

DETERMINATION OF THE DUSTINESS OF RAW MATERIALS USED IN THE CERAMIC SECTOR

**A. López-Lilao ⁽¹⁾, A. Escrig ⁽²⁾, V. Sanfelix ⁽²⁾,
E. Monfort ^(1,2), G. Mallol ^(1,2)**

⁽¹⁾ Dpto. de Ingeniería Química. Universitat Jaume I. Castellón. Spain

⁽²⁾ Instituto de Tecnología Cerámica

Asociación de Investigación de las Industrias Cerámicas

Universitat Jaume I. Castellón. Spain

ABSTRACT

In this study, the dustiness of raw materials used in tile body compositions and frit and glaze compositions in the ceramic sector was determined. The tests were performed with the continuous drop method, one of the reference test methods according to standard UNE EN 15051:2007 "Workplace atmospheres – Measurement of the dustiness of bulk materials – Requirements and reference test methods".

On the other hand, with a view to relating the dustiness of the materials to their properties, the particle size distribution, Hausner ratio, flow factor, specific surface area, and moisture content of these materials were determined.

A database was drawn up of the dustiness of raw materials used in the ceramic sector, together with a classification of these materials according to the criteria established in the above standard. A preliminary evaluation was also made of the influence of certain parameters on the dustiness of particulate materials.

This information is deemed essential for establishing the most efficient preventive and/or corrective measures to reduce the generation of fugitive emissions of particulate matter (PM), both into the outside atmosphere (air pollution) and inside the facilities (occupational health).

1. INTRODUCTION

Recent studies [1-8] show that in areas with high concentrations of industry that handle particulate materials, such as the ceramic industry, fugitive emissions of particulate matter (PM) can contribute significantly to overall PM emissions into the atmosphere.

As a result, in the new legislation on emissions in industrial activities (content of the Integrated Environmental Authorisations, emissions declarations requested by the European Union through EPER-PRTR, and content of the Air Quality Improvement Plans), fugitive emissions are identified as a feature to be controlled and quantified in order to be able to establish and identify actions that will reduce such PM fugitive emissions.

On the other hand, the legislation on occupational health stresses the need to limit the risks inherent to exposure to PM, mainly by inhalation, in occupational environments.

A parameter of great interest for evaluating, controlling, and minimising the risks associated with PM fugitive emissions, generated during the handling of particulate materials, is the dustiness of the materials, i.e. their tendency to produce dust when they are handled.

1.1. DUSTINESS

A great variety of testers [9] are available for the determination of the dustiness of particulate materials. However, in general, these devices contain the following elements:

- Dust generation section
- Dust transfer section
- Sampling section
- Size fractionator or size fractionators
- Dust collection section

As the dustiness assessment of a material will depend on the apparatus used, standard UNE EN 15051:2007 [10] establishes two reference test methods: the rotating drum method and the continuous drop method.

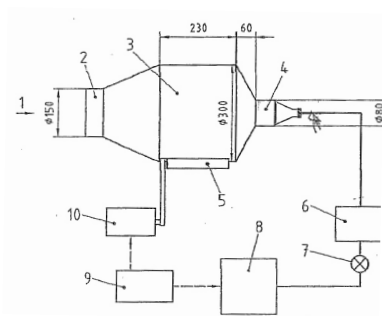
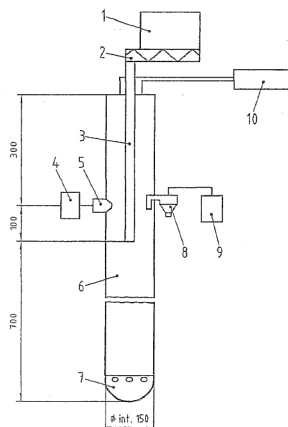
Method	Schematic illustration of the device	
Rotating drum method		1 Sample tank 2 Metering device 3 Drop pipe 4 Pump for sampling the inhalable aerosol fraction 5 Sampling head for the inhalable aerosol fraction 6 Backflow pipe 7 Collector tank 8 Sampling head for the respirable aerosol fraction 9 Pump for sampling the respirable aerosol fraction 10 Main pump
Continuous drop method		1 Sample tank 2 Metering device 3 Drop pipe 4 Pump for sampling the inhalable aerosol fraction 5 Sampling head for the inhalable aerosol fraction 6 Backflow pipe 7 Collector tank 8 Sampling head for the respirable aerosol fraction 9 Pump for sampling the respirable aerosol fraction 10 Main pump

Table 1. Reference test methods according to standard UNE EN 15051:2007.

The test enables the mass fractions of inhalable dust (W_I), thoracic dust (W_T), and respirable dust (W_R) to be determined. In addition, the results obtained allow the materials to be classified in different categories as a function of their dustiness, according to the criteria laid down in the above standard.

Method	Classification	Mass fraction of dust[powder] (mg/kg)		
		W_I	W_T	W_R
Method A Rotating drum method	Very low	<200	<40	<10
	Low	200 to 1000	40 to 200	10 to 50
	Moderate	>1000 to 5000	>200 to 1000	>50 to 250
	High	>5000	>1000	>250
Method B Continuous drop method	Very low	<250	-	<25
	Low	250 to 2500		25 to 125
	Moderate	2500 to 12500		125 to 1250
	High	>12500		>1250

Table 2. Dustiness classification according to standard UNE EN 15051:2007.

1.2. INFLUENCE OF THE PROPERTIES OF THE MATERIALS ON THEIR DUSTINESS

Dustiness depends on several factors, such as the handling to which the materials are subjected and the properties of the processed material. Various studies [11-20] have thus evaluated the influence of these parameters on the dustiness of particulate materials.

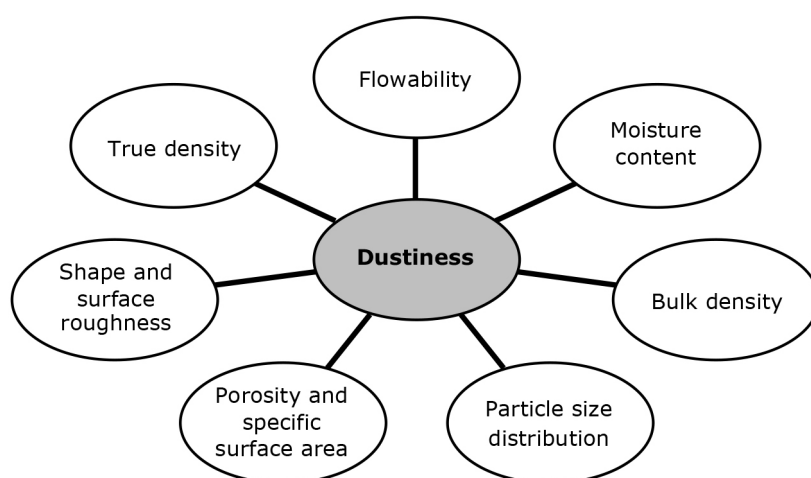


Figure 1. Variables influencing the dustiness of particulate materials.

One of the objectives of the present study was, thus, to determine the dustiness of the raw materials used in the ceramic industry and to evaluate the influence of the above variables on this parameter.

In this first phase of the study, a number of these variables that might a priori be highly influential were selected in order to relate them to the dustiness of the materials. The characterisation performed included the following variables: particle size distribution, flowability, specific surface area, and moisture content.

2. OBJETIVES

The objectives of this study were as follows:

- To draw up a database on the dustiness of raw materials used in the ceramic sector.
- To classify the studied raw materials as a function of their dustiness.
- To evaluate the influence of certain parameters on the dustiness of these particulate materials.

3. MATERIALS AND EXPERIMENTAL TECHNIQUES USED

3.1. SAMPLES

The study was conducted on a selection of 36 materials used in the ceramic tile and frit, glaze, and ceramic pigment manufacturing industry.

3.2. METHODOLOGY

The methodology applied may be divided into two lines of work: determination of dustiness and characterisation of the raw materials.

3.2.1. DUSTINESS

The tests were conducted using the continuous drop method, as this test apparatus was considered to better simulate how particulate materials are handled in the ceramic industry.

The sampling apparatus (Figure 2) consisted, basically, of a cylindrical pipe through which air circulated in an upward direction at a volume flow rate of 53 l/min. The test material was metred from the top of this pipe, at a rate of 6 to 10 g/min. The dust dropped through an inner pipe, concentric to the pipe through which the air rose. This tube was shorter than the outer pipe, so that the dust was released into a countercurrent airflow. A pair of sampling heads for health-relevant particle size fractions were located slightly above the discharge position of the material. The inhalable and the respirable fractions (UNE EN 481:1995) were studied. The inhalable fraction represented the suspended particles capable of being inhaled, whereas the respirable fraction represented the particle fraction that could infiltrate into the non-ciliated respiratory system.



Figure 2. Dustiness tester (ITC).

3.2.2. CHARACTERISATION OF THE RAW MATERIALS

The particle size distribution (PSD) of the studied materials was determined by laser diffraction, using the wet method (v_h). The resulting distribution by volume was then used to obtain parameters d_{10} , d_{50} , d_{90} , and d_{90}/d_{10} .¹

Moisture content was determined by drying, according to the procedure set out in standard UNE EN 15051:2007 [10].

The specific surface area was determined by nitrogen adsorption, according to the BET method.

The flowability of the materials was evaluated on the basis of two parameters:

- Hausner ratio (HR): This was determined from the quotient of the bulk density of the packed particle bed (by vibration or tapping) and the aerated bulk density of the particle bed, which was obtained by dropping powder into a container without stirring or vibration.
- Flow factor (ffc): The yield loci of the studied materials were determined using a rotational shear cell. These functions enabled the flow factor to be determined (ffc), defined as the quotient of the consolidation stress (σ_1) and the compressive strength of the bed for this consolidation stress ($ffc = \frac{\sigma_1}{f_c}$). This factor can be used to compare materials when the consolidation stress is fixed (σ_1). Specifically, for this study, a σ_1 of 20 kPa was used, as this value had also been used in previous studies on powder rheology [21].

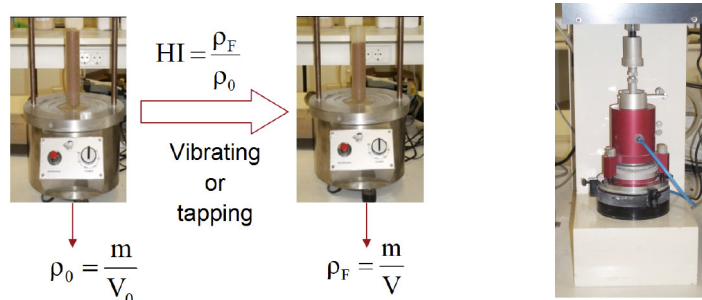


Figure 3. Hausner ratio (left) and picture of the rotational shear cell (right).

The determination of the factors described allows the materials to be classified as a function of their flowability, as detailed in Table 3.

Flow factor (ffc)	Hausner ratio	Behaviour of the material
$\text{ffc} < 1$	$\text{IH} > 1.4$	Non-flowing
$1 < \text{ffc} < 2$	$\text{IH} > 1.4$	Very cohesive
$2 < \text{ffc} < 4$	$\text{IH} > 1.4$	Cohesive
$4 < \text{ffc} < 10$	$1.4 > \text{IH} > 1.25$	Easy flow
$\text{ffc} > 10$	$1 > \text{IH} > 1.25$	Free flow

Table 3. Classification of the materials as a function of their flow factor and Hausner ratio.

¹ Where d_{10} , d_{50} , and d_{90} are the diameters below which lie 10%, 50%, and 90% by volume, respectively, of the total particles

4. RESULTS AND INTERPRETATION

4.1. DATABASE

The results obtained are detailed in Table 4. The table includes the treatment to which the materials were subjected (when applicable), moisture content, and the inhalable and respirable fractions of the selected samples.

Sample	Treatment	Moisture content (%)	w_I (mg/kg)	Classification	w_R (mg/kg)	Classification
Clay 1	-	15.8	1	Very low	0.5	Very low
Clay 2	-	10.3	2	Very low	0.5	Very low
Clay 3	-	14.1	2	Very low	1	Very low
Clay 4	-	14.8	5	Very low	<DL	Very low
Clay 5	-	9.5	60	Very low	3	Very low

Zinc oxide	-	0.2	65	Very low	6	Very low
Quartz 1	-	0.4	743	Low	<DL	Very low
Clay 6	-	7.7	977	Low	5	Very low
Clay 7	-	5.2	1000	Low	19	Very low
Clay 8	-	6.4	1100	Low	13	Very low
Clay 9	-	5.1	1300	Low	17	Very low
Micronised zircon	-	0.4	1847	Low	18	Very low
Alumina 1	-	0.6	1896	Low	11	Very low
Calcium carbonate 1	-	0.3	3570	Moderate	21	Very low
Spray-dried red-body stoneware tile powder	-	5.3	3700	Moderate	19	Very low
Red pigment	-	0.3	3885	Moderate	151	Moderate
Sodium feldspar	-	0.4	6122	Moderate	76	Low
Talc 1	-	0.8	6180	Moderate	132	Moderate
Quartz 2	-	0.2	6873	Moderate	53	Low
Black pigment	-	0.5	8225	Moderate	456	Moderate
Quartz 3	-	0.3	9396	Moderate	132	Moderate
Quartz 4	-	0.4	9666	Moderate	89	Low
Quartz 5	-	0.4	10080	Moderate	119	Low
Calcium carbonate 2	-	0.4	10492	Moderate	186	Moderate
Clay 10*	Drying	<0.1	11600	Moderate	111	Low
Colemanite	-	2.0	13186	High	37	Low
Clay 11 *	Grinding and drying	1.1	14110	High	36	Low
Kaolin *	Grinding	1.1	14475	High	91	Low
Zirconium flour	-	0.2	14646	High	121	Low
Yellow pigment	-	0.1	15378	High	165	Moderate
Clay 12 *	Clay 11 moistening	7.7	15552	High	66	Low
Clay 13 *	Clay 11 moistening	4.1	17695	High	32	Low
Alumina 2	-	0.3	19329	High	65	Low
Clay 14 *	Grinding	1.1	19439	High	142	Moderate

Clay 15 *	Grinding	1.4	20985	High	135	Moderate
Talc 2	-	0.4	63680	High	884	Moderate

Table 4: Dustiness database (increasing order according to the inhalable dust fraction).

(*) All the materials were tested in the as-received state except for the materials indicated.

<DL: Below the detection limit

This database allowed evaluation of the environmental issues associated with the generation of PM fugitive emissions of the different materials.

The results obtained show that the materials used in the ceramic industry encompass a wide range of dustiness. However, dustiness is not an intrinsic material characteristic but depends on the storage and handling conditions.

4.2. INFLUENCE OF THE VARIABLES

4.2.1. Effect of moisture content on the clays

The variation of (ground and unground) clay dustiness with moisture content is plotted in Figure 4.

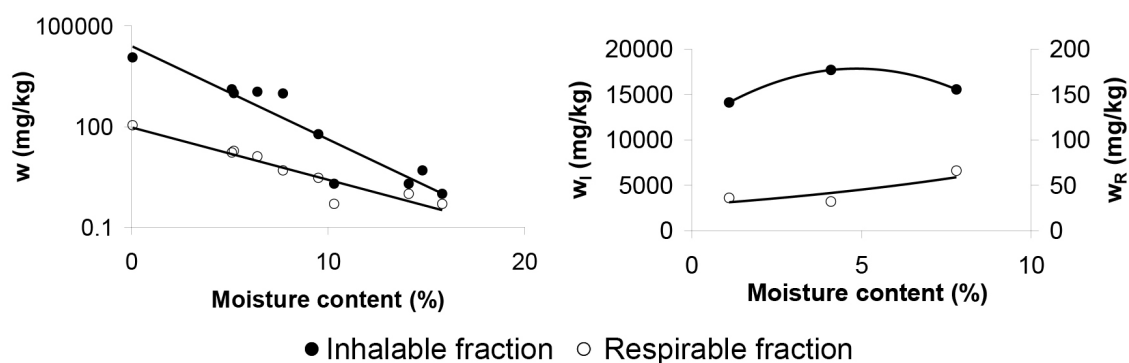


Figure 4. Variation of clay dustiness with moisture content. A) unground clays (left) B) ground clays (right).

It may be observed in Figure 4 that, in the unground clays, independently of the nature of the clay, dustiness decreased with moisture content, an effect noted in numerous previous studies [11-14]. However, the ground clays displayed a more complex behaviour because, at moisture contents of the order of 1%, clay dustiness was less than at higher moisture contents. This effect, which has also been observed in other materials [14-16], could stem from the fact that, in micronised clays, the attractive surface forces are dominant at low moisture contents, so that the addition of water initially has a lubricating effect, decreasing these attractive forces, whereas at higher moisture contents the water acts as a binder, reducing dustiness. These two opposing effects lead the ground clay to exhibit maximum dustiness at intermediate moisture contents.

4.2.2. Behaviour of materials of different nature

In the case of the test clays, the changes in moisture content appeared to explain the variations observed in dustiness (Figure 4). However, Table 4 shows that, when

materials of different nature are involved, other parameters need to be evaluated in order to explain the differences noted in dustiness.

Consequently, in this study, a series of materials displaying a wide range of dustiness were selected from the database. These materials were then thoroughly characterised with a view to identifying the parameters that significantly affected dustiness. The results obtained are detailed in Table 5.

Material	Moisture content (%)	a)PSD (μm)			d_{90}/d_{10}	Flowability		ρ_s (m^2/g)	Mass fractions (mg/kg)	
		b) d_{10}	b) d_{50}	b) d_{90}		HR	ffc		w_I	w_R
Micronised zircon	0.4	0.7	1.8	3.8	5.4	1.60	4.1	4.5	1847	18
Alumina	0.6	2.6	8.4	16.9	6.5	1.76	3.6	4.9	1896	11
Feldspar	0.4	3.8	15.2	40.3	10.6	1.63	4.1	1.1	6122	76
Quartz 1	0.2	3.7	19.7	53.9	14.7	1.56	4.9	0.9	6873	53
Quartz 2	0.3	4.0	23.3	67.3	16.9	1.52	5.0	0.8	9396	132
Quartz 3	0.4	3.8	29.1	80.4	21.2	1.51	6.5	0.6	9666	89
Quartz 4	0.4	4.5	29.9	97.7	21.7	1.47	6.0	0.7	10080	119
Calcium carbonate	0.4	2.9	7.5	19.6	6.8	1.62	5.4	0.7	10492	186
Zirconium flour	0.2	2.2	15.7	58.6	26.2	1.49	4.4	1.1	14646	121

Table 5. Characterisation of the selected raw materials and their dustiness.

- a) PSD: Particle size distribution
b) d_i : diameter below which lies $i\%$ by volume of total particles
c) S: specific surface area

Dustiness is not determined by a single parameter but depends on multiple factors, whose influence on dustiness is quite complex [11-21]. However, the materials studied displayed certain trends, which may be observed in the plots below.

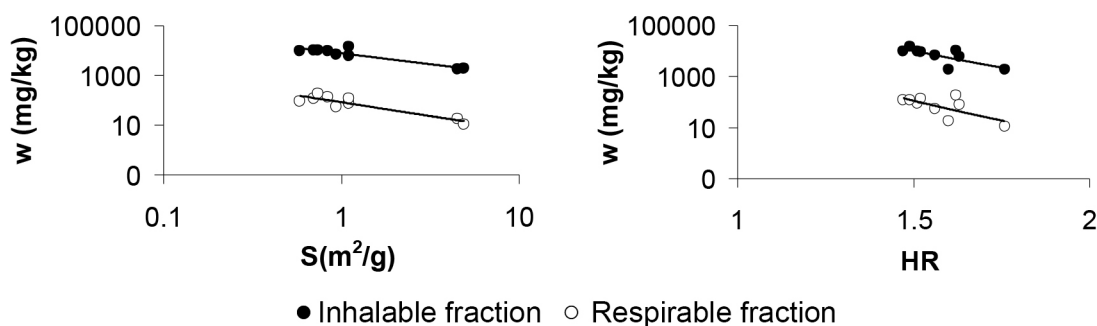


Figure 6. Variation of dustiness with specific surface area (left) and with the Hausner ratio (right).

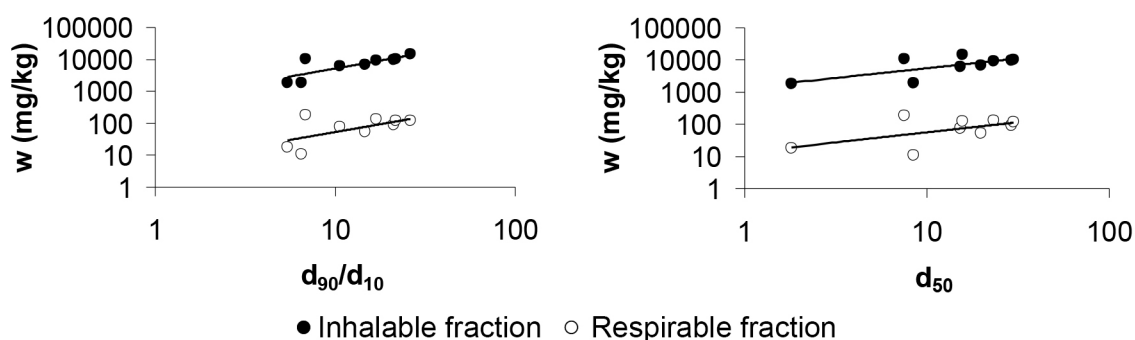


Figure 7. Variation of dustiness with the breadth of the PSD (left) and with d_{50} (right).

The graphs show that, in general, the studied materials exhibited the following behaviour:

- When the Hausner ratio increased, i.e. flowability decreased, dustiness diminished. This might be because materials with lower flowability display greater cohesiveness and therefore, as other authors have indicated [14], lower dustiness.
- A rise in specific surface area entailed a reduction in dustiness. This might be because, in the studied materials, this rise increased the number of surface contact points, which encouraged interparticle adhesive forces.
- With regard to particle size distribution, it was observed that both the increase in PSD breadth (d_{90}/d_{10}) and the increase in average particle size (d_{50}) raised dustiness. The effect of the PSD breadth might be caused, in the absence of a more thorough characterisation, by the variation in flowability observed in the tested materials. The effect of d_{50} , for the studied range of sizes ($d_{50} < 30\mu\text{m}$), might be caused, as other authors have indicated [12,17,18], by the surface forces dominating over the volume forces in the small particles. Consequently, a reduction in size decreased dustiness, as against what might intuitively be expected. If materials with increasing values of d_{50} were included, above a certain size, dustiness might be observed to decrease when particle size increased, as volume forces would dominate over the surface forces.

5. CONCLUSIONS

- The dustiness tester designed according to the specifications of standard UNE EN 15051 allowed particulate materials to be classified as a function of their dustiness. It therefore provided very useful information for the control of PM fugitive emissions and for the control of PM exposure levels in occupational environments. It would also enable a standardised classification to be drawn up of the studied materials as a function of their dustiness.

- A great scatter was observed in the dustiness results of the materials used in the ceramic industry, sample dustiness ranging from very low to high. These differences could even be observed for samples of the same material that exhibited different characteristics (different particle size distribution, flowability, etc.). The results highlight the usefulness of the dustiness test for establishing recommendations for the handling of particulate materials.
- Based on the preliminary evaluation of the influence of the variables studied, in the evaluated clays, an increase in moisture content decreased dustiness, except in the ground clays, which exhibited a more complex behaviour. In the materials selected for the study of the variables, increases in flowability, PSD breadth, and particle size raised dustiness, whereas increases in specific surface area decreased dustiness.

6. ACKNOWLEDGEMENTS

El desarrollo de este trabajo ha sido financiado por el Ministerio de Ciencia e Innovación a través del proyecto de investigación "Cuantificación y caracterización de emisiones difusas de partículas en la gestión y transporte de materiales pulverulentos" dentro del Plan Nacional de I+D+I (2008-2011) con referencia CGL 2009-14680-C02-01 y el Fondo Europeo de desarrollo Regional por la cofinanciación del proyecto citado previamente (CGL 2009-14680-C02-01). Por otro lado, la Generalitat Valenciana ha apoyado dicho trabajo con la concesión de una ayuda con referencia (ACIF/2012/111) dentro del Programa Vali+d para investigadores en formación.

REFERENCES

- [1] Monfort, E., Celades, I., Mestre, S., Sanz, V., Querol, X., 2004. Industrial PM_x data processing in ceramic tile manufacturing emissions. *Key Engineering Materials* 264-268, 2453-2456.
- [2] Monfort, E., Celades, I., Gomar, S., Gazulla, M.F., Sanfelix, V., Martín, F., de Pascual, A., Aceña, B., 2006. Control of fugitive particulate emissions in the ceramic industry. In: *Qualicer 2006: IX World Congress on Ceramic Tile Quality*, Castellón, Cámara oficial de comercio, industria y navegación, pp. PBC137-PBC150.
- [3] Monfort, E., Celades, I., Sanfelix, V., Gomar, S., Martín, F., Aceña, B., de Pascual, A., 2007. Experimental fugitive PM₁₀ emission factors from solid bulk handling areas in the open air. In: *CEM 2007: 8th International Conference on Emissions Monitoring: Proceedings*. EMPA, Dübendorf, pp. 245-250.
- [4] Querol, X., Minguillón, M.C., Alastuey, A., Monfort, E., Mantilla, E., Roig, A., Renau, A., Felis, C., Miró, J.V., Artiñano, B., 2007. Impact of the implementation of PM abatement technology on the ambient air levels of metals in a highly industrialised area. *Atmospheric Environment* 41, 1026-1040.
- [5] Santacatalina, S., Reche, C., Minguillón, M.C., Escrig, A., Carratala, A., Nicolás, J.F., Yubero, E., Crespo, J., Alastuey, A., Monfort, E., Miró, J.V., Querol, X., 2010. Impact of fugitive emissions in ambient PM levels and composition. A case study in Southeast Spain. *Science of the Total Environment* 408, 4999-5009.
- [6] Minguillón, M.C., Querol, X., Alastuey, A., Monfort, E., Mantilla, E., Sanz, M.J., Sanz, F., Roig, A., Renau, A., Felis, C., Miró, J.V., Artiñano, B., 2007a. PM₁₀ speciation and determination of air quality target levels. A case study in a highly industrialised area of Spain. *Science of the Total Environment* 372, 382-396.
- [7] Minguillón, M.C., Querol, X., Alastuey, A., Monfort, E., Miró, J.V., 2007b. PM sources in a highly industrialised area in the process of implementing PM abatement technology: quantification and evolution. *Journal of Environmental Monitoring* 9, 1071-1081.
- [8] Moreno, N., Viana, M., Pandolfi, M., Alastuey, A., Querol, X., Chinchón, S., Pinto, J.F., Torres, F., Díez, J.M., Saéz, J., 2009. Determination on direct and fugitive PM emissions in a Mediterranean harbour by means of classic and novel tracer methods. *Journal of Environmental Management* 91, 133-141.
- [9] Hamelmann, F., Schmidt, E., 2003. Methods of Estimating the Dustiness of Industrial Powders- A review. *Kona* No.21.
- [10] UNE EN 15051: 2007. *Atmósferas en los lugares de trabajo-medición del estado pulverulento de materiales a granel-Requisitos y métodos de ensayo de referencia*.

- [11] Mikula, R. J., Parsons, I. S., 1991. Coal dustiness: characterization and control. *Coal Preparation*, 9(3-4), 199-212.
- [12] Chetan P. Pujara, 1997. Determination of factors that affect the generation of airborne particles from bulk pharmaceutical powders. Faculty of Purdue University. Tesis doctoral.
- [13] Plinke, M. A., Maus, R., Leith, D., 1992. Experimental examination of factors that affect dust generation by using Heubach and MRI testers. *The American Industrial Hygiene Association Journal*, 53(5), 325-330.
- [14] Plinke, M. A., Leith, D., Boundy, M. G., Löffler, F., 1995. Dust generation from handling powders in industry. *American Industrial Hygiene Association*, 56(3), 251-257.
- [15] Hsieh, H. P., 1987. Measurement of Flowability and Dustiness of Alumina. *Light Metals 1987*, 139-149.
- [16] Cowherd, C., Grelinger, M. A., Englehart, P. J., Kent, R. F., Wong, K. F., 1989. An apparatus and methodology for predicting the dustiness of materials. *American Industrial Hygiene Association Journal*, 50(3), 123-130.
- [17] Upton, S. L., Hall, D. J., Marsland, G. W., 1990. Some experiments on material dustiness. In *Aerosol Society Annual Conference*, University of Surrey, Guildford, Surrey, UK (April 9-11, 1990).
- [18] Pensis, I., Mareels, J., Dahmann, D., Mark, D., 2010. Comparative evaluation of the dustiness of industrial minerals according to European standard EN 15051, 2006. *Ann Occup Hyg.* 54(2), 204-16.
- [19] Hjemsted, K., Schneider, T., 1996. Dustiness from powder materials. *J. Aerosol Sci*, Vol. 27, Suppl. 1, pp. S485-S486.
- [20] Petavratzi, E., Kingman, S.W., Lowndes, I.S., 2007. Assessment of the dustiness and the dust Liberation mechanisms of limestone quarry operations. *Chemical Engineering and Processing: Process Intensification*, Volume 46, Issue 12, Pages 1412-1423.
- [21] Amorós, J. L., Mallol, G., Campos, B., Orts, M. J., & Bordes, M. C. estudio del comportamiento reológico de diferentes materiales cerámicos pulverulentos. *Qualicer 2008: X World Congress on Ceramic Tile Quality*, Castellón, Cámara Oficial de Comercio, Industria y Navegación, pp. PBC191-PBC205.