Mixing of a wet granular medium: Effect of the particle size, the liquid and the granular compacity on the intensity consumption

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ABSTRACT

The mixing of a wetted insoluble granular medium was experimentally investigated focusing on the variation of the specific intensity consumption of mixing with addition of liquid in the granular bulk.

Preliminary results showed the influence of the particle average diameter of glass beads and calcites ($CaCO_3$), on the intensity consumption which decreased with the increase of the particles size (for the capillary and funicular states). However, an exception was observed at the level of the funicular state of the calcite, for which there was no particle size influence on the intensity consumption (for particles sizes inferior to 150 μm).

The effect of an inhomogeneous liquid distribution was not clearly demonstrated. However, the funicular intensity consumption was linked with the compacity of the medium.

By stopping the liquid flow at the end of the funicular state, and by continuing the mixing of a calcite, the funicular state intensity consumption and the measured granular compacities were increased. Moreover, this compacity measured during the funicular state, decreased with the particle size.

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1. Introduction

Mixing of wet granular media provides a homogeneous distribution of solids in the mixer and improves the wetting of the granular medium. However, it is a process that induces high energy consumption when the granular medium resists mixing (shearing).

In order to decrease the specific intensity consumption of mixing, a classification can be made of the factors (powders and liquid properties, process parameters) that have an effect on the inter-particular forces (capillary, Van-Der-Waals, gravitational, frictional, viscous forces, etc.) [1–3] and hence on the mixing efficiency.

The use of progressive addition, with a controlled flow, is worth studying because it leads to an increase in the time scale. The specific intensity consumption evolution with the added liquid amount (Fig. 1) characterise the mixing behaviour in different wetting states of the granular medium. The intensity profile may be divided in five sections [4–7]:

At the beginning of the mixing process (I), the first amount of liquid added is distributed at the surface of the particles without the formation of liquid bridges. Then, in the pendular state (II),

the liquid amount at the particle surface is high enough to form capillary bridges between them, while the medium becomes more cohesive and there is a rapid increase in intensity consumption. During the funicular state (III), the porous volume of the previously formed agglomerates is filled by the additional liquid. In this state, the increase in the intensity is much lower because the majority of the capillary bridges have already been formed in the pendular state. When the porosity in the agglomerates is completely filled by the liquid (end of III), the additional liquid forms new liquid bridges between agglomerates until the formation of a single agglomerate. This is the capillary state (IV), characterised by the appearance of a rapid increase in intensity consumption (capillary peak). Finally, in the droplet state, the volume of liquid becomes higher than the inter-particle porosity of the granular medium and the intensity consumption decreases.

In a granular medium wetted by water, the dominant force is the cohesive capillary force "F_C" [3]. This force depend on the particle diameter "d", on the liquid superficial tension "γ_L", and on a function that takes into account the contact angle "θ", the filling angle of the liquid bridge "β" (Fig. 2), and the ratio of the distance between particles with the particles average diameter "a/d" [2,8]:

$$F_C = \gamma_L d f\left(\theta, \beta, \frac{a}{d}\right) \tag{1}$$

A law can be used to take into account the cohesive forces "F_T" in the shear resistance of a granular medium. This is the tensile strength Rumpf's law [9] which is defined for a regular packing of

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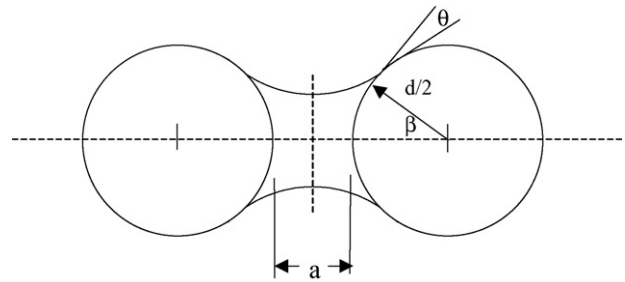
Fig. 2. Liquid bridge between two spheres.

particles and generate a capillary force limiting the relative motion of particles and giving high porosity. The increase of porosity is more pronounced for small than for large particles as a result of an increased capillary force relative to gravity. So, the inter-granular porosity is probably modified and must be taken into account during the mixing.

This paper presents firstly the influence of the particle average diameter on the intensity consumed during the mixing at the levels of the funicular and capillary state. "Two model granular materials were used in this study: glass beads (spherical, monomodal granular distribution, no reactivity with water) and calcites (non-spherical, monomodal granular distribution, no reactivity with water) allowing the observation of different behaviours during mixing."

The second part focuses on the different behaviours present in the funicular state.

monomodal spheres:

$$\sigma_T = \frac{(1-\varepsilon)}{\varepsilon} \frac{F_T}{d^2} \qquad (2)$$

The Eqs. (1) and (2) give the Rumpf's law of a granular medium which is proportional to the inverse of the average particle size [8]:

$$\sigma_{T,C} = \frac{(1-\varepsilon)}{\varepsilon} \frac{\gamma_L}{d} f\left(\theta, \beta, \frac{a}{d}\right) \qquad (3)$$

This law also takes into account the inter-particular porosity "ε" of the granular packing. Several authors have shown that addition of liquid in a dry granular medium modifies the compacity [10–14]. The porosity increases in the pendular state before remaining constant in the funicular state. Moreover, the porosity measured in the funicular state increases with the decrease of the particles size. Adding liquid will lead to the formation of liquid bridges between
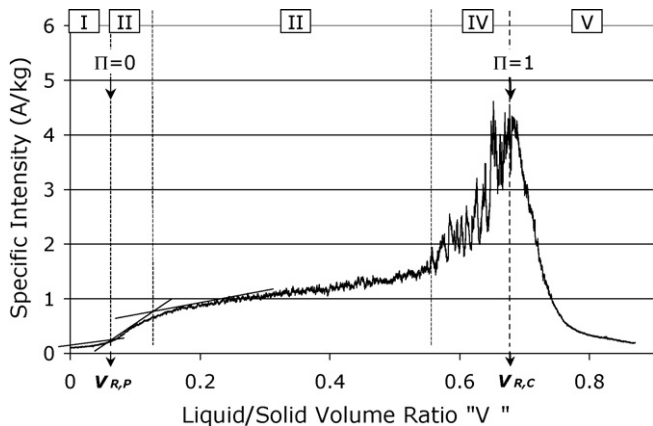

Fig. 1. Intensity consumption profile: mixing of the calcite 2 with a graduated liquid addition (40 ml/min).

2. Experimental set up and materials

The mixer used was a planetary mixer from the CAD® company (Mlx60). It can be used to make a hard paste, and it allows the intensity consumption of the engine to be recorded, at a frequency of 10 points per second. The available rotation velocity is between 0 and 300 rpm and the volume of the bowl is 4 L.

A preliminary study allowed the optimal process parameters to be set: an impeller speed of 120 rpm, a droplet-by-droplet liquid flow rate of 40 ml/min, and a powder volume "V_T" of 1250 cm^3 (This volume take into account the volume of solid "V_S" and the inter-particle porous volume "V_P"). The liquid was added using a peristaltic pump and a feeder pipe, 3 mm in diameter. Moreover, the necessary mass of powder "m_S" filling 1250 cm^3 was determined using the bulk density of the granular medium "ρ", equal to the average of the aerated "ρ_A" and tapped "ρ_T" densities:

$$\rho = \frac{m_S}{V_T} = \frac{m_S}{V_S + V_P} \qquad (4)$$

$$m_S = 1250\rho = 1250\frac{\rho_A + \rho_T}{2} \qquad (5)$$

Materials used (Table 1) were spherical glass beads supplied by SOVITEC® and calcites supplied by OMYA®. The volume granulometric distributions were obtained with a MALVERN® Mastersizer 2000, the true density "ρ_S" of the solids with a helium picnometer (Accupyc 1330-MICROMERITIC®), and the dry porosities "ε" (or the compacity "C") using the values of the true "ρ_S" and bulk "ρ" densities:

$$\rho_S = \frac{m_S}{V_S} \qquad (6)$$

$$\varepsilon = 1 - \frac{\rho}{\rho_S} = 1 - C \qquad (7)$$

The liquid used for all the tests was water.

Table 1
Material properties.

	$d10$ (μm)	$d50$ (μm)	$d90$ (μm)	True density ρ_S (g/cm^3)	Bulk density ρ (g/cm^3)	Porosity ε (%) (dry state)
Calcite 1	1	6	17	2.73	0.87	68
Calcite 2	2	18	47	2.73	1.13	58
Calcite 3	3	29	68	2.73	1.32	51
Calcite 4	58	90	140	2.73	1.36	50
Calcite 5	102	156	225	2.73	1.36	50
Calcite 6	140	220	317	2.73	1.39	49
Calcite 7	181	258	354	2.73	1.38	49
Calcite 8	316	524	803	2.72	1.45	47
Glass beads 1	18	30	48	2.48	1.44	42
Glass beads 2	64	88	122	2.51	1.49	41
Glass beads 3	117	160	219	2.52	1.49	41
Glass beads 4	194	261	353	2.52	1.46	42
Glass beads 5	391	538	738	2.52	1.49	41

3. Results and discussion

3.1. Particle size effect on the specific intensity consumption of mixing in funicular and capillary states

Several glass beads and calcites were mixed in order to study the solids properties effect on the granular medium behaviour during the mixing (shear resistance, wet inter-particular porosity). The measurements of the intensity consumed at the levels of funicular and capillary states are presented as a function of the inverse of the mean particles size (Fig. 3). This representation takes into account the Rumpf's law for which the tensile strength is proportional to the inverse of the particle size.

Firstly, the calcites generate higher intensity consumption than the glass beads. The different particle shapes of the two materials are responsible for this difference in mixing resistance. The calcites are less spherical than the glass beads and generate additional frictional forces that increase the shear resistance [15,16].

The intensity increases linearly with $1/d50$ for $d50 < 150$ μm. This result shows that the shear resistance (or the mixing resistance) is linked to the specific surface of particles. However, the y-intercept is not equal to zero. This difference with the Rumpf's law shows that the conditions for which this model is defined are not fulfilled. The mixing is achieved in a dynamic regime and the calcites are composed of irregular shapes non-monomodal particles whereas the Rumpf's law is defined in a static regime for spherical monomodal particles. On the other hand, this phenomenon is also caused by the consideration of the specific intensity consumption measured in the dry state, before liquid addition.

There is an exception for the funicular state of the calcites for which there is not a particle size effect (for $d50 < 150$ μm). To explain this stability of the intensity with particle size, our hypotheses are an inhomogeneously liquid distribution through the granular medium and/or an increase of the granular porosity when particle size decreases.

3.2. Independence of the funicular intensity consumption with the calcites particle size

To study the independence of the funicular intensity consumption with the calcites particle size, different approaches were used to characterise the liquid distribution and the granular compacity:

- Measurement of the liquid content present in several wet granular samples
- Tests carried out in order to increase the intensity consumption of the funicular state
  - Modification of the liquid flow and the number of liquid feeds
  - Mixing after stopping the liquid flow: observation of the intensity evolution and porosity measurements of granular samples removed, just after stopping the liquid flow and 20 min later
- Inter-particular porosity measurements of granular samples removed during the mixing, from the dry to the capillary state

3.2.1. Measurement of liquid content present in several wet granular samples

In order to measure the liquid content present in several wet samples, three calcites with a mean diameter higher or lower than 150 μm were used (calcite 3, 4 and 7; Table 1). The mixing (liquid flow of 40 ml/min, rotation velocity of 120 rpm) was stopped in the funicular state in order to take five wet granular samples ($\approx$10 cm^3) in different places of the bowl (in the middle or at the wall). With the masses of the wet and dried (24 h at 100 °C) samples, the liquid content present in every sample could be determined and compared with the liquid amount really added in the mixer. The results show that the liquid content added in the mixer and present in every sample are similar. So, at the scale of our samples, the liquid amount is localized homogeneously. However, even if an inhomogeneous liquid distribution is not observed at the scale of the samples, lack of homogeneity could be present at the scale of the agglomerates.

3.2.2. Tests carried out to increase the intensity consumption of the funicular state

The following tests were achieved in order to improve the liquid distribution and/or the granular compacity.

3.2.2.1. Modification of the liquid flow and the number of liquid feeds.
The number of liquid feeds was increased with keeping a constant liquid flow of 40 ml/min. The calcite 2, composed of particles lower than 150 μm, was mixed firstly with one liquid feed delivering 40 ml/min, and then with three liquid feeds spaced out 120°, each delivering 13.3 ml/min.

The results show that there is no effect of the number of liquid feeds. Indeed, the signals of the intensity consumption, obtained with one and three liquid feeds, are perfectly superposed. This test shows that the mixer seems to realize the same liquid distribution whatever the number of the liquid feeds used, and particularly for the smaller calcites like the calcite 2.

Liquid flows between 10 and 80 ml/min were achieved for the mixing of the calcite 1. The intensity consumptions obtained in the funicular state increase lightly with the liquid flow but this effect is too small to conclude that the liquid flow has an effect on the liquid distribution.

3.2.2.2. Mixing after stopping the liquid flow.
In order to study the liquid distribution and the granular compacity during the mixing, the liquid flow (40 ml/min) was stopped in the funicular state of the calcite 2 and the glass beads 1, and the mixing was maintained during 20 min. The intensity consumption evolution was observed,
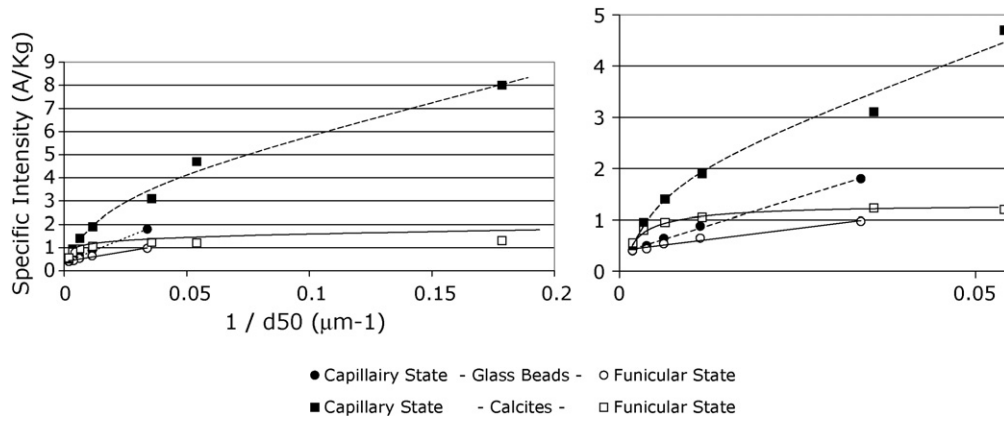
Fig. 3. Specific intensity consumption of mixing for the glass beads and the calcites as a function of the inverse of the mean particle size (1/d50).

after the stop of the liquid flow (Fig. 4). Tests were carried out with a stop of the liquid flow at the middle and at the end of the funicular state.

In addition, the porosity of several granular samples was measured just after stopping the liquid flow and after 20 min of mixing without flow (Table 2). Three different tubular punches (1570, 2260 and 9120 mm³) allowed making the samples, and for every punch volume ($V_{T,E}$), three samples were taken. Then, all the sample were dried in a drying oven (100 °C during 24 h) and the porosity "ε" was determined with the following equation:

$$\varepsilon = 1 - \frac{m_S}{V_{T,E}\rho_S} \tag{8}$$

For the flow stopped at the middle of the calcite funicular state, the intensity consumptions and the porosities determined after stopping the liquid flow and 20 min later are the same. In this case, stopping the liquid flow has no effect as there is not enough liquid in the system to allow a reorganization of the wet granular medium.

After stopping the liquid flow at the end of the calcite funicular state, the intensity increases (from 1.4 to 2.1 A kg⁻¹) and the porosity decreases (from 68% to 42%) with the mixing time. These results show that the liquid distribution in the calcite granular medium is not complete at the end of the funicular state, when the liquid is added progressively. So, an additional mixing time improves the liquid distribution, and consequently increases the intensity consumption and the compacity.

The intensity consumption and the compacity of the glass beads remain constant after stopping the liquid flow at the end or in the middle of funicular state. This result shows that glass beads do not

need more mixing time to distribute the liquid and to reach the maximal compacity, contrary to the calcite.

These first results confirm our hypotheses explaining the independence of the funicular intensity consumption with the calcites particle size. In the funicular regime, the liquid distribution seems to be incomplete. Furthermore, the inter-particular porosity is very important in this state and decrease with the mixing time after stopping the liquid flow.

In the next part, it will be shown that the inter-particular porosity in the funicular state increases when the particle diameter decreases.

3.2.3. Inter-particular porosity of granular samples removed during the mixing

Using the method described in Section 3.2.2.2, measurements of porosity were achieved for several glass beads and calcites granular samples. These samples were taken during the mixing for different liquid saturation rates, from the dry state to the capillary state. In order to calculate corresponding amounts of liquid for the different solids, we used the dimensionless amount of liquid Π proposed by Leuenberger et al. [17]:

$$\Pi = \frac{V_R - V_{R,P}}{V_{R,C} - V_{R,P}} \tag{9}$$

"V_R" is the liquid/solid volume ratio, "$V_{R,P}$" is situated at the beginning of the pendular state, and "$V_{R,C}$" correspond to the capillary state (Fig. 1).

Figs. 5 and 6 show that the two materials present similar porosity evolutions. From the dry ($\Pi = 0$) to the wet state ($\Pi = 0.1$), the
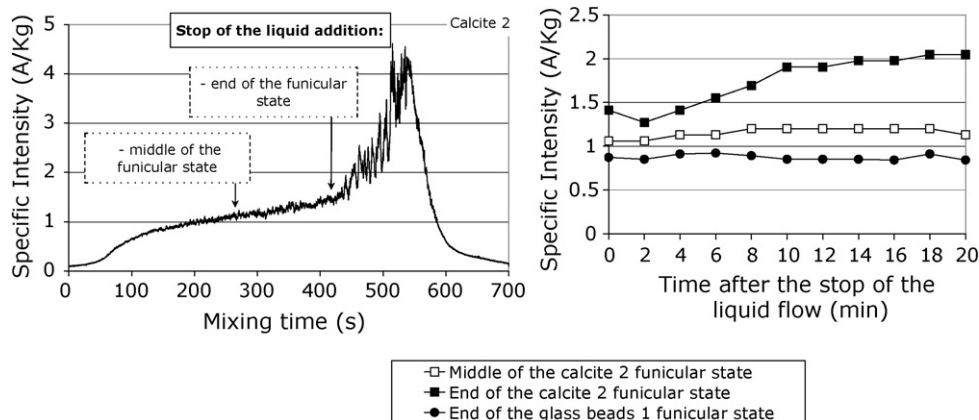

Fig. 4. Evolution of the specific intensity consumption of mixing after stopping the liquid flow in the funicular state.

Table 2
Inter-granular porosities of granular samples taken after having stopped the liquid flow and after 20 min of mixing without flow.

	Dry initial state	Middle of the funicular state		End of the funicular state	
		Time after stopping the liquid flow (min)			
		0	20	0	20
	1	2	3	4	5
Porosity of calcite 2 (%)	59	75 ± 3	74 ± 2	68 ± 4	42 ± 3
Porosity of glass beads 1 (%)	42	59 ± 6	–	54 ± 3	54 ± 4
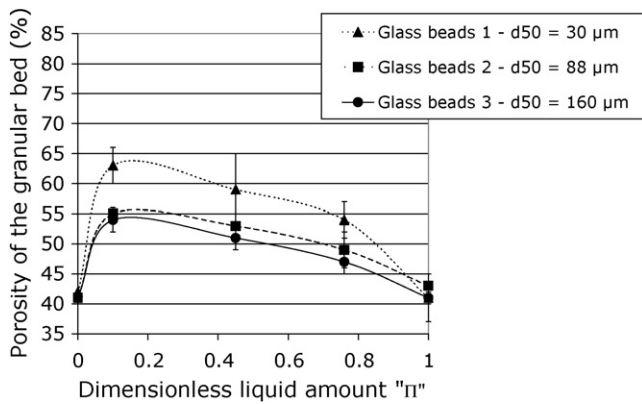
Fig. 5. Evolution of the porosity of glass beads during mixing.

porosity increases (in the pendular state) before remaining constant in the funicular state ($0.1 < \Pi < 0.75$). Then, in the capillary state ($\Pi > 0.75$), it decreases. Moreover, the porosity determined in the funicular state increases with the decrease of the particle size. These observations are in agreement with previous works [10–14] described in Section 1.

However, some differences are observed between the solids. For the bigger particles (glass beads, calcites 4 and 5), the initial porosity (dry state) is the same as the final porosity (capillary state). In this case, the porosities of the calcites are higher than the porosities of the glass beads because of the calcites are less spherical [18]. For the smaller particles (calcite 1–3), the final porosity is lower than the initial porosity. Under a critical particle size, the dry porosity increases with a decrease of the particle size because of the effect of the Van-Der-Waals cohesive forces: a restriction of the relative motion of particles [10–13].

In the funicular state, the porosity of the calcites increases with the decrease of the particle size, to attain values between 70% and 80% for the smaller sizes (under 30 μm). This induces a decrease of the density of contact points and of the frictional and capillary cohesive forces (in a wet state, i.e. pendulary, funicular or capillary
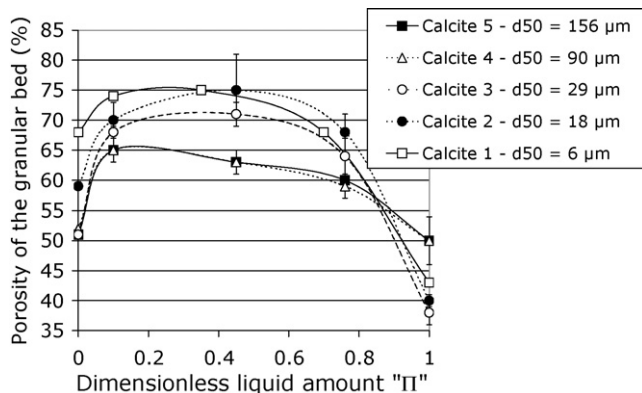
state, the Van-Der-Waals cohesive forces are negligible compared to capillary forces).

4. Conclusion

The mixing of two different wetted insoluble granular media (non-spherical calcite and glass beads) was experimentally studied focusing on the variation of the specific intensity consumption of mixing with addition of liquid in the granular bulk.

According to the Rumpf's law, we firstly showed that the specific intensity consumption is function of the inverse of the particles size (for the capillary and funicular states). However, an exception was observed at the level of the funicular state of the calcite, for which there is no particle size influence on the specific intensity consumption.

By stopping the liquid flow at the end of the funicular state, and by continuing the mixing of a calcite, the specific intensity consumption and the granular compacities were increased. On the contrary, the intensity and the compacity of glass beads remained constant after the stop of the liquid flow. These phenomena explain partly the stability of the calcite funicular intensity when particle size decreases. The liquid distribution and the compacity are incomplete in the funicular state and need more mixing time to be improved.

Moreover, measurements of calcites granular porosities during the mixing (from the dry to the capillary state) showed that the porosity of the funicular state increases until 70–80% with the decrease of the particles size. So, the space between particles is increased and the inter-particle forces (capillary and frictional) are decreased consequently. This phenomenon allows keeping the specific intensity consumption in the funicular state constant.

Otherwise, these measurements during the mixing show that the porosity is not constant from the dry to the capillary state. A dilation of the granular bed takes place in the pendular state, and a densification occurs in the capillary state. So, the compacity depends on the added liquid amount, influences the shear resistance (and consequently the specific intensity consumed by the mixer), and is an important parameter to take into account in the study of the granular medium behaviour during mixing.

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Fig. 6. Evolution of the porosity of calcites during mixing.

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