HAL open science

Robustness of cement-based materials: From dosage variations to yield stress fluctuations

Wenqiang Zuo, Hela Bessaies-Bey, Qian Tian, Changwen Miao, Nicolas Roussel

To cite this version:

Wenqiang Zuo, Hela Bessaies-Bey, Qian Tian, Changwen Miao, Nicolas Roussel. Robustness of cement-based materials: From dosage variations to yield stress fluctuations. Cement and Concrete Research, 2021, 139, pp.106260 -. 10.1016/j.cemconres.2020.106260 . hal-03493240

HAL Id: hal-03493240

https://hal.science/hal-03493240

Submitted on 9 May 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Version of Record: https://www.sciencedirect.com/science/article/pii/S0008884619316679
Manuscript_0d66bff183af0f8802d30a18a38bdf75

Robustness of cement-based materials: from dosage variations to yield stress fluctuations

Wenqiang Zuo [a,b], Hela Bessaies-Bey [a], Qian Tian [c,d], Changwen Miao [b,c,d], Nicolas Roussel [a*]

[a] Laboratoire Navier, Université Gustave Eiffel, ENPC, CNRS, F-77447 Marne-la-Vallée, France

[b] School of Materials Science and Engineering, Southeast University, Nanjing 211189, China

[c] Jiangsu Research Institute of Building Science Co.,Ltd, Nanjing 211100, China

[d] State Key Laboratory of High Performance Civil Engineering Materials, Nanjing 211100, China

[*] Corresponding author:

Email address: nicolas.roussel@ifsttar.fr (N. Roussel)

Abstract

The idea behind the present work is to assess robustness of fresh cement-based materials (*i.e.* their ability to display the same properties when submitted to variations in the way they are produced) from a generic and analytical point of view. First, analytical models from literature relating components proportions and yield stress are combined into a multi-scale and physical approach and compared to our experimental yield stress measurements. In the second part, the derivation of these analytical relationships is used to assess robustness as a function of components proportions, which allows for the most sensitive variations to be mapped onto component dosage from a yield stress point of view. In the case of fluid concretes, this approach showcases the interest of working at slightly higher water content and slightly lower aggregates fraction along with benefits of viscosity modifying agents / plasticizers combination.

© 2020 published by Elsevier. This manuscript is made available under the Elsevier user license
https://www.elsevier.com/open-access/userlicense/1.0/

Keywords: Fresh Concrete (A); Robustness; Yield stress; Rheology (A); Admixture (D)

1. Introduction

In the construction industry, the term “robustness” captures the ability of a product to display the same properties when submitted to variations in the way it is processed or used. However, when it comes to concrete, robustness most often represents the ability of the material to display the same properties when submitted to either some variations in constituent parts physical and chemical properties or some variations in constituent parts dosage.

In most academic or industrial research laboratories, mix design and sample production are carried out using only one unique cement batch. Aggregates are stored in controlled environmental conditions. Dosing and weighting uncertainties are low compared to industrial practice. For a given mixing process, most measurements standard deviations are therefore low. Such low standard deviations allow for the assessment and study of tiny variations in macroscopic properties typical of today’s research. As a consequence, robustness at the laboratory stage is often a fully ignored feature of a given mixture.

The large volumes of raw material required for industrial concrete production mandates the use of successive batches of cement and aggregates, increasing the variation in particle size distribution and fine content. In addition, storage in most often uncontrolled environmental conditions exacerbates control issues and variations in aggregate water content in particular. The accuracy in constituent materials weighing is far lower than in the laboratory. All the above results in higher variations in constituent parts proportions. In turn, these larger variations in constituent parts proportions and properties may induce larger variations in the resulting rheological properties of the fresh material. Consequently, formulations that had seemed fully adequate in terms of macroscopic properties in the laboratory may prove difficult to steadily produce on a daily basis at an industrial level. For instance, Ultra-High Performance Concrete (UHPC) [1] and Self-Compacting Concrete (SCC) [2] are recent examples of advanced cement-based materials, the commercial development of which has been hampered by, among others, some robustness

issues [3, 4].

Robustness of a given cement-based mixture is however an extremely delicate feature to tackle, as it often requires numerous experiments in order to capture variations in macroscopic behavior induced by variations in constituent parts properties and dosages. The most sensitive (or most critical) constituent properties or proportions may vary from one mix design to another. For instance, low W/C ratio concretes are far more sensitive to water content variations than high W/C ratio mixes whereas low slump concretes can be more sensitive to sand/gravel ratio than standard flowable concretes.

The idea behind the present work is therefore to make use of the most recent analytical models from literature allowing for the prediction of fresh properties of cement-based materials as a function of constituent parts properties and proportions. The obvious advantage of such relationships lies in their analytical nature, which offers the possibility to derive them and assess robustness from a generic and analytical point of view. This, in turn, may allow for a reduction of the amount of experimental measurements required to assess the robustness of a given cement based-mixture.

This above generic idea is illustrated in this work by a fluid concrete example (*i.e.* slump flow higher than 50 cm) although the same work could be carried out for other types of cement-based materials. The approach and the analytical relations used here would be the exact same ones. In this study, it is assumed that all components properties are fixed. The variations, to which the material is submitted, are only, therefore, the respective proportions of its constituent parts. This would correspond in practice to a situation, in which the material is produced from the same batches of constituents. Moreover, in this study, the focus is limited to the yield stress of the resulting mixture and its variations, as this rheological parameter is the most documented in literature and the most important in standard casting processes [5].

The objective of the first part of this paper is therefore to show how component proportions variations affect yield stress from the polymer adsorption scale to the

concrete scale. Analytical models from literature relating components proportions and yield stress are combined into a multi-scale and physical approach and compared to our experimental yield stress measurements.

In the second part of this paper, from the derivation of these analytical relations, we assess robustness as a function of components proportions and map the most sensitive variations in components dosage from a yield stress point of view.

Finally, in the specific case of the fluid concrete taken here as an example, we conclude on the potential changes in mix design that may result into a more robust mixture.

2. Background and methodology

First, it is important to make a distinction between material parameters that capture the constituent parts properties and are therefore non-dependent on mix proportions and the mixture variables, which result directly from mix proportions.

In this section, from literature, the number and nature of the variables theoretically required to describe one cubic meter are derived in the case of an optimized fluid concrete with a given yield stress and containing five constituents: water, cement, sand, gravel and a super-plasticizer (SP). The proportion of each of these five constituents is obviously a mixture variable by itself. However, when dealing with the proportions required to produce one cubic meter of concrete, it is therefore possible to describe all components proportions with only four mixture variables. Additionally, as the yield stress of the concrete is specified, this provides an additional relation, which leads to reduce the number of independent mixture variables to three. Finally, it is considered in this study that the studied mix has already been optimized. As a consequence, the aggregates size distribution and the relative proportion between sand and gravel have both already been fixed in order to maximize the packing properties (*i.e.* the maximal packing fraction) of the granular skeleton [6-8]. Such a material should therefore be fully described by using only two independent mixture variables. A mineral filler would often be added to such a mixture in order to reduce carbon foot print, to improve workability and to decrease cost, shrinkage and hydration heat according to [9-11] and ASTM C 1797. This would, in turn, increase the number of variables required to describe the mixture to three. For the sake of simplicity, this situation is however out of the scope of the present paper.

In order to identify and select the two previous variables, the present approach is based on several analytical relations from literature, which have all proved, in previous publications from various authors, their ability to predict the related underlying physics. These analytical relations include:

2.1. YODEL model for cement pastes

The Yodel is a first principle analysis of yield stress of colloidal particles suspension. It was shown to successfully predict the main parametric dependencies of cement suspensions yield stress [12-15]. This model is used here to predict the yield stress τ_p of the constitutive cement paste of the concrete (*i.e.* the matrix phase). The model can be written as:

$$\tau_p \cong m \frac{A_0 a^*}{d^2 H^2} \frac{{\varphi_c}^2(\varphi_c - \varphi_{perc})}{\varphi_{cm}(\varphi_{cm} - \varphi_c)} \quad (1)$$

Where 6 material parameters are:

m, a pre-factor, which depends on the cement particle size distribution; It is possible to compute it from the cement particle size distribution but it is simpler to measure the value of the product mA_0a^* in Eq. (1) on a reference cement paste as shown in [14, 15].

A_0, the non-retarded Hamaker constant taken as 1.6×10^{-20} J [12]*;*

a^*, the typical size of the surface defects of cement grains of the order of several hundreds of nm [15]*;*

d, the cement particle volume-average diameter d_{50} of the order of 10 µm. It can be measured using laser granulometry (see next section);

φ_{cm}, the maximal packing fraction of the cement powder. It can be measured using the water demand method developed by De Larrard (see next section) [6].

φ_{perc}, the percolation volume fraction; It results from the competition between Brownian motion and colloidal attractive forces between cement particles, which, in turn, depend on the SP dosage [15]. *This dependency is however weak. It is therefore considered here as a constant material parameter. It can be extrapolated from a yield stress measurement on a reference cement paste as described in* [14, 15].

And where 3 mixture variables are:

τ_p, the yield stress of the constitutive cement paste of the concrete;

φ_c, the solid volume fraction of cement particles in the cement paste;

H, the average surface-to-surface separating distance between

flocculated cement particles; It depends on the SP dosage as described in sub-section 2.2.

2.2. Statistical average separating distance between cement grains as a function of admixture surface coverage

The average separating distance between cement grains H [14] can be computed as:

$$\frac{1}{H^2} = \frac{\theta^2}{{H_p}^2} + \frac{8\theta(1-\theta)}{(H_p + H_0)^2} + \frac{(1-\theta)^2}{{H_0}^2} \quad (2)$$

Where 2 additional material parameters are:

H_p, the SP layer thickness at full surface coverage. It could be estimated from the molecular structure of the SP [16]*. It is however easier in practice and when faced with an unknown molecular structure to assess its value from yield stress measurements on fresh cements pastes containing a given dosage of SP* [15]*;*

H_0, the average separating distance between cement grains in a system not containing any admixtures (of the order of 1-2 nm) [17].

And where 1 mixture parameter is:

θ, the so-called "surface coverage" or the ratio between the amount of SP adsorbed and the amount of adsorbed SP at surface saturation, which can be extracted from an adsorption isotherm (see below).

2.3. Surface coverage as function of SP dosage

It is now accepted that any mathematical relations showing a proportionality at low dosages between SP dosage and SP adsorption and displaying an adsorption plateau at high SP dosages is able to capture the adsorption behavior of a given SP and the surface coverage evolution as function of SP dosage [18, 19]. Langmuir-type relation is an example of this kind of mathematical relations. In this work, for the sake of simplicity, the following analytical relation is used:

$$\theta = \frac{\kappa \cdot D_{rsp}}{1 + \kappa D_{rsp}} \quad (3)$$

Where D_{rsp} is an additional mixture parameter, defined as the relative dosage of SP with $D_{rsp} = D_{sp}/D_{sat}$, where D_{sp} is the SP dosage expressed as a mass proportion of the cement content per cubic meter and D_{sat} is the so-called saturation dosage (see below);

And where 2 material parameters are:

κ, a dimensionless adsorption and desorption equilibrium rate constant; κ can be assessed from the measurement of the polymer adsorption isotherm;

D_{sat}, the so-called saturation dosage, above which further increase in the dosage of SP does not increase the surface coverage. It can be assessed through SP adsorption measurements or yield stress measurements.

2.4. Chateau and Ovarlez model for mortar and concrete yield stress

The Chateau and Ovarlez model has proven its ability to predict the yield stress of a mixture containing rigid inclusions in a yield stress fluid [20, 21]. It was moreover shown to be able to predict the yield stress of fluid mortars and concretes when direct frictional contacts between aggregates were neglectable (*i.e.* high matrix volume content and/or fluid mixtures [7]). It can be expressed as:

$$\tau_M = \tau_p \sqrt{(1-\varphi)} \left(1 - \frac{\varphi}{\varphi_{div}}\right)^{-1} = \tau_p \sqrt{(1-\varphi)} \left(1 - \frac{\varphi}{0.8\varphi_{am}}\right)^{-1} \quad (4)$$

Where 2 additional mixture parameters are:

τ_M, the yield stress of the mortar or concrete mixture;

φ, the aggregates volume fraction at the level of one cubic meter of concrete;

And where one material parameter is:

φ_{div}, the aggregates volume fraction for which the yield stress diverges. It was shown that, for sand, gravels and fibers, $\varphi_{div} = 0.80\varphi_{am}$, where φ_{am} is the maximal packing fraction of the aggregates [7, 22]. *φ_{am} can be measured using independent compaction measurements carried on the dry*

aggregates [23].

2.5. Integration of all models

The yield stress τ_M of a fluid concrete can therefore, in theory, be predicted combining equations ((1) to (4)). Such an approach would involve the 11 material parameters listed above that do capture both the components properties and their interactions. These 11 parameters can all be experimentally and independently assessed. Because of the way the above analytical relations are built, these parameters are only component-dependent and not dosage-dependent (*i.e.* they do not change when varying components proportions).

The full description of the mixture from the above equations moreover involves seven mixture variables along with four multi-scale relations. This results into 3 independent variables. For these variables, the cement solid volume fraction φ_c at the scale of one cubic meter of the constitutive cementitious matrix (*i.e*. excluding any aggregates), the SP dosage D_{sp} (*i.e.* the mass ratio between SP and cement) and the aggregates volume fraction φ at the scale of one cubic meter of concrete or mortar were selected. As stated previously, only two of these three variables are enough to describe such a system if a target value for its yield stress is first specified.

3. Materials and experimental protocols

3.1. Materials properties

A commercial cement equivalent to ASTM Type I cement was used in this study. The chemical composition of the cement is shown in **Table 1**. The Particle Size Distribution (PSD) was measured by laser diffraction (MALVERN MASTERSIZER S) after dispersion in isopropanol and its volume average diameter d_{50} was around 12 micrometers (Cf. **Fig. 1**). The maximum packing fraction of this cement was 0.58. It was measured following the water demand method developed by De Larrard [6]. It

consists in finding the minimum water content allowing for the transition during mixing between pasty cement granules and a continuous homogeneous paste.

The properties of the aggregates used in this study are shown in **Table 2** and the PSD of the aggregates is shown in **Fig. 1**. The maximal packing fraction of the aggregates was measured using the following protocol. A container with the diameter and height of 16 cm and 32 cm respectively was fixed on a vibration table and was submitted to a 150 Hz vibration. A mass, equivalent to 1 kPa external pressure, was applied above the sample. For each dry batch of aggregates, 7.5 kg of aggregates were poured into the container and, after a 60 s vibration, the height of the sample was measured. Finally, using the mean apparent density of the aggregate mixtures, the maximum packing fraction was computed.

In addition to the independent assessment of sand and gravel, the packing properties of dry mixtures of sand and gravel with varying proportions were also measured (Cf. **Fig. 2**). The sand mass proportion was then chosen in order to optimize the packing properties of the granular skeleton and is therefore equal to 45%.

The SP used in this study is a non-blended commercial polycarboxylate provided under liquid form with a solid content of 29.5%. The recommended highest dosage of this SP is 3.0% by mass of cement.

3.2. Mixing protocols

Cement pastes with different dosages of SP ranging from 0% to 3.5% were prepared. The SP was first added into the mixing water (with controlled temperature of around 22 ± 2℃) prior to the contact with cement. For each dosage of SP, cement pastes with

W/C mass ratio ranging from 0.275 to 0.5 were prepared. For each sample, a volume of around 200 ml of cement paste was mixed for 3 min using a Turbo test Rayneri VMI mixer at the speed of 840 rpm. A total of 53 different cement pastes was prepared.

The mortars were produced from cement pastes prepared using the same protocol as above. Then, the sand was introduced into the cement paste and stirred for 2 min by hand in order to decrease the influence of mixing in the presence of rigid particles on the rheological properties of the interstitial constitutive cement paste [14]. The sand content was varied from 0 to 52.5% volume fraction (*i.e.* around 475 liters of paste per cubic meter) while the W/C ratio and SP mass dosage were varied between 0.275 to 0.50 and between 0 and 3% respectively in order to produce 41 different mortars.

In the case of concrete mixing, a Zyklos-type concrete mixer from Schwelm was used. The corresponding cement paste was first mixed alone for 3 min then the aggregates were introduced and dispersed for another 2 min. We checked that the cement pastes prepared in this mixer had a similar yield stress as the ones prepared with the Rayneri mixer above (results not shown here). Around 10 liters of concrete were produced for each concrete mixture. The total aggregate content was varied from zero to around 62% volume fraction (*i.e.* around 380 liters of paste per cubic meter of concrete) over 5 different concretes mixes.

All experiments were carried out at a temperature of 22 ± 2℃ and at a relative humidity of around 50% ± 5%.

All mix designs are provided as supplementary materials along with measured yield stress values.

3.3. Yield stress measurements

In this study, the values of the yield stress of cement pastes, mortars and concrete mixtures were computed from simple flow tests [7, 24].

At the scale of cement paste and mortar, volumes of around 200 ml and around 500 ml were used respectively to measure flow spread. Each mixture was slowly poured from the mixing cup on a horizontal steel plate. The spread diameter was assessed from the average of two measured diameters. The yield stress value was then computed using the following equation [24, 25]:

$$\tau_0 = \frac{225\rho g \Omega^2}{128\pi^2 R^5} \quad (5)$$

where τ_0 is the yield stress (Pa); ρ is the bulk density of the mixture (kg/m^3); Ω is the volume of the mixture (m^3); R is the spread radius (m).

At the scale of concrete, 6 l of material were slowly poured from a bucket into one end of the LCPC box and the spread length was measured [26]. The yield stress of the concrete mixtures was computed from the existing analytical correlation between the measured spread length in the LCPC box and the yield stress of the tested material [26, 27] (Cf. Eq. (6) and (7)).

$$V = \frac{{l_0}^3}{4A}[LN(1 + \mathrm{U}_0)] + \frac{U_0(U_0 - 2)}{2} \quad (6)$$

$$L = \frac{h_0}{A} + \frac{l_0}{2A} LN(\frac{l_0}{l_0 + 2h_0}) \quad (7)$$

where V is the tested concrete volume (L); l_0 the width of the LCPC box (20 cm); L the measured spread length of concrete in the box (cm); h_0 the height of the sample at the point where concrete is poured at the end of the test (cm); $A = 2\tau_0/\rho g l_0$, $U_0 = 2h_0/l_0$, where τ_0 is the yield stress of the concrete (Pa), ρ the apparent density of the concrete (kg·m^{-3}), g the gravity (m·s^{-2}). Solving the above equations simultaneously provides the value of the concrete yield stress. An abacus is also available in [26].

3.4. Polymer adsorption measurements

A total organic carbon (TOC) analyzer manufactured by Shimadzu was used in this work. Cement pastes with W/C equal to 0.4 and dosages of SP ranging from 0 % to 3.5% by mass of cement were mixed as described above. The cement pastes were then centrifuged and the extracted interstitial fluid was diluted and analyzed. By comparing the TOC values of the extracted interstitial fluid with a reference polymer solution, the amount of polymer adsorbed on cement grains was computed and the surface coverage (*i.e.* the ratio between adsorbed polymer and adsorbed polymer at saturation) as a function of the polymer relative dosage (i.e. ratio between dosage

and saturation dosage) is plotted in **Fig. 3**. The amount of organic carbon in the interstitial fluid of a reference cement paste without polymer was also measured to account for the presence in the cement powder of organic compounds such as grinding agents.

4. Experimental results and analysis of results

4.1. Yield stress of cement paste

The yield stress of cement paste as a function of cement volume fraction for various SP dosages is plotted in **Fig. 4**. Only non-bleeding cement pastes (visual observation) are shown in **Fig. 4**. The values of the measured yield stress range from around 1 Pa to around 100 Pa. It should be noted that the higher the SP dosage, the higher the minimum cement volume fraction required to obtain a non-bleeding mixture.

The prediction of the yodel model is also plotted in **Fig. 4** (continuous lines). It is worth noting that the only parameter changing from one SP dosage to another in the Yodel is the average surface-to-surface separation distance (H) between cement particles (not shown here). This distance is varied from 2 nm (reference paste) to 9 nm (full surface coverage). As already shown in previous papers [12, 13], the YODEL analytical model is able to capture the evolutions of a cement paste yield stress as a function of solid volume fraction and inter-particle distance over more than two orders of magnitude.

4.2. Yield stress of mortar

Fig. 5 shows the relative yield stress of the tested mortars (*i.e.* the ratio between the mortar yield stress and the yield stress of the corresponding constitutive cement paste) as a function of sand volume fraction for sand volume factions varying from 0% to around 50%. It can be seen that the values of the relative yield stress range from 1 to around 50 times the yield stress of the corresponding constitutive cement pastes. Interestingly but as already shown in [23] and as predicted by the Chateau and Ovarlez model, the relative yield stresses of the studied mortars does not depend on

the yield stress of the constitutive cement pastes. The amplification of the constitutive paste behavior by the rigid sand particles is well predicted in **Fig. 5** by the Chateau and Orvarlez model without any need for fitting nor corrections.

As predicted by Eq. (4), when the relative solid volume fraction (*i.e.* the ratio between the sand volume fraction and the maximum packing fraction of the sand as measured in section 3.1) approaches $0.80\varphi_{am}$ (dotted vertical line in **Fig. 5**), the yield stress of the tested mortars shows a sharp increase of a couple orders of magnitude [7, 22]. This feature indicates that frictional contacts between sand grains start to percolate and dominate the macroscopic rheology of the mixtures [20, 21, 23].

4.3. Yield stress of concrete

Fig. 6 represents the relative yield stress of the concrete (*i.e.* the ratio between the yield stress of the concrete and the yield stress of the constitutive cement paste) as a function of the total aggregate volume fraction (*i.e.* sand and gravel). The relative yield stress predicted by the Chateau and Ovarlez model is also plotted in **Fig. 6**. As expected from the previous section, the model captures the experimental data and shows a divergence as the aggregates volume fraction reaches the divergence volume fraction around 62% (vertical dotted line in **Fig. 6**).

5. Robustness assessment

5.1. Robustness index definition

In order to assess the robustness of a given mixture, we define here a robustness index R_τ for a component dosage variation. As yield stress is the parameter of interest in this study, such an index can be expressed as follows:

$$R_\tau = \frac{d\tau_m}{dx} \cdot \frac{\Delta x_{max}}{\tau_m} \tag{8}$$

where R_τ is the robustness index from the yield stress point of view (dimensionless). τ_m is the yield stress of the mixture (Pa); x stands for the dosage of one of the mixture components while Δx_{max} is the maximum variation of this dosage in

industrial practice. In order to define Δx_{max}, the average variations of components dosage in concrete plants from various technical recommendations and studies [28, 29] are considered (Cf. **Table 3**). It has to be noted that the derivation in Eq. (8) includes the fact that, if the volume of one component is increasing or decreasing, then the volume of all other components has to be decreased or increased respectively in order to cover for the total volume change at the level of the considered one cubic meter of concrete. This effect is neglectable for SP and cement dosage variations but plays a non-neglectable role for aggregates and water dosages variations. Finally, it has to be kept in mind that, in the following, the higher the robustness index, the less robust is the mix.

It can first be noted that the more restrictive values from the ACI standards [28] seem to be closer to real industrial practice when compared to the industry-representative values reported in literature [30-32]. We choose therefore to consider these limits in this study.

However, the water dispenser accuracy from the ACI standard [28] in **Table 3** does not take into account some potential uncertainties on the moisture content of the sand [4, 33]. These are reported to be measured with a 0.5% accuracy for most existing sensors as reported in technical data sheets for this kind of sensors. Some more accurate water-moisture online assessment technologies do exist but we do not consider that they are representative of the common practice.

Moreover, we choose here to turn these relative uncertainties requirement for standard concrete production into absolute uncertainties in order to extrapolate the consequences on robustness of these variations for non-standard component dosages. In order to do so, we consider that the previous uncertainties are given for one cubic meter of standard concrete that is considered here to contain 300 kg of cement, 150 kg of water, 800 kg of sand, 1200 kg of gravel and 3 kg of super-plasticizer. The absolute variation for water dosage is then computed as the square root of the sum of the water dispenser accuracy (4.5 kg) at the power two and the uncertainty of the sand moisture sensor (4 kg) at the power two resulting in an overall absolute uncertainty of 6 kg. Due to the fact that the accuracy of adding SP

might be difficult to guarantee during the opening and closing of the SP feed port, a higher absolute variation for SP dosage (i.e. 150 g) is considered according to the EHE limits [29].

Finally, we choose here to ignore any potential variations in the sand-to-gravel ratio and only consider variations in the total aggregate content. The reason behind is that aggregates influence on yield stress in mostly driven by total solid fraction and maximum packing fraction of the sand-gravel mixture. However, variations in sand to gravel ratio for an optimized mixture of aggregates do not induce strong variations in maximum packing fraction. Indeed, around the optimal sand-gravel proportions, variations in maximum packing properties of the resulting granular skeleton are neglectable as illustrated in this work in **Fig. 2**. Similar to water dosage variations, the overall absolute uncertainty on the total aggregate content is then computed from the combined variations in sand and gravel content and reported in **Table 3**.

5.2. Robustness maps

5.2.1. SP dosage variations

Fig. 7 represents the robustness index for SP dosage variations. It is plotted for the 150g SP dosage variation from **Table 3** for W/C between 0.3 and 0.55 and relative aggregates content (*i.e.* the ratio between aggregates volume fraction and maximum packing fraction) between 74 and 79%. This W/C range is typical of most fluid concretes as it covers high strength materials (excluding ultra-high-performance material) for civil engineering application along with standard concretes for housing applications. Obviously, lower W/C shall lead to better mechanical and transfer properties while higher aggregates volume fraction shall result in cheaper mixes as they lead to lower matrix and cement content. Two features bound the range covered here by the aggregates relative fraction variation. On one hand, for the highest aggregates relative fraction, as illustrated in **Fig. 6**, it is not possible to mix design a stable fluid material as direct frictional contacts between aggregates dominate [7]. No matter the amount of water or SP, the material shall either display a high yield stress or be unstable [34]. On the other hand, for lowest aggregates

relative fraction, the matrix volume becomes high inducing potential shrinkage, hydration heat and/or creep issues [35].

For a SP saturation dosage taken as 2% of the cement mass, **Fig. 7** shows that the highest robustness index is around 20% (*i.e.* 10 Pa variation) and occurs for the highest W/C and lowest relative aggregates volume fraction. It is indeed in this mix design range that the dosage of SP is the lowest. As a consequence, the relative variation in SP dosage for a given dosage accuracy is the highest. This feature is amplified by the shape of the adsorption isotherm in **Fig. 3**. Indeed, as the dosage of SP gets closer to the saturation dosage, the sensitivity of surface coverage to SP dosage decreases and the system becomes more robust from a SP dosage point of view. This is one of the reasons why admixtures manufacturers often recommend a minimum SP dosage. This minimum dosage ensures that the resulting mix is not too sensitive to SP dosage variation in industrial production. As a consequence, at high W/C, it is obviously safer to use a less efficient SP that would require a higher dosage to get a similar yield stress. For instance, the robustness index values around 20% in **Fig. 7** become of the order of 15% if the saturation dosage of the SP used increases from 2% to 3% (*i.e.* less effective SP).

5.2.2. Cement dosage variations

Fig. 8 represents the robustness index for cement dosage variations. It is plotted for the 3 kg cement dosage accuracy from **Table 3**. Firstly, it can be noted that the mixture is far less sensitive to standard cement variations than it is to standard SP variations. Moreover, it can be noted that the trend here is more complex with different regimes clearly appearing, namely for low and high W/C and for high relative aggregates volume fractions.

It is first obvious that the system is more sensitive to cement dosage variation when the W/C is low. Indeed, at high cement volume fractions (*i.e.* close to the cement powder maximum packing fraction), as shown in **Fig. 4**, despite the log scale, yield stress shows an extremely high sensitivity to solid volume fraction.

At high W/C, on the other hand, the dosage of SP and the resulting surface coverage

is low. A variation in cement content result therefore in a variation in surface coverage, which, in turn, induces variations in yield stress as discussed above for **Fig. 7**.

Finally, at high relative aggregates volume fractions, our analysis suggests that the higher sensitivity of the mixture finds its origin in the fact that, at the level of one cubic meter, a decrease in cement dosage results in a relative increase in aggregates content. Such systems are however close to the divergence aggregates volume fraction illustrated in **Fig. 6** and are therefore very sensitive to small fluctuations in aggregate content.

Overall, however, it will be seen further that fluctuations in yield stress induced by cement dosage variations are neglectable compared to the other sources of fluctuations.

5.2.3. Water dosage variations

Fig. 9 represents the robustness index for water dosage variations. It is plotted for the 6 kg water dosage accuracy from **Table 3**. The high sensitivity of the system to water dosage compared to SP and cement can first be noted. Moreover, as W/C is decreasing, the sensitivity of the mixture to water variations increases. This finds its origin in the robustness of the cement paste itself that decreases at low W/C as shown in **Fig. 4** where the yield stress, for high solid fractions, shows an extremely high sensitivity to solid volume fraction.

We moreover note that, when the relative aggregate volume fraction increases, the sensitivity of the mixture to water increases. This unexpected trend finds its origin in the indirect consequence of a decrease in water dosage at the scale of one cubic meter. If the water content decreases, the proportions of all other components increase. A 6 liters variation in water is indeed able to induce a 15 kg variation in aggregate content or around 20 kg in cement dosage. As shown previously, cement content variations only had minor consequences. This is not the case for aggregates content, especially at high aggregate fractions when yield stress becomes extremely sensitive to aggregate content as shown in **Fig. 6**.

5.2.4. Aggregate dosage variations

Fig. 10 shows the robustness index for aggregate dosage variations. It is plotted for the 29 kg total aggregate dosage accuracy from **Table 3**. Firstly, it can be noted that this graph shows the second highest sensitivity after water variations (Cf. **Fig. 10**). The robustness index does not depend on the W/C ratio but only on the relative aggregates solid volume fraction. For high aggregate fractions, yield stress becomes extremely sensitive to aggregate content as shown in **Fig. 6**.

5.2.5. Practical consequences on the mix design of robust fluid concretes

Fig. 11 shows the overall sensitivity map of the fluid concrete taken as an example here by computing the quadratic average of all the above robustness indexes. Unsurprisingly, the overall answer is dominated by the influence of water variations and aggregates dosage variations. As a consequence, although the most economical mixtures shall be obtained for aggregates relative solid fraction close to 80%, **Fig. 11** suggests that mixtures with acceptable robustness are obtained for lower aggregates dosages than the economical optimum.

Moreover, **Fig. 11** suggest that more robust mix could be produced by moving to higher W/C ratio (when obviously mechanical strength and durability are not an issue). This could reach a limit at high W/C ratio combined with lower aggregates dosage as it would lead to a decrease in the SP dosage required and bring the system to the darkest zone of Fig. 7 where the mixture becomes very sensitive to SP dosage. One solution would then be to combine the SP with a viscosity modifying agent (VMA). The resulting adsorption competition between these molecules [36, 37] would increase the required SP dosage and decrease therefore the sensitivity of the mixture to SP dosage. It should be noted that this solution seems to be adopted in many countries where Self-compacting Concretes are designed with VMAs. Our results suggest, however, that VMAs do not increase robustness directly as sometimes suggested [38] but, instead, allow for the design of mixtures with higher water content or higher SP dosages, which are, in turn, more robust.

Finally, as both aggregates content and water dosage are strongly correlated to aggregates water content and aggregates water content measurement, it can be extrapolated that, from a concrete production point of view, it is on its ability to measure aggregates water content that the concrete plant technology performance has the strongest influence on a fluid concrete robustness.

5 Conclusions

The idea behind the present work was to assess robustness of fresh cement-based materials from a generic and analytical point of view.

In a first part, analytical models from literature relating components proportions and yield stress were combined into a multi-scale and physical approach and compared to our experimental yield stress measurements.

In the second part of this paper, from the derivation of these analytical relations, we assessed robustness as a function of constituent parts proportions and mapped the most sensitive variations in constituent parts proportions from a yield stress point of view.

The results in this investigation demonstrated the dominant sensitivity of the mixture to water and aggregates dosages and, indirectly, to aggregates moisture variations. Although these results are known in practice, the work presented here allows, for the first time, for the quantitative prediction of concrete robustness.

Moreover, the results, obtained here in the case of the fluid concrete taken as an example of the methodology, provide some physical explanations for the positive effect on robustness of some trends in SCC mix design. For instance, working with slightly lower fractions of aggregates than the economical *optimum* brings the system further from the aggregates contact percolation packing fraction where the sensitivity of the system to aggregates dosage is the highest. Similarly, increasing slightly the water content brings the system further from the cement powder maximum packing fraction where the sensitivity to water dosage is the highest. However, increasing water dosage and decreasing aggregate content both induce a

decrease in the required plasticizer dosage. This, in turn, increases the sensitivity of the mixture to plasticizer dosage. Combining viscosity modifying admixtures with plasticizers, because of their competitive adsorption, is a potential way to go back to standard plasticizer dosages and improve robustness.

Acknowledgement

The authors would like to acknowledge the financial support from National Natural Science Foundation of China (No. 51890904, No. 51708108).

Conflict of interest

The authors declare that they have no conflict of interest.

Reference

[1] B.A. Graybeal, Material property characterization of ultra-high performance concrete, in, United States. Federal Highway Administration. Office of Infrastructure …, 2006.

[2] H.J.H. Brouwers, H.J. Radix, Self-Compacting Concrete: Theoretical and experimental study, Cement and Concrete Research, 35 (2005) 2116-2136.

[3] I. González-Taboada, B. González-Fonteboa, F. Martínez-Abella, N. Roussel, Robustness of self-compacting recycled concrete: analysis of sensitivity parameters, Materials and Structures, 51 (2018) 8.

[4] S. Naji, S.-D. Hwang, K.H. Khayat, Robustness of self-consolidating concrete incorporating different viscosity-enhancing admixtures, ACI materials journal, 108 (2011) 432.

[5] N. Roussel, Rheology of fresh concrete: from measurements to predictions of casting processes, Materials and Structures, 40 (2007) 1001-1012.

[6] F. De Larrard, Concrete mixture proportioning: a scientific approach, CRC Press, 2014.

[7] J. Yammine, M. Chaouche, M. Guerinet, M. Moranville, N. Roussel, From ordinary rhelogy concrete to self compacting concrete: A transition between frictional and hydrodynamic interactions, Cement and Concrete Research, 38 (2008) 890-896.

[8] F. De Larrard, T. Sedran, Mixture-proportioning of high-performance concrete, Cement and concrete research, 32 (2002) 1699-1704.

[9] F.V. Mueller, O.H. Wallevik, K.H. Khayat, Linking solid particle packing of Eco-SCC to material performance, Cement and Concrete Composites, 54 (2014) 117-125.

[10] W. Zuo, W. She, W. Li, P. Wang, Q. Tian, W. Xu, Effects of fineness and substitution ratio of limestone powder on yield stress of cement suspensions, Materials and Structures, 52 (2019) 74.

[11] G. Ye, X. Liu, G. De Schutter, A.-M. Poppe, L. Taerwe, Influence of limestone powder used as filler in SCC on hydration and microstructure of cement pastes, Cement and Concrete Composites, 29 (2007)

94-102.
[12] R.J. Flatt, P. Bowen, Yodel: a yield stress model for suspensions, Journal of the American Ceramic Society, 89 (2006) 1244-1256.
[13] R.J. Flatt, P. Bowen, Yield stress of multimodal powder suspensions: an extension of the YODEL (Yield Stress mODEL), Journal of the American Ceramic Society, 90 (2007) 1038-1044.
[14] J. Hot, H. Bessaies-Bey, C. Brumaud, M. Duc, C. Castella, N. Roussel, Adsorbing polymers and viscosity of cement pastes, Cement and concrete research, 63 (2014) 12-19.
[15] A. Perrot, T. Lecompte, H. Khelifi, C. Brumaud, J. Hot, N. Roussel, Yield stress and bleeding of fresh cement pastes, Cement and Concrete Research, 42 (2012) 937-944.
[16] R. Flatt, I. Schober, Superplasticizers and the rheology of concrete, in: Understanding the rheology of concrete, Elsevier, 2012, pp. 144-208.
[17] G. Gelardi, R. Flatt, Working mechanisms of water reducers and superplasticizers, in: Science and Technology of Concrete Admixtures, Elsevier, 2016, pp. 257-278.
[18] D. Marchon, S. Mantellato, A. Eberhardt, R. Flatt, Adsorption of chemical admixtures, in: Science and Technology of Concrete Admixtures, Elsevier, 2016, pp. 219-256.
[19] R.J. Flatt, I. Schober, E. Raphael, C. Plassard, E. Lesniewska, Conformation of adsorbed comb copolymer dispersants, Langmuir, 25 (2008) 845-855.
[20] X. Chateau, G. Ovarlez, K.L. Trung, Homogenization approach to the behavior of suspensions of noncolloidal particles in yield stress fluids, Journal of Rheology, 52 (2008) 489-506.
[21] F. Mahaut, X. Chateau, P. Coussot, G. Ovarlez, Yield stress and elastic modulus of suspensions of noncolloidal particles in yield stress fluids, Journal of Rheology, 52 (2008) 287-313.
[22] L. Martinie, P. Rossi, N. Roussel, Rheology of fiber reinforced cementitious materials: classification and prediction, Cement and Concrete Research, 40 (2010) 226-234.
[23] H. Hafid, G. Ovarlez, F. Toussaint, P. Jezequel, N. Roussel, Effect of particle morphological parameters on sand grains packing properties and rheology of model mortars, Cement and Concrete Research, 80 (2016) 44-51.
[24] N. Roussel, P. Coussot, “Fifty-cent rheometer” for yield stress measurements: from slump to spreading flow, Journal of rheology, 49 (2005) 705-718.
[25] N. Roussel, C. Stefani, R. Leroy, From mini-cone test to Abrams cone test: measurement of cement-based materials yield stress using slump tests, Cement and Concrete Research, 35 (2005) 817-822.
[26] N. Roussel, The LCPC BOX: a cheap and simple technique for yield stress measurements of SCC, Materials and structures, 40 (2007) 889-896.
[27] T. Nguyen, N. Roussel, P. Coussot, Correlation between L-box test and rheological parameters of a homogeneous yield stress fluid, Cement and Concrete Research, 36 (2006) 1789-1796.
[28] A. ACI, 117-Standard Specifications for Tolerances for Concrete Construction and Materials, American Concrete Institute International, (2006).
[29] E.I.E. de Hormigón Estructural, EHE-08 (Spanish Code Structural Concrete), Ministerio de Fomento, Madrid, (2008).
[30] D. Bonen, Y. Deshpande, J. Olek, L. Shen, L. Struble, D. Lange, K. Khayat, Robustness of self-consolidating concrete, in: Proceedings of the Fifth International RILEM Symposium on Self-Compacting Concrete, 2007, pp. 33-42.
[31] J. Rigueira, E. Taengua, P. Serna, 13. Robustness of SCC dosages and its implications on large-scale production, in: 5th International RILEM Symposium on Self-Compacting Concrete, RILEM

Publications SARL, 2007, pp. 95-101.
[32] L. Lohaus, P. Ramge, H. Höveling, S. Anders, 12. Superplasticizer-based approach for optimized paste-composition and robustness of SCC, in: 5th International RILEM Symposium on Self-Compacting Concrete, RILEM Publications SARL, 2007, pp. 89-94.
[33] S. Nunes, H. Figueiras, P.M. Oliveira, J.S. Coutinho, J. Figueiras, A methodology to assess robustness of SCC mixtures, Cement and Concrete Research, 36 (2006) 2115-2122.
[34] N. Roussel, A theoretical frame to study stability of fresh concrete, Materials and structures, 39 (2006) 81-91.
[35] K.H. Khayat, G. De Schutter, Mechanical properties of self-compacting concrete, state-of-the-art report of the RILEM technical committee, (2016).
[36] H. Bessaies-Bey, R. Baumann, M. Schmitz, M. Radler, N. Roussel, Organic admixtures and cement particles: Competitive adsorption and its macroscopic rheological consequences, Cement and Concrete Research, 80 (2016) 1-9.
[37] H.B. Bey, J. Hot, R. Baumann, N. Roussel, Consequences of competitive adsorption between polymers on the rheological behaviour of cement pastes, Cement and Concrete Composites, 54 (2014) 17-20.
[38] P. Billberg, K.H. Khayat, Use of viscosity-modifying admixtures to enhance robustness of SCC, in: The Third North American Conference on the Design and Use of Self-Consolidating Concrete, 2008.

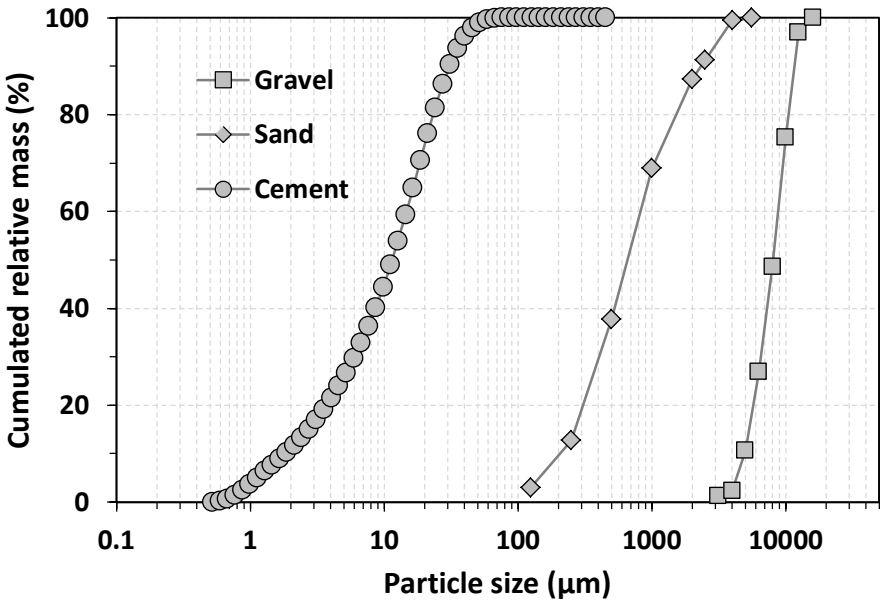

Figure 1. Particle size distribution of the cement and sand used in this study.

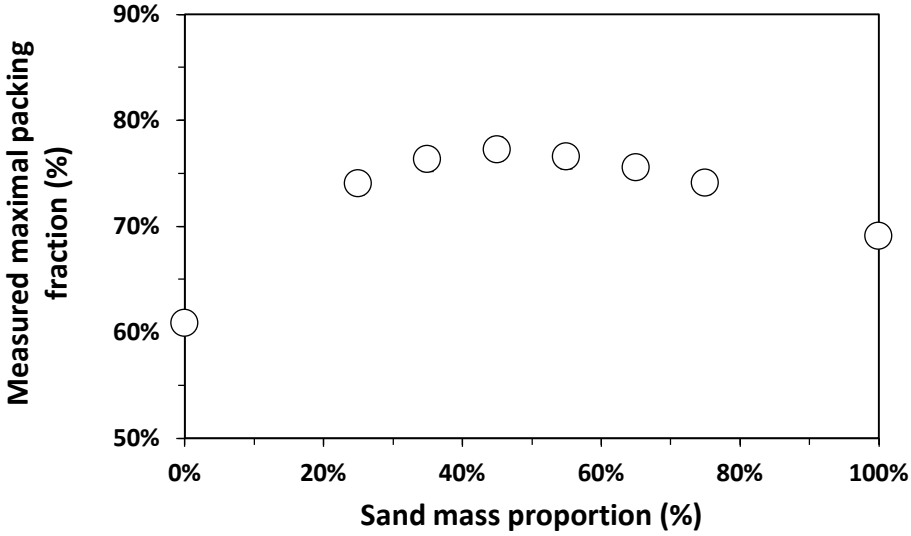

Figure 2. Maximal packing fraction of aggregate mixtures as a function of sand mass proportion at the level of the granular skeleton.

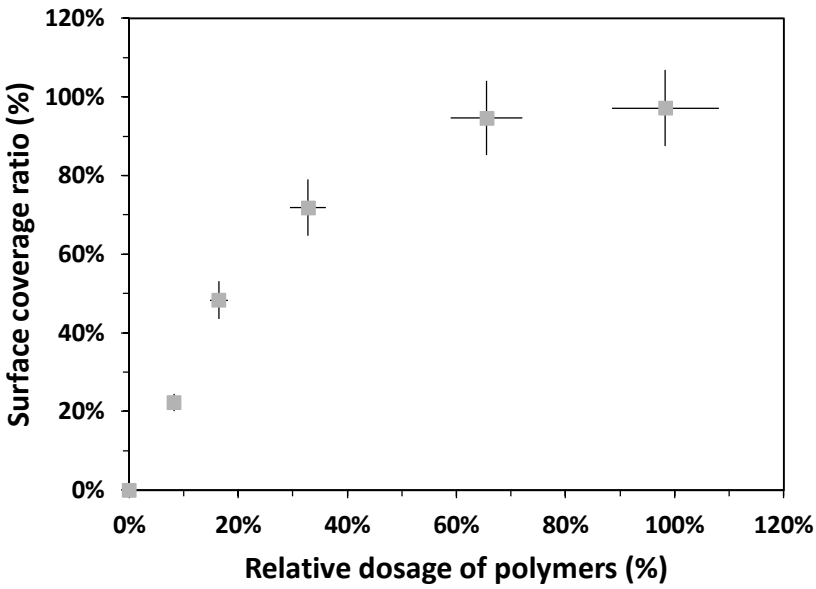

Figure 3. Surface coverage ratio (i.e. ratio between adsorbed polymer and adsorbed polymer at saturation) as a function of D_{rsp}, the polymer relative dosage (i.e. ratio between dosage and saturation dosage).

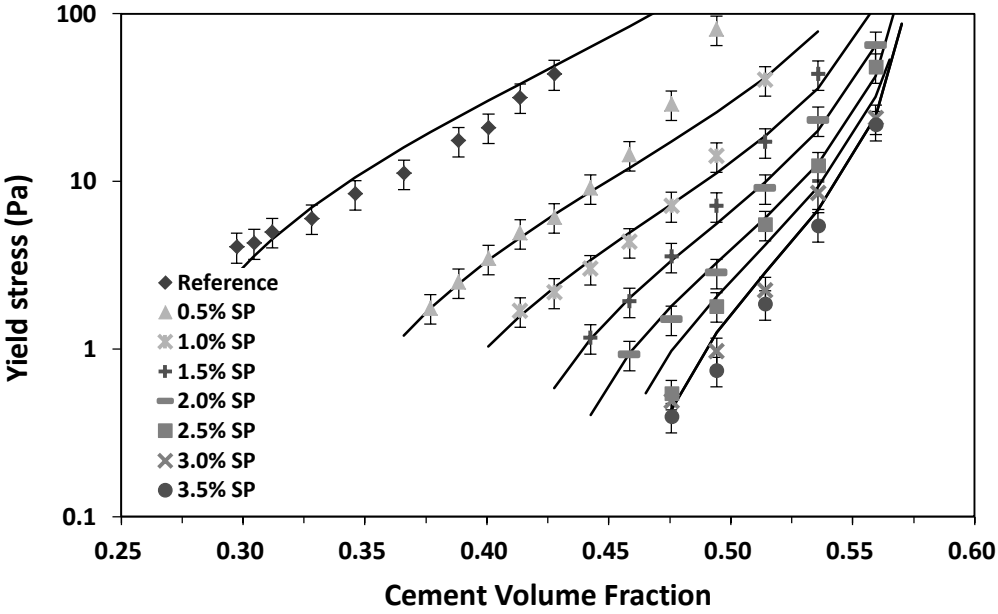

Figure 4. Yield stress of cement paste as a function of cement volume fraction for various SP dosages for the two types of the cement. The solid lines are the computed predictions of the YODEL.

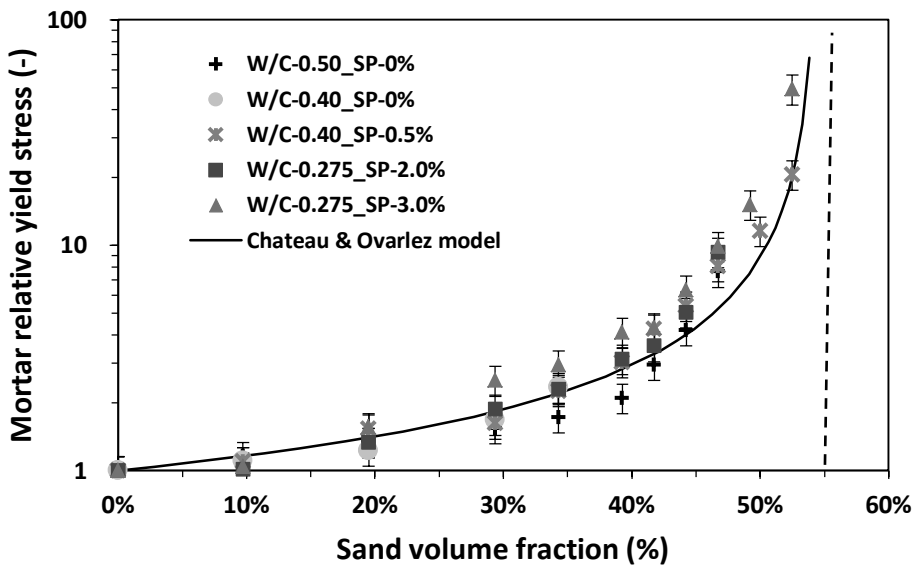

Figure 5. Mortar relative yield stress (i.e. ratio between the mortar yield stress and the constitutive cement paste yield stress) as a function of the sand volume fraction. The dotted vertical line corresponds to the theoretical divergence volume fraction (see text).

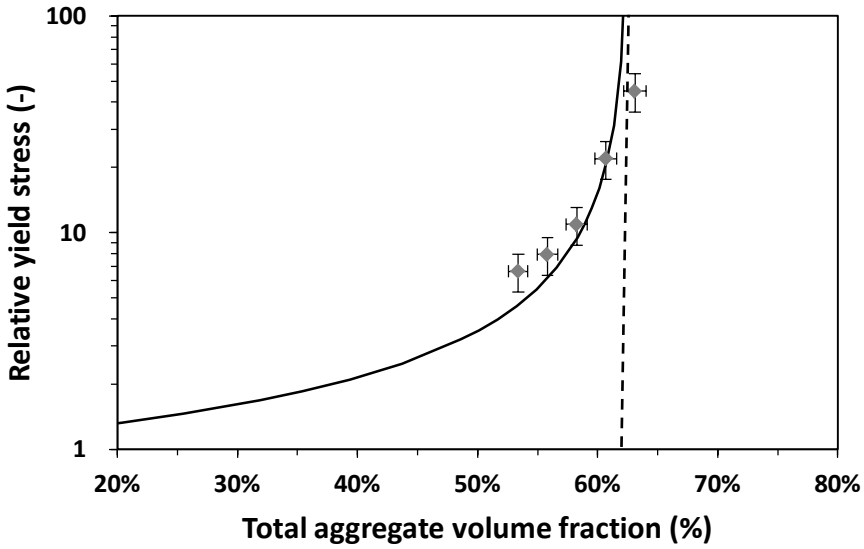

Figure 6. Relative yield stress of concrete as a function of aggregate volume fraction. Continuous line corresponds to the fitting of the Chateau and Ovarlez model (see text).

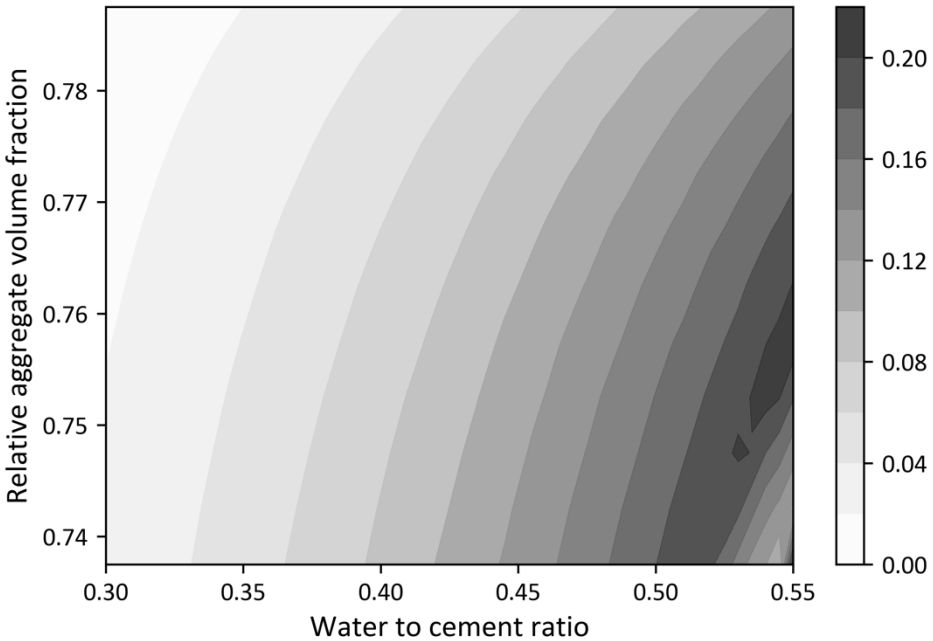

Figure 7. Robustness index as a function of W/C ratio and relative aggregates volume fraction for standard SP variations in industrial production. SP saturation dosage taken as 2% of the cement mass.

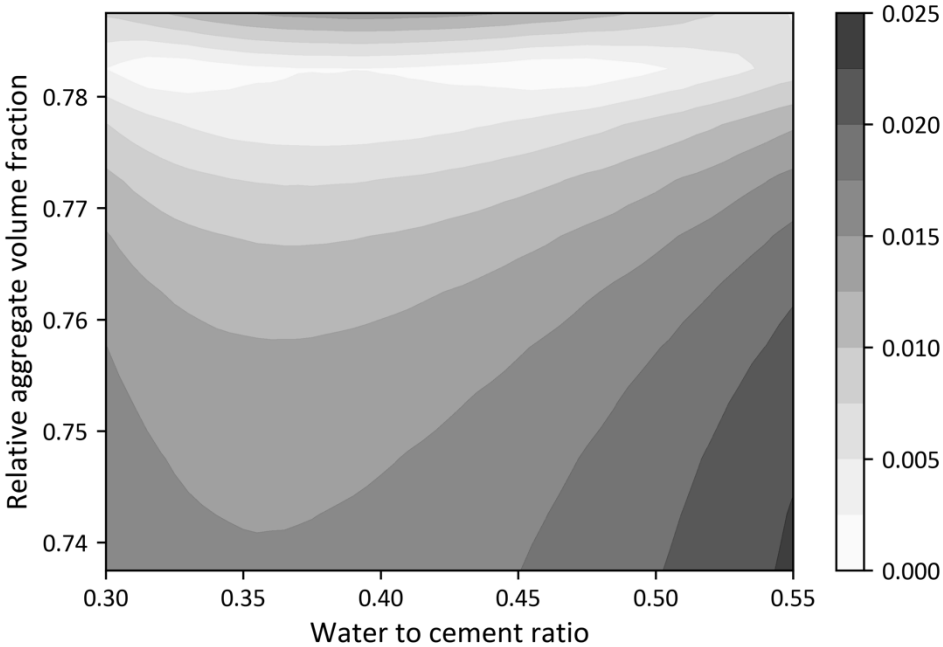

Figure 8. Robustness index as a function of W/C ratio and relative aggregates volume fraction for standard cement variations in industrial production.

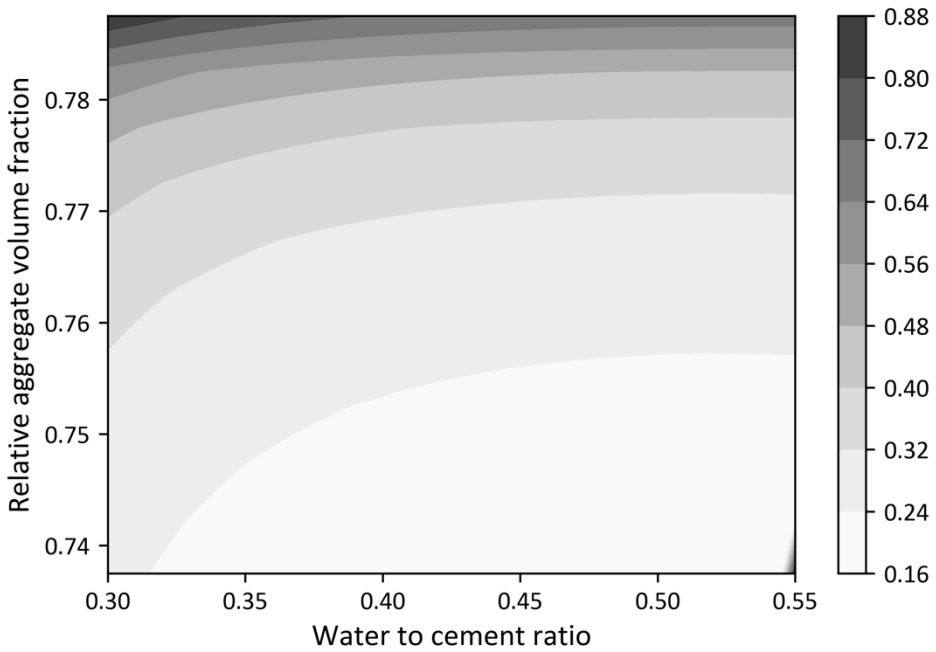

Figure 9. Robustness index as a function of W/C ratio and relative aggregates volume fraction for standard water dosage variations in industrial production.

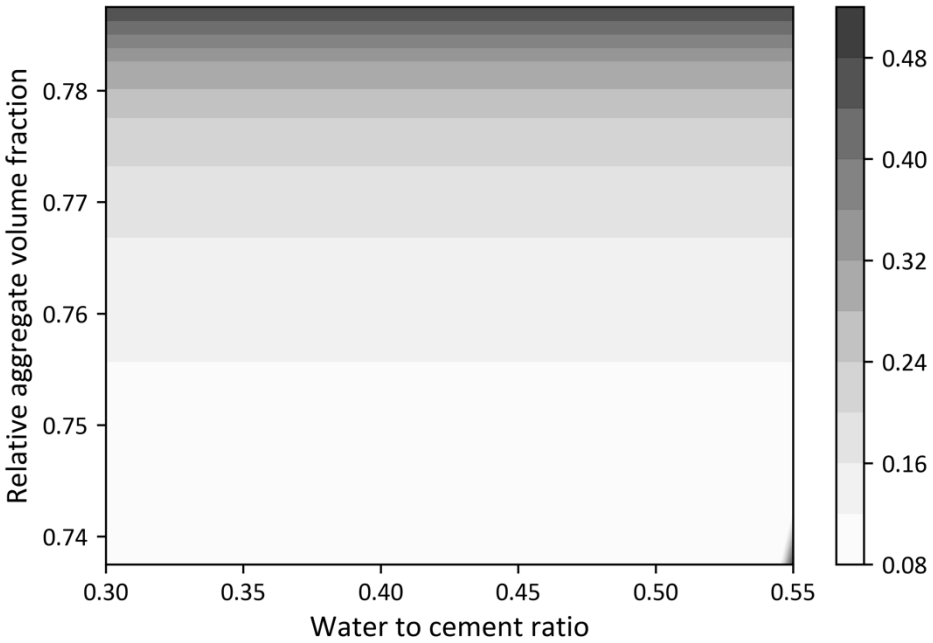

Figure 10. Robustness index as a function of W/C ratio and relative aggregates volume fraction for standard aggregates dosage variations in industrial production.

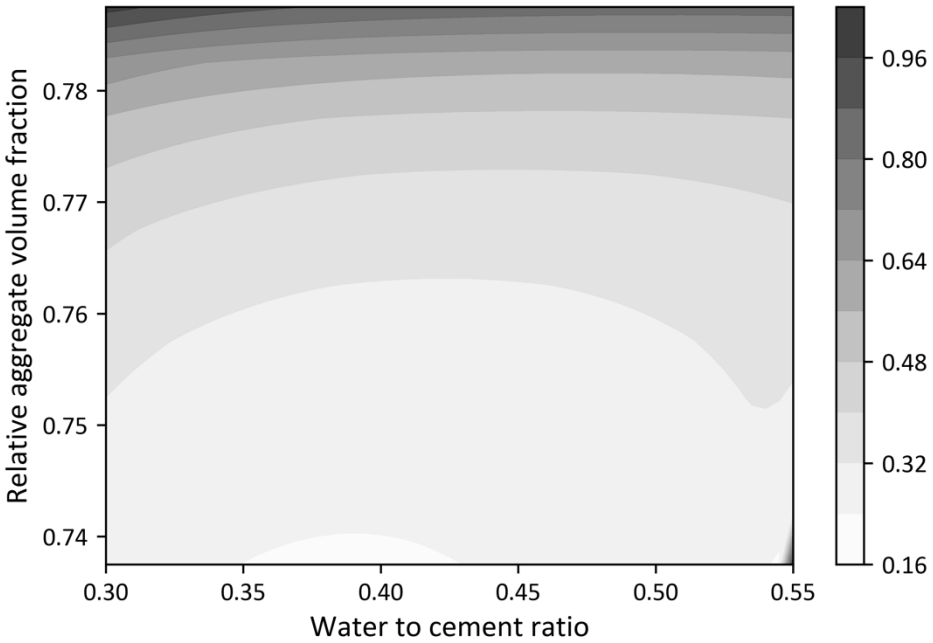

Figure 11. Overall robustness index as a function of W/C ratio and relative aggregates volume fraction for standard dosage variations in industrial production.

Table 1 Chemical composition of the cement.

Composition	%Weight
SiO2	21.81%
Al2O3	3.67%
Fe2O3	5.54%
CaO	64.10%
MgO	1.31%
SO3	2.41%
Cl	0.04%

Table 2 Aggregates properties.

Aggregates	Origin	Size range/mm	Specific density	Water absorption ratio/%	Maximum packing fraction/%
Sand	Palvadeau	0 – 4	2.64	0.6±0.05	68.3
Gravel	Seine	4 – 12	2.56	< 0.5	60.9

Table 3 Minimum required dispenser accuracy for concrete components in a ready-mix concrete plant from existing standards.

Components	ACI limits [28]	EHE limits [29]	Absolute variation (kg/m^3)
Cement	*1.0%*	*3.0%*	*3*
Sand	*2.0%*	*3.0%*	*16*
Gravel	*2.0%*	*3.0%*	*24*
Total aggregate content	-	-	*29*
Water	*3.0%*	*3.0%*	*6*
Admixture	*3.0%*	*5.0%*	*0.15*