensions.

Table 1
Chemical compositions of the raw materials obtained via X-ray fluorescence (X-Ray PANalytical Axios Advanced).

Oxides (%)	Cem1	D1	D2	D3	C1	C2	C3	Q1	Q2	G1	G2	G3	G4
CaO	65.4	30.0	29.3	28.3	53.8	54.7	55.2	–	–	1.09	6.45	0.98	3.65
SiO_2	18.9	4.59	4.45	8.76	2.62	1.69	0.44	98.9	98.1	72.6	54.8	70.5	63.3
Al_2O_3	3.09	0.39	0.80	0.85	0.41	0.05	0.05	–	–	13.8	14.4	14.5	15.5
Fe_2O_3	5.27	0.58	0.53	0.66	0.15	0.11	0.05	–	–	1.97	10.70	2.33	6.53
MgO	0.68	21.40	21.30	21.20	0.19	0.15	0.87	–	–	0.32	3.44	0.55	1.83
SO_3	4.19	–	–	–	–	–	–	–	–	–	–	–	–
K_2O	0.78	0.05	0.21	0.28	0.05	0.05	0.05	–	–	5.22	2.56	5.68	3.78
Na_2O	0.09	0.05	0.05	0.05	0.05	0.05	0.05	–	–	3.24	2.63	2.81	2.84
TiO_2	0.22	0.05	0.05	0.05	0.05	0.05	0.05	–	–	0.25	1.76	0.22	0.84
Other	0.43	0.19	0.21	0.05	0.18	0.35	0.34	0.95	1.48	0.97	1.69	0.92	0.72