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# Research paper Clay hydration mechanisms and their effect on dustiness

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## ABSTRACT

Clays are employed in a wide variety of industries such as ceramic industry, manufacture of paper, rubber, etc. In this sense, it is well known that at industrial processes in which clayey materials are used, such as ceramic industry, in order to carry out some specific stages, the wetting of clays is commonly required. Moreover, it is also long established that wetting is an appropriated measure to reduce particulate matter emissions during clays storage and handling.

The present study was undertaken to assess the influence of moisture on clay dustiness because, though the complex behaviour of the clay-water system has been known since antiquity, the mechanisms involved in clay hydration and their influence on dustiness are still not well understood. To encompass a wide range of specific surface areas, three clays and a kaolin were studied. Chemical and mineralogical analysis of these four raw materials was performed and their particle size distribution, flowability, true density, plastic limit, and specific surface area were determined. Raw materials dustiness was determined using the continuous drop method.

As against what might intuitively be expected, the results showed that the relationship between moisture and dustiness was quite complex and strongly related to the hydration mechanisms. In this regard, to better understand the phenomena involved in the clay hydration process, a specific methodology was developed to estimate the critical points of the clay hydration process (regarding dustiness). This methodology can be readily applied to other clays or even to materials of different nature to predict the optimum moisture and, therefore, it could be employed to propose specific measures which could entail an improvement of outdoor and indoor air quality.

#### 1. Introduction

#### 1.1. Dust generation associated with clay use

Clays are used in a wide variety of industries, such as the ceramics, paper, rubber, and absorbent industries (Mukherjee and Ghosh, 2013). A vast number of workers are thus potentially exposed to particulate matter emitted from dry mechanical processes such as screening and grinding and clay handling and transfer operations (Mukherjee and Ghosh, 2013).

Some industrial activities, such as ceramics manufacture (floor and wall tiles, sanitary ware, tableware, ceramic refractories, etc.) and cement manufacture (grey and white Portland cement, concrete, etc.), are particularly at issue, as many of these industrial goods are clay-based. The raw materials mixture used in manufacturing these products includes considerable amounts of clay, which play a vital part in the different manufacturing process stages. By way of example, Fig. 1 presents a break-down of the use of national clays in Spain (8.5 million - tonnes) in 2012. About 86% of these clays were used in the ceramic

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industry, 10% in cement manufacture, and 4% in other industries.

A parameter of great interest in evaluating, controlling, and minimising the risks associated with the particulate matter emissions generated during clay handling is clay dustiness, i.e. clay tendency to produce dust when handled. In this regard, as the resulting dustiness depends on the apparatus used, standard EN 15051:2013 proposes two test methods: the rotating drum method and the continuous drop method. The test enables the mass fractions of inhalable dust ( $w_I$ ), thoracic dust ( $w_T$ ), and respirable dust ( $w_R$ ) to be determined. The test results 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.

#### 1.2. Water as dust suppressor

It is long established that moisture content increases the interparticle binding forces, which leads to less dust generation (WHO/SDE/ OEH/99.14). Dustiness was observed to decrease with increasing moisture content in different experimental and observational field





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Fig. 1. Use of national clays in Spain, 2012. (Instituto Geológico y Minero de España, IGME).

studies in which water was added to dusty solids to reduce their dustiness (Alwis et al., 1999; Farrugia et al., 1989; Hjemsted and Schneider, 1996; Plinke et al., 1992; Teschke et al., 1999; Westborg and Cortsen, 1990). Industrial examples of the use of water as a dust suppressor include using water as a wetting agent in the bulk outdoor storage of certain dusty materials, wet processing of minerals, and the use of slurries and wetted materials in the ceramic industry (WHO/SDE/OEH/99.14). Water is the most common dust suppressor because of its availability, low cost, and chemical compatibility.

However, other studies (Cowherd et al., 1989; Plinke et al., 1995) found that the dustiness index of some materials rose when moisture content increased slightly, dustiness subsequently decreasing when moisture content was further raised.

Moisture may therefore be considered to have a rather complex effect, which may vary from one material to another, depending on the extent to which water is absorbed internally or is attached to the particle surface (Leith, 1991). That is, its effect depends on the material and on the material's surface properties and hygroscopicity (WHO/SDE/OEH/99.14).

#### 1.3. Clay hydration mechanisms

In order to understand the effect of moisture on clays dustiness, it is needed to know the mechanisms involved in the hydration of clays (Fig. 2). Clay water retention takes place by two mechanisms: adsorption and capillary condensation, and the following domains may be distinguished (Prost et al., 1998):

#### Domain 1

In this adsorption domain, water is first adsorbed on the material's hydrophilic sites (exchangeable cations, edge cations, and surface OH



groups). It can induce the creation of pillars between clay layers, expanding the layers (interlamellar swelling) and eventually, results in a creation of a film of water on surfaces. Capillary condensation at contact points between particles may also occur in this domain.

#### Domain 2

Multilayer adsorption then occurs on "free surfaces", which are either unsaturated pore walls or surfaces of layers or particles that can expand freely. This does not necessarily mean that the entire water structure adsorbed by this process is affected by the surface: only about 2–3 layers of adsorbed water are perturbed by clay surfaces (Prost et al., 1998; Revil and Lu, 2013).

In this regard, Haggymassy (1970) obtained the statistical number of water layers versus relative pressure  $(P/P_0)$  for a wide variety of nonporous adsorbents. Hagamassy classified the obtained results in different groups based on the heat of adsorption of water on the adsorbents, to this regard each group was characterised by its BET constant (C), which is an index linked with the affinity between adsorbate and adsorbent. Hagamassy results can be summarised as follows: for low relative pressures (P/P $_0$  < 0.5) four groups were established (C = 5.2, C = 10-14.5, C = 23, C = 50-200), whilst for high relative pressures  $(P/P_0 > 0.5)$  two groups were distinguished (C = 5.2 and C = 10-200) The C values obtained in our study were in the range 6.9-7.2 and, therefore, the results obtained by Hagamassy for the group with C = 5.2 have been considered the most appropriated for all the range of relative pressures (Table 1). Ormerod and Newman (1983) and Moore and Lockner (2007) postulated that water sorption on the external surfaces of clays was similar to that on reference non-porous oxides (Haggymassy, 1970), so that the relation between relative pressure and the statistical water layers may also be assumed to be valid for porous materials.

#### Domain 3

In this domain, capillary condensation of water in pores must be involved (Benchara, 1991; Carrott et al., 1982; Prost et al., 1998). The capillary condensation phenomenon occurs in pores that correspond to the fabric of hydrated particles.

As the amount of water increases, the added water is attracted by the water already adsorbed at the solid surface by the cohesion forces while tending to minimise the water–air interface area, so that particles start to be held together by liquid bridges at their contact points (**pendular state**). If more water is added, some pores become fully saturated by liquid, while air-filled voids still remain (**funicular state**). Note that the transition between the pendular and the funicular state (5% saturation degree) entails about a threefold increase in granule tensile strength from the pendular state (Oliva, 2015). Water then starts to fill the interparticle voids until all these voids are filled with liquid, but the surface liquid is drawn back into the pores under capillary action (capillary state).

#### Domain 4

This domain is related to the multilayer adsorption process on grain or particle "free surfaces" and to a capillary condensation mechanism in

Table 1Statistical water layers versus relative pressure using the proce-<br/>dure proposed by Hagamassy (C = 5.2).

Relative pressure P/P <sub>0</sub> (%)	Water layers
0.35	1
0.725	2
0.90	3
0.95	4
0.98	5
1	6

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