Refereed Proceedings

The 13th International Conference on Fluidization - New Paradigm in Fluidization Engineering

Engineering Conferences International *Year* 2010

AERATION AND MIXING BEHAVIOURS OF BINARY MIXTURES OF NANO-SIZED POWDERS UNDER SOUND VIBRATION

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ABSTRACT

The aeration behaviour of three different nano-sized powders, Al_2O_3, Fe_2O_3 and CuO, fluidized under the application of acoustic fields of different intensities (125-150 dB) and frequencies (50-300 Hz) has been characterized. The characterization of the fluidization of binary mixtures of Al_2O_3 and Fe_2O_3 has been also investigated under the application of acoustic fields of 120 Hz and of two different intensities, 130 and 135 dB, and varying the relative amount of two powders from 17 to 50 %wt of Fe_2O_3. The addition of Fe_2O_3 has a beneficial effect on fluidization quality of Al_2O_3 even for the lowest amount of Fe_2O_3. Under the effect of sound, mixing between Al_2O_3 and CuO nanopowders has been also quantitatively characterized by the Scanning Electron Microscopy with X-ray microanalysis (SEM/EDS) analysis of captured samples. Mixing between aggregates of the two powders takes only few minutes. However, mixing also occurs inside aggregates but this process requires times of the order of 80-150 minutes, depending on the feeding order of two powders.

INTRODUCTION

Nanopowders (< 100 nm) have received increased attention due to their unique characteristics (1) in different industrial sectors such as the manufacturing of pharmaceuticals, cosmetics, chemicals, ceramics and electronics. However, whatever the process, a key aspect not entirely investigated is how these nanoparticles can be effectively handled. In fact, the processing of ultrafine powders is very challenging due to extremely strong inter-particle forces, responsible for the formation of relatively large aggregates (1,2). Fluidization is one of the most promising technologies to treat a large amount of nanoparticles in a gas–solid system (1-3), especially when some external devices, such as the application of mechanical vibration (3), acoustic (2,4), centrifugal (5), electric (6) or magnetic (7) fields, are used. In particular, under the influence of appropriate acoustic fields, channeling or slugging tend to disappear and the beds expand uniformly, with a reduction of the minimum fluidization velocity (8). Zhu et al. (8) reported that the optimal sound frequency range is 20-1000 Hz; in particular, from 200 to 600 Hz bubbling fluidization has been observed. On the other hand, Guo et al. (2,4,9) reported that the optimal sound frequency range is 40-70 Hz and the fluidization

behaviour appears similar to that of Geldart group A particles with no bubbles and negligible elutriation. At sound frequency exceeding these respective ranges, sound has almost no impact on the fluidization: the aeration process encounters slugging and fluidization process is accompanied by much elutriation. As regards the effect the sound pressure level, below a critical value, 115 dB (8) or 100 dB (2,4,9), there is no effect. Moreover, the fluidization quality generally improves as the sound pressure level increases (2,4,8,9). Only few studies can be found in literature on mixing behaviour of different nanopowders (5,10). In particular, Nakamura and Watano (5) analyzed the mixing of two nanopowders, SiO_2 and Al_2O_3, under the effect of a centrifugal acceleration. They found that the mixing occurred at a micron-scale.

Present work is focused on the characterization of the behaviour of binary mixtures of nanopowders fluidized under the application of different acoustic fields. A preliminary assessment of the efficiency of sound application to promote the mixing between two different nanopowders has been also carried out. As regards the last point, both the mixing of aggregates of the two powders and the presence of mixed aggregates made of single particles of the two powders have been investigated analyzing samples of the bed taken at different times by means an “ad hoc” non-destructive sampling procedure.

EXPERIMENTAL

Apparatus and materials

The experimental apparatus consists of a laboratory scale fluidization column (40 mm ID and 500 mm in height), made of Plexiglas, equipped with a porous plate gas distributor, a 300 mm high wind-box filled by Pyrex rings to ensure an uniform distribution of gas flow, a set of filters for the collection of elutriated fines at the column exit, a pressure transducer (Hartmann & Braun) installed at 5 mm above the gas distributor to measure the pressure drop across the bed, a sound wave guide at the top of the freeboard, a sound-generation system and a data acquisition system Details on the sound generation system are reported elsewhere (11). Pure dry nitrogen from a compressed tank is used as the fluidizing gas. The flow rate of gas is controlled by a mass flow controller (Bronkhorst). Both the fluidization and the mixing tests have been carried out at room temperature and ambient pressure conditions. The behaviour of three different nano-sized powders, Al_2O_3, Fe_2O_3 and CuO (Sigma-Aldrich), has been investigated. The powders have primary particle average sizes lower than 50 nm and primary particle densities of about 4000, 4500 and 6300 kg/m^3, respectively.

Procedure for characterization of fluidization

Aeration tests of single nanopowders have been performed in correspondence of an initial bed height (H_0) fixed at about 15 cm, corresponding to a bed of about 48, 33 and 35 g for Al_2O_3, Fe_2O_3 and CuO, respectively. Pressure drop and bed expansion curves have been obtained in two different experimental conditions: i) aeration with a fluidizing gas; ii) aeration with both a fluidizing gas and the application of acoustic fields of different intensities (125-150 dB) and frequencies (50-300 Hz).

The fluidization tests of binary mixtures of nanopowders have been carried out using the Al_2O_3 and the Fe_2O_3 materials, which showed different fluidization behaviours. Two different relatively low sound intensities (130 and 135 dB) at a fixed frequency (120 Hz) have been applied. Tests have been performed by feeding 30 g of the binary mixtures and varying the relative amount of two powders from about 17 to 50 %wt of Fe_2O_3. The effect of the feeding order of two powders has been also investigated.

Procedure for characterization of mixing

Mixing tests have been carried out using the Al_2O_3 and the CuO powders, which showed similar fluidization behaviours, with application of an acoustic field (140 dB – 120 Hz) able to fluidize both powders. The superficial gas velocity has been fixed at about 0.45 cm/s, enough to fluidize the materials under the application of the above acoustic field. For all the experimental conditions tested, the starting point was obtained by feeding 30 g of Al_2O_3 nanopowder (white powder) and 5 g of CuO nanopowder (black powder), used as a tracer. Two different feeding orders of the powders inside the column have been considered: in the A case the Al_2O_3 was firstly fed and then the CuO, whereas in the B case the CuO was firstly fed and then the Al_2O_3. Each test has been carried out for about 120 minutes. The visual observation of the bed, also recorded by a video camera, gave some preliminary rough information on the uniformity of mixing and the mixing characteristic time at a global scale (11). During experiments, samples of the fluidized materials have been taken at different times by means an “ad hoc” non-destructive sampling procedure. In particular, a probe made of a silicon tube, linked to the adhesive sample disk used for SEM analysis, has been inserted from the top of the reactor, so that the fluidized materials present in the upper part of the bed sticked on it. The different samples have been then analyzed by SEM/EDS analysis in order to determine the shape and the chemical composition of aggregates.

RESULTS

Characterization of fluidization

The fluidization quality of Al_2O_3 and CuO nanopowders was very poor without the application of acoustic fields, as confirmed by the presence of channels across the bed, the irregular surface of the beds together with the absence of particle motion and very low dimensionless bed expansions (< 1.1). However, it was possible to individuate a critical value of the superficial gas velocity (about 4 cm/s), above which the dimensionless pressure drop approaches the unity. On the contrary, the Fe_2O_3 nanopowder showed a better fluidization quality without the application of acoustic fields, the critical value of the superficial gas velocity being about 1 cm/s and the dimensionless bed expansion approaching values as high as 1.5. The application of acoustic fields of different intensities and frequencies generally resulted in more regular pressure drop and bed expansion curves for three powders. However there were some differences that make possible to identify the ranges of intensities and frequencies of acoustic fields most effective to stabilize a good fluidization regime. The maximum values of dimensionless pressure drop ($\Delta P_{max}/\Delta P_0$), where ΔP_0 is the pressure drop equal to buoyant weight of particles per unit area of bed, and of the bed expansion ratio (H_{max}/H_0), derived from experimental pressure drop and bed

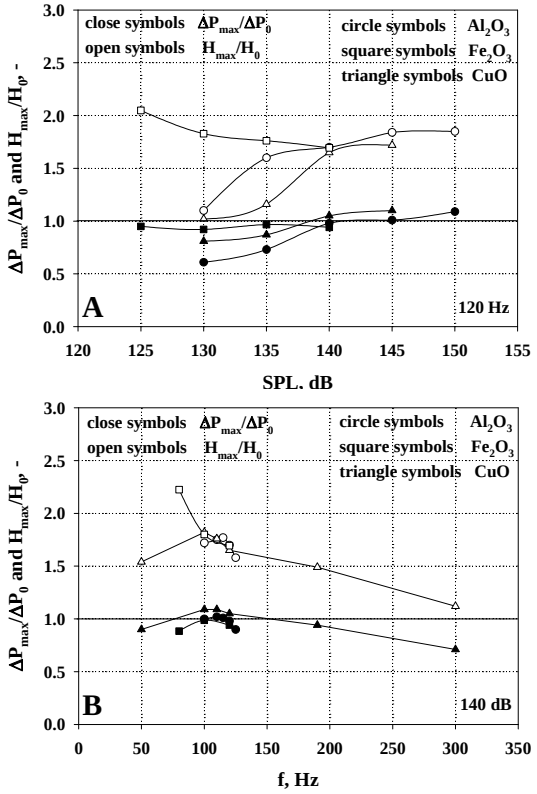

Figure 1. A) Effect of sound intensity (f = 120 Hz) and B) effect of sound frequency (SPL = 140 dB) on $\Delta P_{max}/\Delta P_0$ and H_{max}/H_0 on fluidization of single nanopowders.

expansion curves, could be considered as an index of the fluidization quality, i.e. $\Delta P_{max}/\Delta P_0 \rightarrow 1$ and higher values of H_{max}/H_0 suggested a better fluidization quality. Fig. 2 reports the values of $\Delta P_{max}/\Delta P_0$ and H_{max}/H_0 as functions of SPL at fixed frequency f = 120 Hz (A) and as a function of f at fixed sound pressure level SPL = 140 dB (B), for three nanopowders. The fluidization quality of both Al_2O_3 and CuO improves, i.e. both the $\Delta P_{max}/\Delta P_0$ and H_{max}/H_0 increased, as the sound intensity increased. However, under the experimental conditions tested, sound intensities at the top of the bed higher than 135 dB were required to obtain values of $\Delta P_{max}/\Delta P_0$ close to unity and of H_{max}/H_0 higher than 1.6. On the contrary, the fluidization quality of Fe_2O_3 nanopowder was quite insensitive to the effect of SPL and intensities higher than 125 dB were enough to achieve a good fluidization quality, probably due to the intrinsic better fluidizability of this powder with respect to the other two powders. Similar results have been obtained for other frequencies. However, it must be noted that these critical values of sound intensity were larger than those found by other Authors, 100 dB (2,4,9) or 115 dB (8). It is likely that both the differences in particle size range of tested nanoparticles or of their aggregates and sound attenuations in deeper beds could account for such differences. On the other hand, the effect of frequency was not monotone, according to literature data (2,4,8,9), and an optimum condition, in terms of a highest values of $\Delta P_{max}/\Delta P_0$ and H_{max}/H_0, has been found, whatever the powder, for 80 Hz < f < 125 Hz. Similar behaviours have been also observed for different sound intensities. The explanation of this trend was not straightforward; it could be related to the ability of sound to penetrate the bed as well as to promote aggregates reduction into a scale which depends also on powder structure. For frequencies higher than about 125 Hz the acoustic field was not able to propagate inside the bed, while for frequencies lower than about 80 Hz the relative motion between smaller and larger sub-aggregates, i.e. the break-up of the large aggregates originally present in the bed, was practically absent. Between these values there was a range of optimal frequencies able to promote a maximum aggregates break-up.

The characterization of the fluidization behaviour of binary mixtures of Al_2O_3 and Fe_2O_3 nanopowders has been investigated under the application of acoustic fields of 120 Hz and of two different intensities, 130 and 135 dB, corresponding to a good and a poor fluidization quality of Fe_2O_3 and Al_2O_3, respectively. In particular, the

effect of increasing amount of Fe_2O_3 (the more fluidizable powder), from 17%wt to 50%wt, on fluidization quality of Al_2O_3 (the less fluidizable powder) has been tested. The effect of different order of feeding of two powders has been also investigated. Figure 2 reports the dimensionless pressure drop $\Delta P/\Delta P_0$ and bed expansion H/H_0 curves obtained for the different binary mixtures under the effect of different acoustic fields. The same curves obtained during the fluidization of single powders (only Al_2O_3 and Fe_2O_3) have been also reported for comparison. The addition of Fe_2O_3 had a beneficial effect on fluidization quality of Al_2O_3. A strong improvement of fluidization quality of Al_2O_3 has been already observed for the lowest amount of Fe_2O_3, increasing amounts of Fe_2O_3 having a reduced beneficial effect. This beneficial effect was more evident in the case of 130 dB than 135 dB, being the fluidization quality of single Al_2O_3 better at 135 dB. The order of feeding of two powders had a negligible effect.

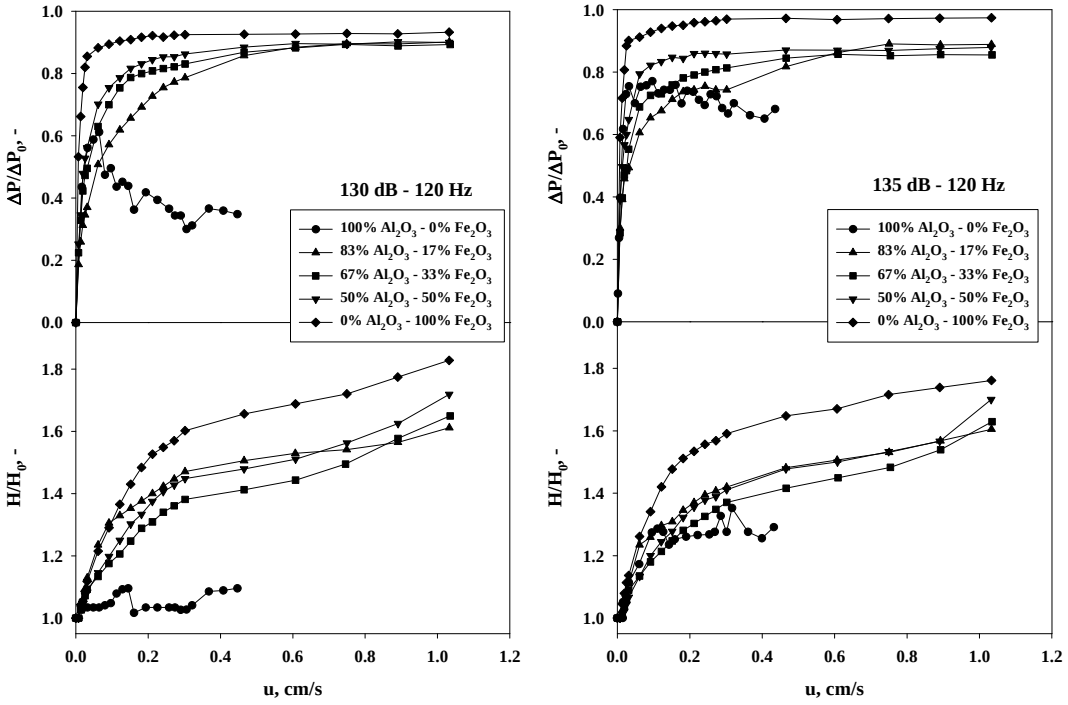

Figure 2. Dimensionless pressure drop ($\Delta P/\Delta P_0$) and bed expansion ratio (H/H_0) as a function of superficial gas velocity during aeration with application of different acoustic fields for binary mixtures of Al_2O_3 and Fe_2O_3.

Characterization of mixing

The possibility that there was an efficient mixing between different nanopowders during aeration, promoted by the application of a suitable acoustic field, has been highlighted by experiments carried out using a powder (CuO) as a tracer in a bed made of the other powder (Al_2O_3). The visual observation of the bed, also recorded by a video camera, already gave some preliminary qualitative information on the uniformity of mixing and the mixing characteristic time at a global scale (11). The application of a suitable acoustic field (140 dB – 120 Hz) resulted into a relatively

large bed expansion and promoted a solid mixing. After few minutes the entire bed turned grey and appeared well mixed. Even exchanging the order of feeding of powders, similar mixing was approached in few minutes of aeration under the same experimental conditions.

In order to determine the shape and the chemical composition of aggregates, a SEM/EDS analysis of samples of the fluidized materials taken from the upper part of the bed at different times has been performed. On the basis of the initial bed composition (30 g of Al_2O_3 and 5 g of CuO), the Al and Cu weights corresponded to 80% and 20%, respectively. The EDS analysis performed on entire areas of bed samples showed that the Al weight composition was about 83-86% already after 1 min of sound assisted aeration whatever the order of feeding of powders and after 120 min this value reduced to 80-82%. These results confirmed the information obtained by the visual observation of the bed, i.e. at a global scale the mixing of the bed really occurred within very short times.

The mixing at the global scale could be associated to two possibilities of mixing at a local scale inside the aggregates: the sound assisted aeration could promote only a mixing of aggregates of single powders or a mixing of sub-aggregates, due to a continuous break-up and re-forming of single aggregates, leading to the formation of mixed aggregates. Fig. 3 reports SEM images and the EDS analysis of aggregates taken at different times during both A and B mixing tests. Analysis of the figure showed that all aggregates were formed of both alumina and copper oxide. Their compositions were variable in a wide range. Aggregates richer both in alumina and in copper oxide were present, but their shapes and sizes appeared to be similar to each other, therefore it was not possible to distinguish the different composition of aggregates on the basis of morphological analysis. Fig. 4 reports the cumulative distribution of aggregates composition at different times obtained for A (Fig. 4A) and B (Fig. 4B) mixing tests. N denotes the number of aggregates having an Al weight

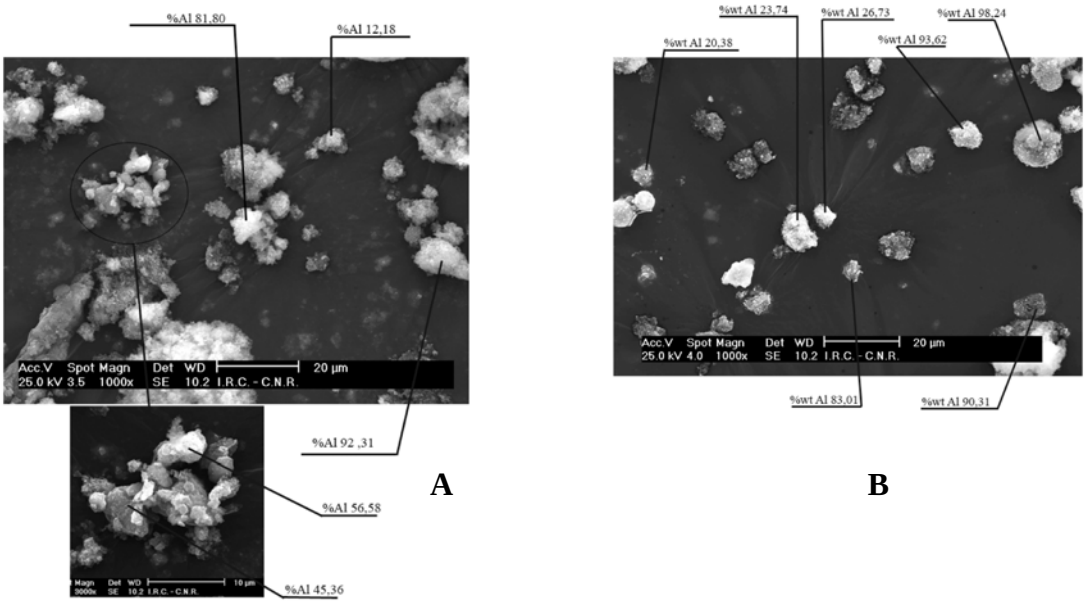

Figure 3. SEM images and EDS analysis of nanoparticle aggregates. A) sample taken after 14 min during the A mixing test; B) sample taken after 30 min during the B mixing test.

composition lower than the x-axis value and N_t represents the total number of aggregates. The analysis of the curves highlighted that all aggregates were mixed even after 1 min of sound assisted aeration even if their composition was variable in a wide range.

Aggregates composition data reported in Fig. 4 have been worked out to obtain the curves, reported in Fig. 5, representing the time dependence of aggregates mixing degree M(t) for both A and B mixing tests. The aggregates mixing degree M at a fixed time t has been defined as the ratio between the number of aggregates with the Al composition in the range 70-90% (i.e. near the limit value of 80%) and the total number of aggregates analyzed at time t. Each data series has been fitted with an exponential rise-to-maximum law: $M(t) = a\left[1 - \exp\left(-\frac{t}{b}\right)\right]$. The parameters a and b have been obtained by least squares minimization. The two dependence laws $M_A(t)$ and $M_B(t)$ obtained for A and B mixing test respectively were also reported in Fig. 5. The analysis of the curves showed that the dynamic phenomena of local mixing evolved in times longer than those characteristic of the global mixing. The maximum aggregates mixing degree was the same (52-53%) for both A and B mixing tests, i.e. half of the aggregates had a composition close to the limit value. The order of placement of the two powders affected the time dependence of the aggregates mixing degree: in the B case (the CuO tracer was initially located under the Al_2O_3 bed) the characteristic time of local mixing (about 150 min) required to reach the maximum mixing degree was twice as big as the one (about 80 min) obtained for A case (the CuO tracer was initially located under the Al_2O_3 bed).

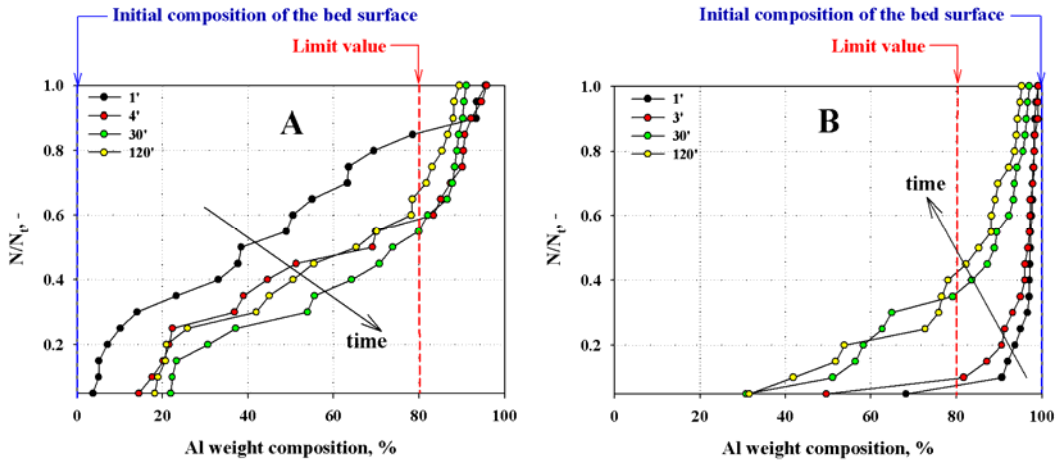

Figure 4. Cumulative distribution of aggregates at different times. A) A mixing test; B) B mixing test.

CONCLUSIONS

The aeration behaviour of three different nano-sized powders, Al_2O_3, Fe_2O_3 and CuO, fluidized under the application of acoustic fields of different intensities (125-150 dB) and frequencies (50-300 Hz) has been firstly characterized.

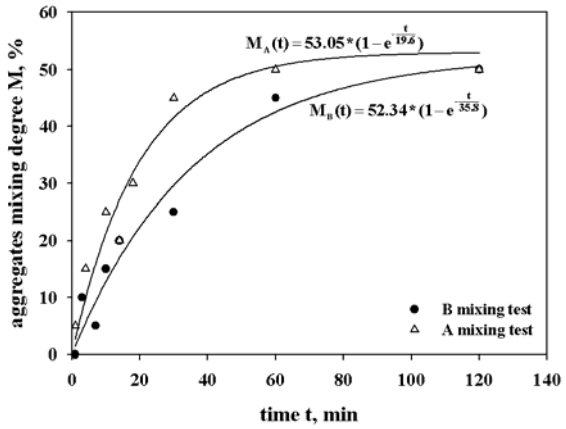

Figure 5. Time dependence of aggregates mixing degree for A and B mixing tests.

The characterization of the fluidization of binary mixtures of Al_2O_3 and Fe_2O_3 has been also investigated under the application of acoustic fields of 120 Hz and of two different intensities, 130 and 135 dB, and varying the relative amount of two powders from 17 to 50 %wt of Fe_2O_3. The addition of Fe_2O_3 had a beneficial effect on fluidization quality of Al_2O_3 even for the lowest amount of Fe_2O_3.

The investigation on the efficiency of sound to promote the mixing of two nano-sized powders (Al_2O_3 and CuO) has been also carried out. The visual observation of the bed and the SEM/EDS analysis of captured samples showed that: i) sound was able to promote the powders mixing on a global scale, whereas, at a local scale aggregates of particles were characterized by a maximum mixing degree of about 50%, whatever the initial feeding order of powders; ii) the time required to reach the global mixing of the two powders was of few minutes, smaller than those required to obtain a rather stationary condition in mixing inside aggregates which required 80-150 minutes depending on the feeding order of powders.

ACKNOWLEDGMENT

Giovanni Gaggiano and Federica Raganati are gratefully acknowledged for their assistance during experiments. Authors are also grateful to Sabato Russo for performing the SEM/EDS analysis.

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