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Mechanical properties and failure probability of compact agglomerates

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ABSTRACT

Handling of compact agglomerate products such as granules which are typically inhomogeneous porous and mechanically (contact) history-dependent, is usually carried out with excessive costs and energy requirements to achieve merely a satisfactory process efficiency and product quality, owing to their statistically distributed probability of failure (by plastic yield or breakage). Although their failure probability may depend on several material properties and process conditions, qualitative and statistically reliable quantitative information about the influences of such factors other than size is hardly known.

This communication presents a pragmatic analysis of model-based data evaluation of experimental force–displacement and breakage behavior of granules under quasi-static uniaxial compression, that commonly occurs during product processing and handling. The coupled influences of moisture content and loading rate on the distributed micro and macro mechanical properties of granules are described. A comparison of the mechanical properties of fresh and fatigue-affected granules is also reported. Furthermore, the macroscopic breakage and fragmentation pattern recorded using X-ray micro-computed tomography is also presented.

1. Problem description and state-of-the-art

Most of the today's particle-based speciality products of process industries such as pharmaceutical, agricultural, chemical and food industries are produced as agglomerates (i.e., particle–particle semi-permanent compounds of size $0.1 < d < 10$ mm, usually composed of ultrafine to fine cohesive solid powders of size $10 < d < 100$ μm with needful binding additives). Known factors that affect the mechanical product quality of agglomerate products during storage and handling are their non-uniform, anisotropic and (contact) history-dependent mechanical behavior. In addition, insufficient apparatus and system reliability of processing as well as handling units always exist. Moreover, as agglomerate products are handled (transported) and stored as bulk solids, these two conventional unit operations that occur in day-to-day industrial practice are based on scale-up estimations of the continuum (bulk) behavior predicted from single agglomerate behavior. However, due to the ubiquitously existing dispersity of agglomerates and their random packing states within an agglomerate collective (bulk product), these continuum-approach based scale-up estimations suffer from severe limitations. For instance, the compressive strength of a bulk product is always over-estimated, as the content of material flaws (pores, micro-cracks) and voids due to packing states are not taken into account. Due to such a comprehensive list of reasons, in addition to the ubiquitously varying process and environmental conditions, conventional unit operations that involve day-to-day industrial handling of agglomerate products advance with excessive costs and energy requirements to achieve a merely satisfactory process efficiency and product quality.

An efficient and applicable solution to this practical and fundamental problem can be approached by a two-step process, which consists of the *problem diagnosis* followed by the *technological therapy* (see Luding and Tomas [1]). The problem diagnosis begins by clearly studying microscopically — the primary particle–particle, agglomerate–agglomerate and agglomerate–wall interactions to evaluate the micro-mechanical property distribution functions (with respect to material properties and process conditions) and thereby studying the bulk behavior using numerical simulations by the discrete element method (DEM) based on physically accurate and intricate contact models. The technological therapy should subsequently advance by designing suitable processing apparatuses and systems to ensure excellent product quality with merely negligible engineering compromises. It is true, that several practical situations in the three most common mechanical unit operations — comminution, agglomeration and fluidization, have been well-studied using DEM simulations for non-bonded particle/powder collectives (see for instance, Cleary and Sinnott [2], Skrinjar and Larsson [3] and Thornton et al. [4] respectively). Moreover, by scaling down from the continuum level to the microscopic particle level, merely breakage at comminution has been well-studied for a confined collective of bonded particles i.e., a single agglomerate (see for instance, Khanal and Tomas [5]). However, to develop DEM simulations of the dynamics of agglomerate collectives at realistic processing and handling conditions — the

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intrinsic mechanical property distributions with high statistical accuracies have to be evaluated from experimental results of *representative* test conditions and test samples that closely mimic real-time product processing and handling.

The mechanical behavior of agglomerate products from the process industry have been traditionally studied using granules (i.e., compact agglomerates mostly of size[1] $1 \le d \le 3$ mm) as typical test samples and either impact (stressing at one contact) or compression (stressing between two contacts) as typical test conditions. The past decade has seen remarkable progress towards the aforementioned problem diagnosis, especially for granules stressed by compressive loads within the quasi-static deformation range. The decreasing contact strength and surprisingly decreasing granule stiffnesses due to increasing content of inhomogeneties and material flaws (such as pores, micro-cracks, etc.,) with increasing granules sizes have been clearly established (see Refs. [6–8]). Similarly, the decreasing contact stiffnesses and granule strengths due to the increasing lubricating and damaging effects with increasing moisture content present in the granule microstructure have also been clearly established (see Refs. [8,9]). Nevertheless, systematic evaluation of the micro and macro mechanical properties of granules with respect to influencing factors other than size is yet in its infancy.

One such influencing factor regarding which limited information remains, is the loading rate (see also Peukert [10]). It is known, that for dry granules, the load required to initiate plastic micro-yielding of the contact as well as to initiate macro-breakage of the granule varies directly with the rate at which they are loaded [11]. Concurrently, it is also known that the load required to initiate plastic micro-yielding of the contact as well as to initiate macro-breakage of the granule varies inversely with the incompressible moisture content present in the granule microstructure [8,9]. Nevertheless, the mechanical behavior of granules which are simultaneously influenced by both these factors have not been studied as of yet. The primary purpose of this communication is to point out the qualitative and (at least approximately) the quantitative coupled influences of moisture content and loading rate on evolving trends of the mechanical property distribution functions during uniaxial compressive loading of single granules within the quasi-static deformation range. The mechanical property distribution functions represent highly reliable statistical information that can be directly used for numerical DEM modeling of granule systems. This communication also describes the breakage pattern of spherical granules under compression, using X-ray micro-computed tomography (μ-CT) for the first time. Furthermore, the deteriorated mechanical properties of granules (that break after several cycles of repeated uniaxial compressive loading with 90% of load required to initiate breakage in fresh granules, or in other words — granules affected by fatigue) are described.

2. Materials and methods

2.1. Test material

Synthetic zeolitic molecular sieves of the type 4A in the form of granules (Köstrolith® 4AK, Chemiewerk Bad Köstritz GmbH, Bad Köstritz) had been selected as test materials since they are ideal test materials i.e., typical representatives of industrial compact agglomerate products. They are almost spherical, highly hygroscopic, water insoluble and porous with a uniform pore diameter of $d_{pore} = 4.2$ Å (for comparison: the distance between the two hydrogen atoms in a water molecule is 1.5 Å). Furthermore, they can be classified as Geldart D particles for fluidization behavior [12] and besides that, they show elastic–plastic mechanical behavior and brittle breakage [7–9].

The industrial production of these granules is carried out as follows [13]:

- firstly, by mixing fine powders of zeolite (primary particles) and attapulgite (binder),
- followed by pelleting (growth/built-up agglomeration) the mixture with the addition of water (and other appropriate additives) and
- finally, by thermally activating the pelleted intermediates at temperatures $T > 500$ °C.

Thus, the final product possesses an 'onion-like' layered structure[2] with micro-cracks and macro-voids developing between layers due to relative expansion and shrinkage of correspondingly hot and cold layers during thermal activation [8]. Furthermore, since zeolite in itself contains a well-defined pore network, the final product is significantly porous.

2.2. Sample selection and pre-treatment

In order to achieve comparable results with the authors' previous communication [11], the same samples had been used. It is worth to note that two dissimilar size fractions $d_{g,min}$–$d_{g,max} = 1.25$–2.24 and 2.50–4.00 mm had been chosen to validate the independency of granule size in the presented results. The granule size distribution and sphericity had been examined using a dynamic particle size analyzer (Camsizer, Retsch Technologies GmbH, Haan), while the solid and granule densities had been measured using a helium gas pycnometer (Ultrapyknometer 1000, Quantachrome GmbH & Co. KG, Odelzhausen) and a powder pycnometer (GeoPyc 1360, Micromeritics GmbH, Aachen) respectively. Thereby, the porosity was calculated from the relation

$$\varepsilon_g = \frac{V_{pore}}{V_s} = \frac{V_s - V_g}{V_s} = 1 - \frac{\rho_g}{\rho_s} \tag{1}$$

as the mass m_g is constant. The mass-specific surface area was determined by evaluating a single-point from the adsorption isotherm measured by a BET-analyzer (Areameter II, Ströhlein Instruments GmbH & Co., Kaarst) using nitrogen gas at a temperature of $T = -195.15$ °C. For detailed descriptions of each of the measurement techniques and the evaluation procedures followed in this study, the reader is referred to Hintz et al. [14]. Table 1 presents the measured physical and the granulometric properties of the test samples. The binder content X_b, porosity ε_g and sphericity ψ_g values in Table 1, clearly depict the material inhomogeneity, content of flaws and surface roughness respectively.

In order to study the influence of moisture on the material behavior, the samples were moistened using wet air in an air-conditioned climatic chamber maintained at a temperature of $T = 25$ °C and a relative humidity of $\varphi = 95\%$ over a period of $t = 60$ h. Subsequently, a part of the samples were immersed in a water bath for a period of $t = 0.5$ h. After such pre-treatment, the samples had been carefully stored in air-tight containers to prevent any loss of the generated moisture contents. The generated moisture contents X_W (see Table 2) in the climatic chamber is calculated from the mass-increase after moistening as

$$X_W = \frac{m_W}{m_{DS}} = \frac{m_{wet,g} - m_{dry,g}}{m_{dry,g}}, \tag{2}$$

where m_W represents the mass of the water, m_{DS} represents the mass of the dry solid, and $m_{wet,g}$ and $m_{dry,g}$ represent the mass of the granules after and prior moistening respectively. But, the generated moisture content in the water bath is back calculated from the porosity of the granules assuming complete pore saturation ($S = 1$; corresponding to

[1] This size is typical for most industrial granules used as absorbents, detergents, flow additives, fertilizers, catalysts, etc.

[2] An onion-like layered structure is typical of particle products produced by the growth/built-up agglomeration techniques due to the characteristic layer-wise growth processes.