



**HAL**  
open science

## Assessing the homogeneity of powder mixtures by on-line electrical capacitance

N Ehrhardt, A Montagne, Henri Berthiaux, B Dalloz-Dubrujeaud, Cendrine  
Gatumel

► **To cite this version:**

N Ehrhardt, A Montagne, Henri Berthiaux, B Dalloz-Dubrujeaud, Cendrine Gatumel. Assessing the homogeneity of powder mixtures by on-line electrical capacitance. *Chemical Engineering and Processing: Process Intensification*, 2005, 44 (2), pp.303-313. 10.1016/j.cep.2004.03.016 . hal-01649523

**HAL Id: hal-01649523**

**<https://hal.science/hal-01649523>**

Submitted on 7 Nov 2019

**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.

# Assessing the homogeneity of powder mixtures by on-line electrical capacitance

N. Ehrhardt<sup>a</sup>, M. Montagne<sup>a</sup>, H. Berthiaux<sup>a,\*</sup>, B. Dalloz-Dubrujeaud<sup>b</sup>, C. Gatumel<sup>a</sup>

<sup>a</sup> *Laboratoire de Génie des Procédés des Solides Divisés, UMR CNRS 2392, Ecole des Mines d'Albi-Carmaux, Campus Jarlard, route de Teillet, 81 000 ALBI, France*

<sup>b</sup> *Département de Mécanique Energétique, UMR CNRS 6595, Ecole Polytechnique Universitaire de Marseille, Technopôle de Château-Gombert, 5, rue Enrico Fermi, 13 453 Marseille, France*

---

## Abstract

In this paper, we try to improve our comprehension of one of the most important challenge in the particulate solids industry: the assessment of the homogeneity of powder mixtures. A two-electrode electrical capacitance method is used to measure the permittivity of mixtures while flowing in a 1-D set up. An emphasis is placed on the possibility of following homogeneity evolution through permittivity measurement, without the necessity to calculate the volumetric proportions of each component from an effective medium formulation. The methodology is applied to examine segregation in a funnel, as well as the efficiency of a laboratory drum mixer.

*Keywords:* Homogeneity; Capacitance; Powder mixing

---

## 1. Introduction

With the increased variety of formulated products of the particle form that appears every day on the market and that are issued for a wide range from industry (pharmaceutical, agro-food, cement, plastics, . . .), particulate processes in general, and mixing processes in particular, are pushed to a never-ending battle against their own performances. This is due to the fact that the end-used properties of the products that are manufactured nowadays are more and more complex, especially because these must cover more and more functions. Because product engineering had become a major engineering science producing its own tools both at the research and the educational levels, a product ought to become an “intelligent delivery system”, a “small process” able to optimise its own action when it is used.

However, it is of common knowledge that mixers, as much other equipment dealing with particles, are all designed and operated from the purest empiricism. Indeed, there exists an

important mismatch between the refinement attained at the formulation step and the development of processes able to manufacture the designed and desired product. If we take biodisponibility of a pharmaceutical tablet as an example, the trust that we have in formulation must be contrasted with the following “traditional chemical engineering” interrogation: “How to make sure that the process will deliver pills, all having the same biodisponibility?”. In practice, this difficulty in controlling the product has led the companies and some regulatory institutions, such as the FDA, to emphasise on “freezing the process” by avoiding any change. In many process industries, the main keyword concerning powder or granular material mixers is undoubtedly “reproducibility”, with the meaning that once an equipment is qualified and validated to perform a certain function, operating conditions are fixed and the cost of any variation in the process parameters may become prohibitive [1]. For example, in the pharmaceutical field, even a slight change in the mixing process would lead to clinical trials for ensuring that the drug’s therapeutic effect has not been affected. In addition, current sampling procedures are widely contested [2–4], especially because of the limited number of samples – typically thirty – that can be assumed in the context of production with

---

\* Corresponding author. Fax: +33 5 63 49 30 25.

*E-mail address:* berthiau@enstima.fr (H. Berthiaux).

respect to the overall number of possible samples – about one million.

In other words, the technological improvement of the products does not guarantee the same progress at the manufacturing level at all, mostly because of a lack of instrumentation and control of the processes, more deeply linked to the lack of understanding and reliability of particle characterisation in general. If we except the mineral industry, the industrial applications of particle control systems are still scarce, the only well-known example being particle size distribution measurement through LASER diffraction. However, the situation is not so dramatic if we look at the progress made in the field of particulate instrumentation research in the last decade, with a constant special insight provided by the international conference series “Control of Particulate Processes” [5].

Optical techniques were developed – both in situ and on line – over a broad range of applications and various types of light sources. Main examples are solids concentration measurements in particle fluidisation [6,7], as well as gas–solid flow [8], or sample composition in dry blending operations, this latter beginning from the pioneering work of Harwood et al. [9] up to more recent advances in this scientific field [10–12]. The main interest of these techniques lays in the fact that optical probes are easily available on the market, so that qualitative monitoring of mixers is possible at an industrial scale. However, these probes only provide local information of the mixture (typically 1/40 of a tablet), so that in essence, they still “sample” the powder flow in a way that may be intrusive and not always representative of the overall stream.

Electrical capacitance tomography (ECT), as well as electrical resistance tomography (ERT) for liquid–solid systems, experienced a flourishing development these recent years for a wide variety of applications [13–18]. Their major advantage is that they provide, in a non-intrusive way, a cross-sectional view of a stream that may contain at the same time a liquid, a solid and a gas. In that sense, they are able to give a radial image, which in turn, and through the help of other multiple electrode sensors in the axial direction, can be reconstructed in three dimensions. However, this huge amount of information must be contrasted with the low resolution at which it is given. This has probably limited the application of this technique to powder mixtures that require an analysis at a very fine scale. X-ray,  $\gamma$ -ray, as well as magnetic resonance imaging (MRI), must also be cited in the category of the tomographic techniques. They were mostly used to investigate fluidisation processes, but still seem difficult to employ at the level of a factory [19–21]. LASER-induced fluorescence (LIF) technique must finally be cited, especially because some pharmaceutical ingredients can naturally provide a signal when excited by the adequate beam [22].

In this work, we develop an electrical capacitance sensor that consists of only two electrodes placed around a glass tube. This set up is considered not in the view of imaging

the media (such as in classical ECT methods), but with the objective of obtaining a global dielectric information and follow its variation while powder is flowing into the tube, with the aim to measure the composition of binary or ternary mixtures in the axial direction of the powder flow stream. It is waited that this method will provide, through specific iterative algorithms, a quantitative and structural information about a binary mixture flowing out of a mixer (either batch or continuous) in relation with its own processing.

## 2. Theoretical and conceptual developments

### 2.1. Powder blend homogeneity: macro and micro considerations

Homogeneity is a key concept for many chemical engineering applications, especially those in which a transformation occur (precipitation, chemical reaction, grinding, . . . ), because of the coupling between kinetics aspects and contact time of the “reagents” due to the flow conditions. In the field of powder mixing, in most cases, no transformation takes place and the quality of the product is the fact of the particulate flow dynamics itself. Also, the main difference with fluid mixing lays in the fact that a perfect mix cannot be performed at a molecular scale, and is still difficult to assess at the scale of a particle. This is due to a wide variety of particle–particle interactions, resulting in a conflict between mobility of single particles, which is essential to achieve a good mix, and resistance to segregation, which is a fact of the mixture’s structure. In that sense, cohesive particulate media (typically below 50  $\mu\text{m}$ ) can be viewed as “macrofluids”, while free-flowing systems (typically superior to 500  $\mu\text{m}$ ) can be viewed as “microfluids”. Between these two extreme behaviours – that sometimes may occur in a single product (case of aerable powders) – particle and particle packets trajectories and motion are both influenced by gravity and surface forces.

As felt above, the concept of powder blend homogeneity cannot be dissociated from the scale at which a mixture is observed. For continuous mixers, homogeneity must be defined at the level of the blend flowing out of the vessel, and falling down to a conveyor or any other transportation equipment. In the case of a batch mixer, the mixture structure can be particularly complex to define, but in practice and when the mixer is emptied, the content of the vessel is discharged through a “pipe” to a container or a tableting machine. Hence, in both cases and if we take into account the entire cross-section of the pipe, the mixture flow can be assimilated as a 1-D particle stream.

Thus, let us consider a powder mixture flow as a mono-dimensional layer divided into  $N$  consecutive “elementary” samples. In a first approach, the size of these can be arbitrary fixed, not too small to prevent from statistical errors (inversely proportional to the square root of the particle number) and not too large for ensuring the validity

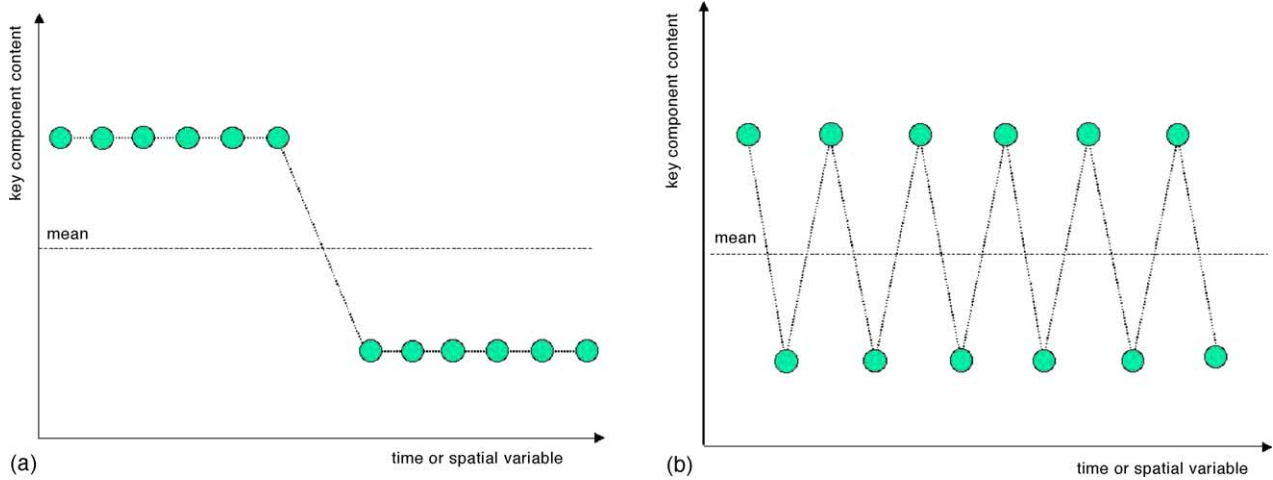


Fig. 1. Two mixtures (a and b) of identical macromixing state can exhibit different micromixing structures that may result in products of different quality and effects.

of the following analysis. From a global point of view, the distribution of the compositions is a first indicator of the mixture's homogeneity, a perfect blend (at the scale of scrutiny of the sample's size) corresponding to a peak centred on the mean value. Hence, the following standard deviation (Eq. (1)) can be considered as a good macromixing index (the coefficient of variation can also be employed).

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (x_i - \mu)^2}{N}} \quad (1)$$

This latter formula, which is commonly used in the industrial practice, also holds for the case of an estimation of the mixture quality using random sampling. However, the main problem in this global analysis lays in the well-known (since the famous Danckwert's example in chemical reactors [23]) fact that two different structures can correspond to the same intensity of segregation criteria. Fig. 1 illustrates this idea, which has a very practical sense in the context of pharmaceutical blending.

In effect, let us assume that the two processes depicted here concern pharmaceutical tablets produced in packages containing six consecutive unit doses. These are of identical macromixing state, as standard deviations are equal. But in one case (namely Fig. 1a), a patient will take a slightly under-dosed treatment, and another one a slightly overdosed treatment, while in the second possibility (Fig. 1b), an equilibrium of the dose will be reached after the second take. Indeed, structure defaults can provoke negative therapeutic effects that cannot be diagnosed with a macroscopic index of segregation intensity.

As introduced by Danckwerts in the mid-fifties [24,25], and more recently reviewed by Gyenis [26], the mixing structure can be accounted for by the use of the autocorrelation function  $R(r)$  (Eq. (2)). The distance  $r_0$ , corresponding to the equation  $R(r) = 0$ , at which two sample compositions can be considered to be uncorrelated, is a main characteristic of the structure of the mixture and has an important

practical consequence: tablets should probably be produced at this – segregation – scale to statistically ensure that these have the right (desired) composition. However, the figure given here is still global, as we do not know anything about the location and the distribution of the segregated zones in the mixer, and also on zones of acceptable defaults. As a consequence, the size of the tablets that corresponds to the segregation scale may be not so adequate as it is said to be.

$$R(r) = \sqrt{\frac{\sum_{i=1}^{N-r} (x_i - \mu)(x_{i+r} - \mu)}{\sum_{i=1}^N (x_i - \mu)^2}} \quad (2)$$

An illustrating example for this idea is provided in Fig. 2, in which a series of consecutive elementary samples containing an active ingredient has been artificially generated. One may easily find that the mean content is  $\mu = 5\%$ , while the global standard deviation is  $\sigma = 1.34\%$ . If we refer to a pharmaceutical standard, this is not a good mixing, being the corresponding coefficient of variation equal to 26%. As it can also be seen from the data, an important deviation from the mean occurs for sample nos. 11 and 12, which have the particularity of being corrected at a small scale (sample nos. 13 and 14). From the point of view of monitoring of the mixing process, it is certainly interesting to search for this local default that makes the overall mixture probably inadequate, despite of the fact that other regions seem to be much better mixed.

If we look at the variations of the corresponding autocorrelation function (Fig. 3), the characteristic "distance"  $r_0$  covers between 1 and 2 elementary samples, which means that these must be grouped two by two or three by three (for more security) to avoid any correlation. Doing so, six or ten tablets can be defined depending on the way the elementary ones were grouped together (see also Fig. 2). However, this procedure did not really change the situation, as it gave rise to an overdosed (no. 4) and an under-dosed tablet (no. 5) and four perfectly dosed tablets (nos. 1–3 and 6). This is due to

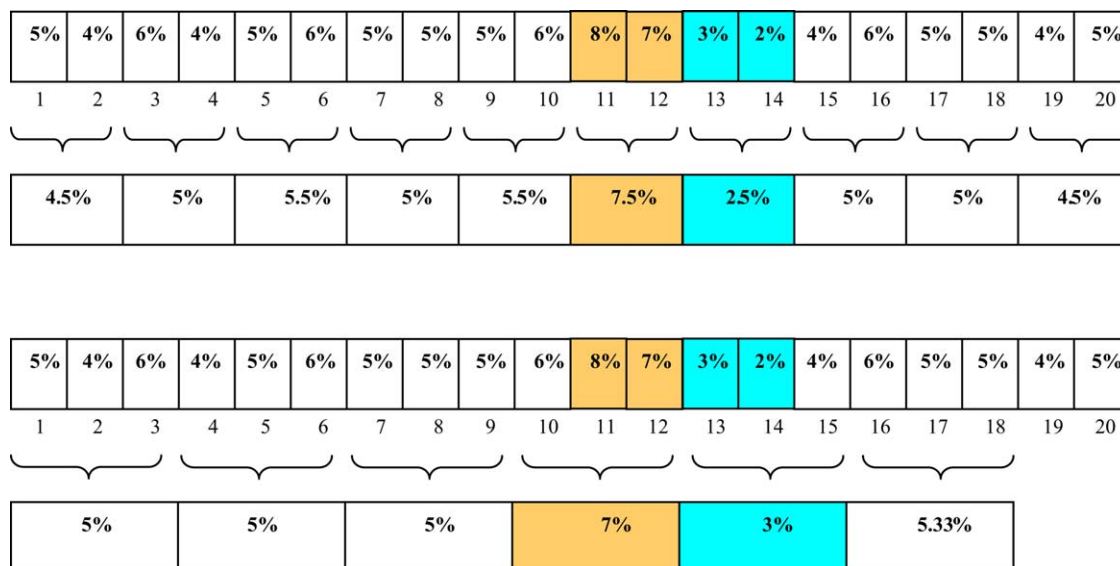


Fig. 2. Numerical example showing that a non-randomised structural default (heterogeneous zone between sample nos. 11–14) cannot be cleared by increasing sample size either by a factor 2 (top case) or 3 (bottom case).

the fact that this structural default induces segregation at a relatively short scale, and cannot be adequately detected by the criteria  $r_0$ . The reason for this failure is fundamentally linked to the global (but structural) character of this criterion that characterises homogeneity at the scale of all the  $N$  samples. In other words, this analysis would give a better result if the “problematic” elementary samples were not placed coast to coast in the mixture, but dispatched along the whole data set.

## 2.2. Methodology for detecting structural defaults

Because of the above-mentioned type of problem, it is advisable to have a better insight on the local structure of the

mixture, with the aim of disregarding samples of inadequate composition rather than grouping them. For this reason, we derived another type of analysis, which we claim to be more sensible to structural defaults and also easier to adapt to on-line measurement of continuous processes in which the series of data is, in principle, infinite. Let us first define a “window” that consists of  $n$  consecutive elementary samples (of course  $n < N$ ) placed at the beginning of the data series. For this packet of samples, a mean  $\mu_{1,n}$  and a standard deviation  $\sigma_{1,n}$  can be defined by the following Eqs. (3) and (4).

$$\mu_{1,n} = \frac{1}{n} \sum_{i=1}^n x_i \quad (3)$$

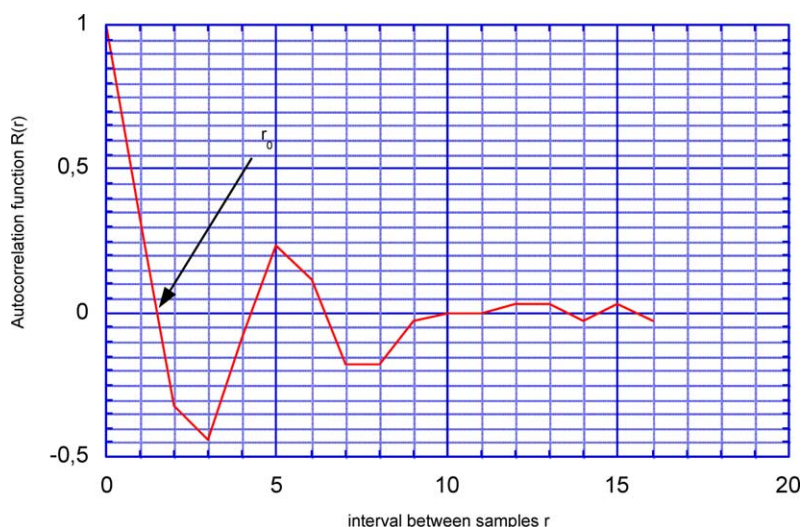


Fig. 3. Autocorrelogram corresponding to the present example, and indicating that a characteristic interval of two samples should be considered for ensuring homogeneity.



$$\sigma_{1,n} = \sqrt{\frac{\sum_{i=1}^n (x_i - \mu_{1,n})^2}{n}} \quad (4)$$

Let us further displace this window from one sample to the right (of, say, Fig. 2), defining thus another mean  $\mu_{2,n}$  and another standard deviation  $\sigma_{2,n}$ . If the composition of the newly considered sample (number  $n + 1$ ) is close to  $x_1$ , then both  $\mu_{2,n}$  and  $\sigma_{2,n}$  will be close to  $\mu_{1,n}$  and  $\sigma_{1,n}$ , respectively. In the contrary, if a small structural default exists, it will be detected by this analysis because the variation of the corresponding mean and standard deviation will be important, even if it concerns segregation at a small scale. Generalisation of Eqs. (3) and (4) for packets beginning with sample number  $k$  is obviously given by Eqs. (5) and (6).

$$\mu_{k,n} = \frac{1}{n} \sum_{i=k}^{n+k-1} x_i \quad (5)$$

$$\sigma_{k,n} = \sqrt{\frac{\sum_{i=k}^{n+k-1} (x_i - \mu_{k,n})^2}{n}} \quad (6)$$

For the above example, one can observe that the seventh packet first considers the small segregated region, while it is not touched after the fourteenth if the size of the window is equal to five samples. This results in a clearly marked peak on a graph representing the variation of these local variances with time, while the means are affected at a much lower level (see Fig. 4).

The reason for this lays in the fact that even a small deviation in composition has a consequence on the standard deviation value, so that the local standard deviations are as great as many samples in the packet demonstrate a deviation from the mean. In our case, it can be seen that the number  $n_p$  of packets that are affected by the perturbation is equal to 8 out of 16, which also helps in magnifying the default. For instance, it is felt that a smoothed local standard deviation

curve is the consequence of a good mixture achieved at a small scale, while a “rough” one corresponds to the existence of heterogeneous regions in the blend.

From the graph, it is also possible to calculate the number of samples  $n_0$  that form part of the default, and should probably be taken away from the production line. In effect, it is easy to demonstrate Eq. (7). In the present example, as  $n_p = 8$  and  $n = 5$ , one will verify that the heterogeneous zone effectively covers four consecutive elementary samples.

$$n_0 = n_p - n + 1 \quad (7)$$

### 3. Experimental set up development and methodologies

Like in any micromixing state consideration, the experimental illustration of the concept needs the development of a sensor. Because even slight structural defaults must be detected, it can be felt that this technique may cover the following properties:

- (1) Be adapted on-line: The three dimensional knowledge of the repartition of the components in a vessel (as in a batch mixer) is generally of a few interest for the practitioner, especially because an important disruption of the mixture occurs when emptying the mixer.
- (2) Be non-intrusive: This is the better way to avoid any “cutting” error due to the insertion of a probe in the powder flow.
- (3) Be exhaustive: As all defaults are to be screened, no partial sampling procedures that would induce a statistical bias can be developed. For instance, the derivation of a certain part of the powder flow for analysis cannot be considered here.
- (4) Have a good spatial and temporal resolution.

On the other hand, as only the global content in the sample is to be derived, imaging and tomographic techniques

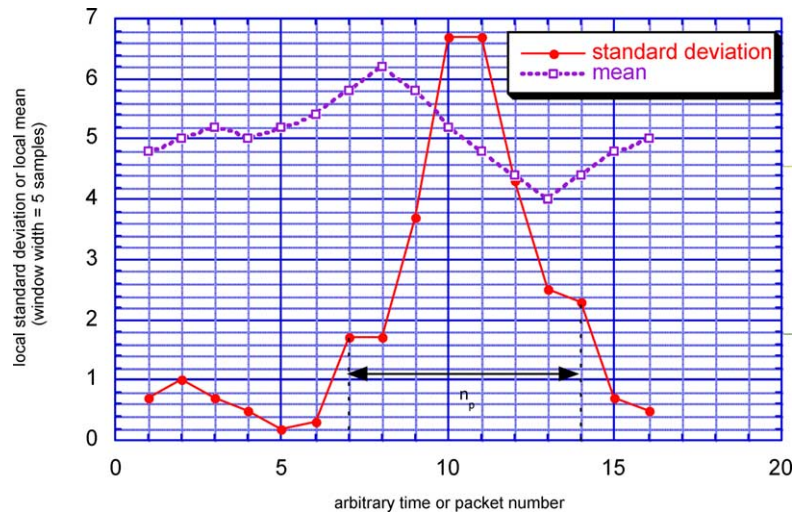


Fig. 4. Evolution of the local means and standard deviations with the number of packet considered (packet width equal to five elementary samples), magnifying the existence and the location of the heterogeneous zone.

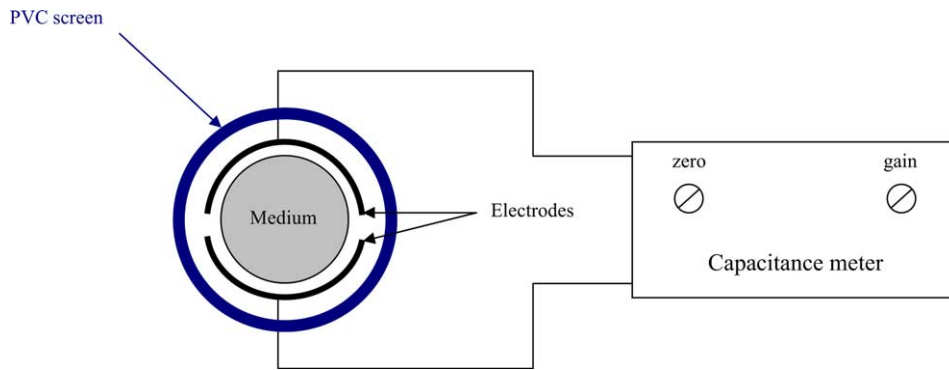


Fig. 5. Capacitance sensor developed.

that also provide spatial distributions are perhaps too powerful tools to be employed with respect to the information effectively needed. From the above-mentioned techniques, it can be understood that the best method profile should combine the advantages of fibre-optics (flexibility, global information) and ECT (non-intrusive, no sampling) sensors.

### 3.1. Electrical capacitance set up

Classical ECT set ups are composed by a ring of electrodes (typically 12, see [27,28]), which is generally placed around the external part of the pipe itself. The correlation between the dielectric measurements provided by the electrodes allows for a tomographic mapping of the pipe in the radial direction. In the present case and as commented above, the radial distribution of the components is of a few interest (at least in a first approximation) as we are only concerned with the overall composition in the measuring volume. For this reason, we developed a capacitance sensor with the aim of measuring the variations of the global capacity of a mixture during its own flow. It thus consists of only two copper electrodes covering practically the whole

section of a glass tube of external diameter 12 mm (see Fig. 5). A PVC screen allows isolating the electrodes from the aluminium full cylinder (of dimensions 40 mm × 30 mm) that covers the overall measuring cell. This latter also prevents from external disturbances, as well as the coaxial cables, making the liaison with the capacitance meter.

The sensor is connected to a capacitance meter consisting of the electronic circuit depicted in Fig. 6, and further to a computer through an analogic/numeric converter. Measurements are taken at a frequency of 5000 scans per second and the sensibility of the circuit is high enough (close to 0.1 pF) to detect the capacity variations of some picofarads provoked by changes in the composition of the measured volume. Preliminary results also show that capacity and dielectric permittivity could be linked to each other through a linear relation over the whole measuring range in which we work. It must be stated that such a statement is only true if the measured space between electrodes is fully filled with material. Otherwise, the measurements would show very strong non-linear character.

The calibration of the sensors is performed with a liquid (namely *n*-butyl acetate) of known dielectric permittivity,

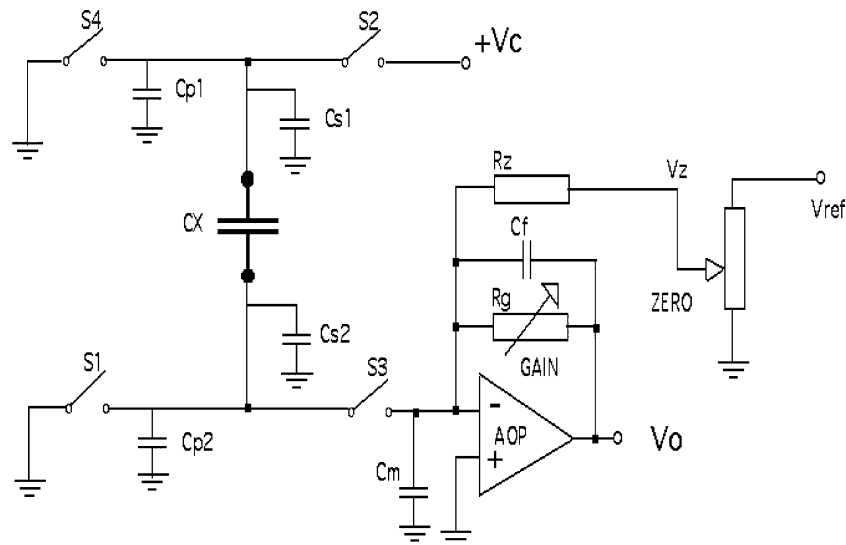


Fig. 6. Sketch of the electronic circuit for capacitance measurements.

and is repeated every week. Three sensors of exactly the same characteristics were constructed and can be linked to the capacitance meter at the same time. As capacitance is strongly influenced by temperature and humidity, a cooling air equipment is also placed near the sensors, ensuring then a temperature comprised between 15 and 16 °C. In the same time, humidity of the laboratory is controlled to be sure it is rigorously the same for each experimentation. Each sensor proceeds to measurements on samples whose volume is approximately 2.3 cm<sup>3</sup> and the electrode area is approximately 0.38 cm<sup>2</sup>.

### 3.2. The effective medium theory

The effective medium theory consists in considering the real medium, which is quite complex, as a fictitious model medium (the effective medium) of identical properties. Bruggeman [29] had proposed a relation linking the dielectric permittivity of the medium to the volumetric proportions of each component of the medium, including the air through the porosity of the powder mixture. This formula has been rearranged under a symmetrical form by Landauer (see Eq. (8)), where  $\epsilon_i$  is the permittivity of powder  $i$  at a dense state,  $\epsilon_m$  is the permittivity of the mixture and  $P_i$  the volumetric proportion of powder  $i$  and cited by Guillot [30] as one of the most powerful model.

$$\sum_{i=1}^{N_c} \frac{\epsilon_i - \epsilon_m}{\epsilon_i + 2\epsilon_m} P_i = 0 \quad (8)$$

### 3.3. Particulate system considered and characterisation

Because the objective of the study is to calculate the proportion of a definite component in a binary or in a ternary mixture from the data, the particulate products to be chosen may first demonstrate a certain permittivity contrast at a “dense state”. It must be stated that in the case of a particulate material, this does not correspond to the dielectric constant of a massive block of the material for which electric contacts between grains do not exist. For instance, the “dense state” corresponds to a hypothetical block of material having the same quality of dielectric contact as a granular material for which porosity is occupied by vacuum.

The method used in determining the dense dielectric permittivity of a definite material consists in defining a fluid in which the material cannot dissolve and further to measure the effective permittivity of various particulate media/fluid mixtures corresponding to various porosity. The interception point of the curves representing the variation of the permittivity of the material with porosity (through the effective medium equation, see section 3.2) gives the “dense state” permittivity.

It is clear that this method does not work in the case of a particulate medium/fluid mixture that saturates the sensor. In this way, we developed a second method that consists in filling the sensor with powder, measuring the blend

Table 1  
Some characteristics of the products used

Products	True density	Dense state relative permittivity	$d_{50}$ ( $\mu\text{m}$ )
Salt	2.21	5.43	565
Silicon carbide	3.15	44.44	565

permittivity, weighting the powder in order to isolate the porosity and, thanks to the effective medium equation (see Eq. (8)), to deduce the dense permittivity of the powder. This method is less precise than the previous one, but we proceeded to several measurements and we averaged them, thus obtaining better results.

In this study, commercial sugar, silicon carbide and powder that enter in the composition of cement (A–C) were finally chosen for their high permittivity difference. Sugar and SiC were both sieved between 500  $\mu\text{m}$  and 630  $\mu\text{m}$  in order to avoid segregation by particle size. Another advantage is that silicon carbide particles can be separated from sugar in the blends by water dissolution of the latter, filtration of SiC and finally drying. Table 1 gives the physical properties of these powders. Note that true densities were measured by a classical pycnometric method.

### 3.4. Experimental rig and mixers

The mixture to be analysed is placed inside vertical walls in a vibrating channel operating at low amplitude to avoid

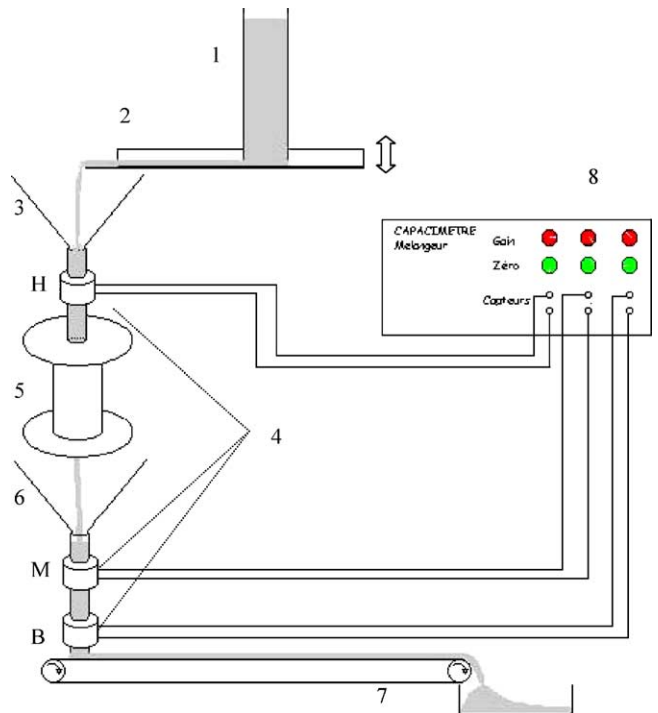


Fig. 7. Sketch of the experimental rig showing the initial mixture (1), the vibrating channel (2), the upper funnel (3), the sensors (4), the static mixer (5), the lower funnel (6), the belt conveyor (7) and the capacimeter (8).



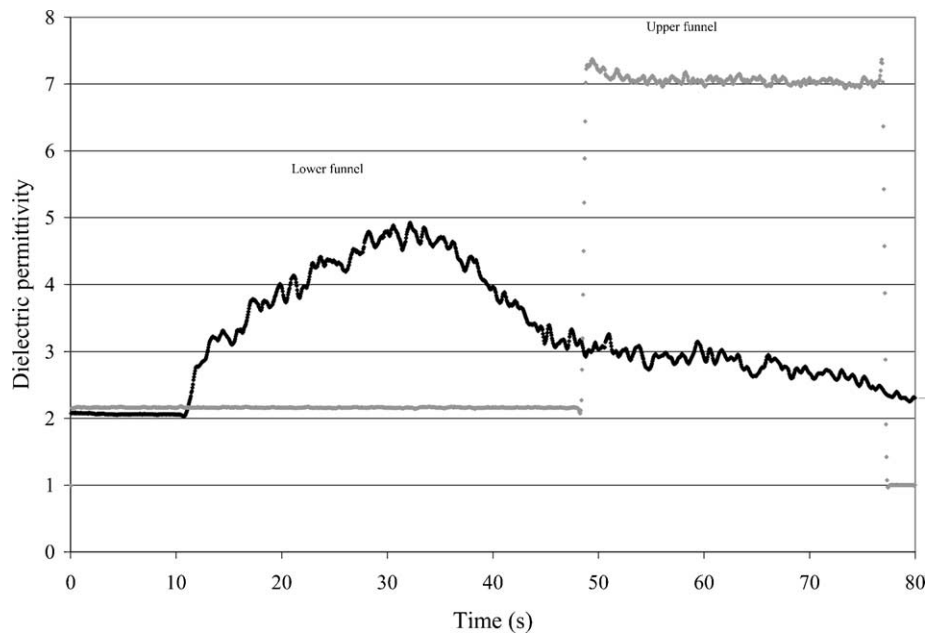


Fig. 8. View of the discharge profiles through a funnel.

particle segregation during transport, as well as ensuring mass flow of the blend. The particles are then falling at the centre of a funnel (without accumulation) that is connected to the glass tube surrounded by a first capacitive sensor (denoted sensor H). At the end of the tube, a Sulzer® static mixer consisting of six mixing elements is placed. These mainly allow for a radial motion of the particles and few axial mixing occurs in normal flow regime (see [31]). The mixture further flows by gravity to another funnel placed in the upper part of another glass tube that supports the last two sensors separated by a known distance (see Fig. 7). Assuming mass flow in this last section, the correlation between the responses of these sensors (denoted M and B) allows for the calculation of the flow rate and the frequency to consider avoiding any sample crossing.

For instance, the glass tube must always be full of solids with a constant flow. To ensure this, the end of each tube was squeezed and the bottom of these tubes were placed at a short distance from a conveyor belt operating at constant speed ( $5 \text{ m min}^{-1}$ ), thus gently depositing the mixture as a layer onto the belt. This set up also allows a validation of the method by comparing the sample compositions in the belt to the compositions given by the closest sensor. With this special set up, all particle flow rates become quickly equal from the vibrating channel to the conveyor belt (in fact, the amplitude of the vibrations can be adjusted for this task).

## 4. Results and discussion

### 4.1. Following the discharge profile through a funnel

In this experiment, we studied the effect of a funnel on the discharge profile of two different and successive layers

(sugar at the bottom and SiC at the top). This preparation flowed through the glass tube equipped by the first sensor and then through a lower funnel equipped by the two other sensors at its outlet. The discharge profiles are represented in Fig. 8.

The profile in the upper funnel clearly shows the step of permittivity applied to the system, whereas the profile after the lower funnel is clearly spread. This shows us that the

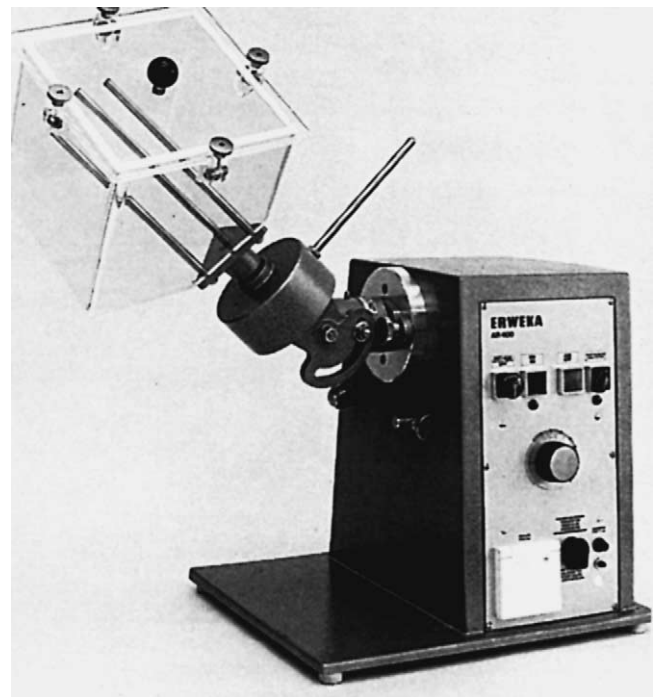


Fig. 9. Photograph of the Erweka Rotocube® mixer.

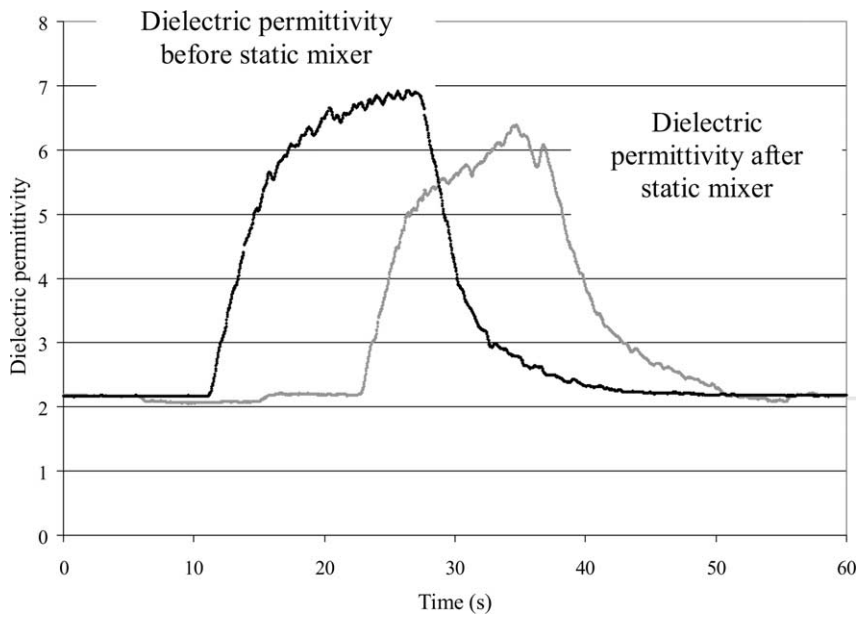


Fig. 10. View of the initial discharge profiles before and after the static mixer.

SiC is mixed with sugar at the beginning of the experiment because it flows preferentially in the middle of the funnel, whereas components situated near the walls are flowing at the end.

We can also conclude that this experimental device generates a high segregation by density difference that give rise to a preferential flow in the centre of the funnel. It thus greatly perturbs the measures and the mixing process. In order to prevent from segregation, it is therefore particularly important not to fill the funnels, so that a solution could be to convey powders directly to the centre of it.

#### 4.2. Kinetics of mixing in a laboratory drum mixer: the Rotocube®

In this section, we apply the above methodology for the case of the Rotocube® mixer (see Fig. 9), and in particular, we examine the influence of the number of revolutions at a fixed rotational speed (30 rpm) on the mixture quality.

The first experiment was realised without any action of the Rotocube® and we just divided the initial preparation into three successive layers placed in a vibrating conveyor: the lower one was constituted by half of sugar, the middle one

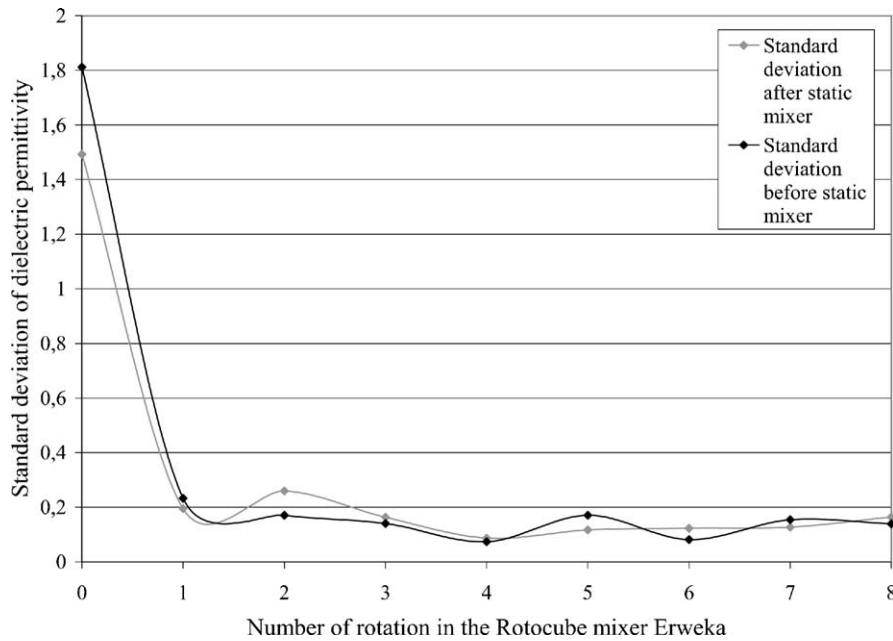


Fig. 11. Standard deviation of dielectric permittivity before and after the static mixer versus the number of revolutions in the Rotocube®.

by all the SiC and the upper one by the rest of sugar. All the preparations constituted of 20% by mass of SiC and 80% by mass of sugar. Fig. 10 clearly shows that this experimental device prevents from segregation inference, even if there is still left some, probably due to the conveyor's vibrations. It must be commented that the nominal value in SiC content is not reached because of the small thickness of the SiC layer and of a slight phenomenon of mixing between SiC and sugar when passing from the first layer to the second one.

Experiments were then realised with several number of revolutions of the mixer. The entire mixture flowed through the device and the standard deviation of the whole mixture (3600 samples) could be derived for each experiment. Fig. 11 shows the evolution of the standard deviation of the permittivity versus the number of revolutions. We can immediately see that the two plots are quite close, which shows that the static mixer mixes in a radial way, an effect that cannot be detected by the sensors we used. Concerning the shape of the plots, it is clearly shown that the mixing quality is improved by the Rotocube® since the first revolution. Then, quality is slowly bettered up to four revolutions and begins slowly to decrease after four revolutions. This can be due to a change in the mixing mechanism in the Rotocube® (passage from a shearing mechanism to a diffusion one), which may change the mixing structure. Note that the presence of oscillations, which are softened by the static mixer, may be due to segregation in the Rotocube®, but also in the vibrating system.

## 5. Conclusions

Hopefully, dielectric constants of current materials are covering a wide range of values, which also means that such a method can be applied in various industrial applications.

This study has shown the possibility to measure segregation problems when discharging mixtures through a funnel. It has also shown the effect of the number of drum revolutions at a fixed speed on the quality of a mixture, as well as the effect of a static mixer. The axial structure of the mixtures through autocorrelation functions could also be studied from these data, but this has not been reported here for clarity of the paper. In addition, it must be remembered that we just studied the evolution of the dielectric permittivity, and that in most cases, we will have to follow the volumetric compositions of each component in order to characterise the homogeneity of the medium. The capacitive method is indeed full of promise for particulate systems, and it would be interesting to explore it much into details, particularly for determining the proportions of each component of the mixture.

## Appendix A. Nomenclature

$d_{50}$	median diameter at 50%
$n$	consecutive elementary samples defining a window (of course $n < N$ )

$n_0$	number of samples that form part of the default and should probably be taken away from the production line
$n_p$	number of packets that are affected by the perturbation
$N$	number of considered consecutive elementary samples into which the powder mixture flow is divided
$N_c$	number of components in the powder mixture, including the air trough the porosity of the medium
$P_i$	volumetric proportion of the component $i$ in the powder mixture
$r$	spatial (or temporal) variable corresponding to the distance (or time elapsed) between two samples
$r_0$	distance (or time elapsed) between two samples corresponding to the equation $R(r) = 0$
$R(r)$	autocorrelation function
$x_i$	weight composition in a key component in the elementary sample $i$

## Symbols

$\mu$	mean observed for the $N$ consecutive samples
$\sigma$	standard deviation for the $N$ consecutive samples
$\mu_{k,n}$	mean for a packet of $n$ samples beginning at sample number $k$
$\sigma_{k,n}$	standard deviation for a packet of $n$ samples beginning at sample $k$
$\varepsilon_i$	dense dielectric permittivity of the component $i$ in the powder mixture
$\varepsilon_m$	dielectric permittivity of the powder mixture

## References

- [1] F.J. Muzzio, T. Shinbrot, B.J. Glasser, Powder technology in the pharmaceutical industry: the need to catch up fast, *Powder Technol.* 124 (2002) 1–7.
- [2] J. Berman, A. Schoeneman, J.T. Shelton, Unit dose sampling: a tale of two thieves, *Drug Dev. Ind. Pharm.* 22 (11) (1996) 1121–1132.
- [3] F.J. Muzzio, P. Robinson, C. Wightman, D. Brone, Sampling practices in powder blending, *Int. J. Pharm.* 155 (1997).
- [4] S. Massol-Chaudeur, Caractérisation de l'état de mélange de poudres – cas de mélanges faiblement dosés, Ph.D. Thesis, INPL Nancy, 2000.
- [5] J. Lister, Forward to special edition of powder technology from “Control of Particulate Processes VI”, *Powder Technol.* 124 (2002) 175.
- [6] T. Wang, Z.J. Lin, C.M. Zhu, D.C. Liu, S.X. Saxena, Particle velocity measurements in a circulating fluidized bed, *AIChE J.* 39 (8) (1993) 1406–1410.
- [7] J. Aguilon, K. Shakourzadeh, P. Guigon, A new method for local solid concentration measurements in circulating fluidized bed, *Powder Technol.* 86 (1996) 251–255.
- [8] J.J. Nieuwland, R. Meijer, J.A.M. Kuipers, W.P.M. van Swaaij, Measurements of solids concentration and axial solids velocity in gas–solid two phase flows, *Powder Technol.* 87 (1996) 127–139.
- [9] C.F. Harwood, R. Davies, M. Jackson, E. Freeman, An optic probe for measuring the mixture composition of powders, *Powder Technol.* 5 (1971/1972) 77–80.

- [10] R. Weinekötter, L. Reh, Characterization of particulate mixtures by in-line measurements, *Part. Part. Syst. Charact.* 11 (1994) 284–290.
- [11] D. Steinmetz, B. Hocine, M. Poux, C. Laguérie, Analyse du mélange de poudres en fluidisation gazeuse à l'aide de la méthode de défluidisation et par capteur à fibre optique, *Powder Technol.* 89 (1996) 139–147.
- [12] O. Berntsson, L.G. Danielsson, B. Lagerholm, S. Folestad, Quantitative in-line monitoring of powder blending by near infrared reflection spectroscopy, *Powder Technol.* 123 (2002) 185–193.
- [13] T. Dyakowski, L.F.C. Jeanmeure, A.J. Jaworski, Applications of electrical tomography for gas–solids and liquids–solids flows – a review, *Powder Technol.* 112 (2000) 174–192.
- [14] K.L. Ostrowski, S.P. Luke, M.A. Benett, R.A. Williams, Application of capacitance electrical tomography for on-line and off-line analysis of flow pattern in horizontal pipeline of pneumatic conveyer, *Chem. Eng. J.* 77 (2000) 43–50.
- [15] E. Brunazzi, A. Paglianti, S. Pintus, A capacitance probe and a new model to identify and predict the capacity of columns equipped with structured packings, *Ind. Eng. Chem. Res.* 40 (2001) 1205–1212.
- [16] W. Warsito, L.S. Fan, Measurement of real-time flow structures in gas–liquid and gas–liquid–solid flow systems using electrical capacitance tomography (ECT), *Chem. Eng. Sci.* 56 (2001) 6455–6462.
- [17] T. Loser, M. Geweke, D. Mewes, Measuring the Phase Distribution in a Transient Gas–Solid Flow Using Capacitance Tomography, 3rd Israeli Conference for Conveying and Handling of Particulate Solids, Dead Sea, Israel, 2000, pp. 15.11–15.15.
- [18] T. Dyakowski, A.J. Jaworski, Application of tomographic techniques for imaging fluidized beds, 3rd Israeli Conference for Conveying and Handling of Particulate Solids, Dead Sea, Israel, 2000, pp. 15.1–15.10.
- [19] A. Kantzas, Computation of holdups in fluidized and trickle beds by computer-assisted tomography, *AIChE J.* 40 (7) (1994) 1254–1261.
- [20] D. Newton, M. Fiorentino, G.B. Smith, The application of X-ray imaging to the developments of fluidized bed processes, *Powder Technol.* 120 (2001) 70–75.
- [21] N. Sommier, P. Porion, P. Evesque, B. Leclerc, P. Tchoreloff, G. Couarraze, Magnetic resonance imaging investigation of the mixing-segregation process in a pharmaceutical blender, *Int. J. Pharm.* 222 (2001) 243–258.
- [22] C.K. Lai, D. Holt, J.C. Leung, C.L. Cooney, G.K. Raju, P. Hansen, Real-time noninvasive monitoring of dry powder blend homogeneity, *AIChE J.* 47 (11) (2001) 2618–2622.
- [23] J. Villermaux, Génie de la réaction chimique – Conception et fonctionnement des réacteurs, Tec&Doc, Lavoisier ed, 1984, p. 204.
- [24] P.V. Danckwerts, Continuous flow systems: distribution of residence times, *Chem. Eng. Sci.* 2 (1953) 1–11.
- [25] P.V. Danckwerts, Theory of mixtures and mixing, *Res. (London)* 6 (1953) 355–361.
- [26] J. Gyenis, Assessment of mixing mechanisms on the basis of concentration patterns, *Chem. Eng. Process.* 38 (1999) 655–674.
- [27] R.C. Waterfall, W.W. Loh, Arko, J. Covy, G.P. Lucas, Electrical tomography for multiphases flow applications, 3rd Israeli Conference for Conveying and Handling of Particulate Solids, Dead Sea, Israel, 2000, pp. 17–25.
- [28] UMIST, Industrial Process Tomography at UMIST, <http://www.tomography.umist.ac.uk>.
- [29] D.A.G. Bruggeman, *Ann. Phys. (Leipzig)* 24 (1935) 636.
- [30] T. Guillot, Contribution à la modélisation des propriétés électromagnétiques des mélanges aléatoires du type diélectrique/conducteur, ONERA, Note technique 3, 1992.
- [31] S. Muerza, H. Berthiaux, S. Massol-Chaudeur, G. Thomas, A dynamic study of static mixing using on-line image analysis, *Powder Technol.* 128 (2002) 195–204.