Particuology xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Particuology

journal homepage: www.elsevier.com/locate/partic

Compression curve analysis and compressive strength measurement of brittle granule beds in lieu of individual granule measurements

Matti Järveläinen[a,*], Aaretti Kaleva[a], Annikka Kaitajärvi[b], Jarmo Laakso[a], Ulla Kanerva[b], Erkki Levänen[a]

[a] Department of Materials Science, Tampere University of Technology, PO Box 589, FIN-33101 Tampere, Finland
[b] VTT Technical Research Centre of Finland, Sinitaival 6, PO Box 1300, FI33101 Tampere, Finland

ARTICLE INFO

Article history:
Received 1 June 2015
Received in revised form 28 August 2015
Accepted 3 October 2015
Available online xxx

Keywords:
Granule
Mechanical characterization
Pressing
Compressive strength
Sintering
Compression curves

ABSTRACT

Powders in granulated form are used in various processes to facilitate convenient usage. The durability of the formed granules is a crucial parameter, typically evaluated by the compressive strength of the granules. However, especially for granules with a diameter in the order of tens of microns, statistically relevant testing of individual granules is not a feasible alternative, and in such cases uniaxial bed compression is required.

There has not been consensus on whether uniaxial compression of a granule bed can be used to study the fracture of micron size or brittle granules. In our case study of a bed of sintered kaolinite granules with diameters under 100 μm, we show how the compressive strength of individual granules can be obtained from the compressive measurement of the entire bed by plotting the relative density versus the logarithmic pressure scale.

We compressed the kaolinite powder with different loads; microscopy confirmed that below the analyzed strength the granules are intact, though the granules start to fracture in the curved region on the compression curve. We found that angle-fitting can be used to locate the average compressive strength on the compression curve and to follow the evolution of strength with sintering temperature. The experiments in unison demonstrate that compression curve analysis is applicable for strength analysis of brittle granules.

Introduction

Materials in granular form are used in process industries, among others, in the production of mineral, concrete, metallurgical (Huang, Yi, & Jiang, 2012), pharmaceutical (Yohannes et al., 2015), and food products (Kwan, Wong, & Fung, 2015). Granules are also used as adsorbents, catalysts (Couroyer, Ning, & Ghadiri, 2000; Subero-Couroyer, Ghadiri, Brunard, & Kolenda, 2003), and in ceramic production, pesticides, fertilizers (Leszczuk, 2014), nuclear fuels, and sludges (Antonyuk, Tomas, Heinrich, & Mörl, 2005; Carneim & Messing, 2001). Granulation is needed to improve the flow properties of a powder and at the same time to prepack the powder to increase its processability (Richerson, Richerson, & Lee, 2005). Lower flowability in non-granulated powders as compared to granules is due to spontaneous agglomeration and relatively higher interaction forces between each particle (Briscoe & Özkan, 1997). Granulation also makes it possible to limit a powder's dusting (Leszczuk, 2014), and control properties such as: shape, chemical composition, size distribution, porosity, internal surface area (Antonyuk et al., 2010), and rate of dissolution (Rahmanian, Ghadiri, Jia, & Stepanek, 2009). Granulation also allows the processor to mix a binder with the granules before compaction, in the earliest processing steps (Müller, Russell, & Tomas, 2015; Terpstra, Pex, & De Vries, 1995). The binders provide lubrication and green-state strength prior to densification (Briscoe & Özkan, 1997).

The mechanical durability of a granule determines how well it can endure stresses that are induced during manufacture, e.g., collisions to spray dryer walls, transport, and during use (Antonyuk et al., 2005). However, the granulate is not always the final desired form; for instance in ceramic production a controlled breakage via pressing of the granules is needed for homogeneous products (Terpstra et al., 1995). Should the granules not break in an orderly form during compaction, the end-product will contain voids and local density changes (Honda & Nonaka, 1998). The granule

---

* Corresponding author. Tel.: +358 408490194.
E-mail address: matti.jarvelainen@tut.fi (M. Järveläinen).

http://dx.doi.org/10.1016/j.partic.2015.10.006

Please cite this article in press as: Järveläinen, M., et al. Compression curve analysis and compressive strength measurement of brittle granule beds in lieu of individual granule measurements. *Particuology* (2016), http://dx.doi.org/10.1016/j.partic.2015.10.006

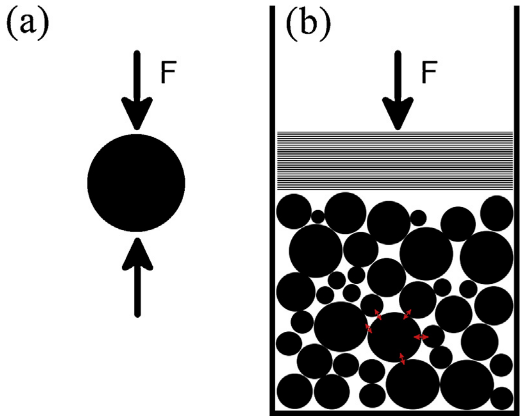

Fig. 1. Compression of (a) a single granule, and (b) a bed of granules. Red arrows show the force distribution of a granule. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

durability is usually tuned with additives, such as binders and lubricants, and careful control of the processing parameters. The durability of powder agglomerates (granulates) can also be important for processes such as injection molding, where powders in their as-synthesized form may aggregate and cause discontinuities in the final product. In such cases, the durability of the agglomerates must be known and accommodated, to ensure that the preprocessing will deagglomerate the powder (Song & Evans, 1994).

Mechanical durability of granules can be analyzed with functional measurements such as impact testing (Antonyuk et al., 2010), Pfosta-apparatus (Leszczuk, 2014), or drop weight testing (Salman & Tomas, 2014). For general analysis, the compressive strength of granules can be modeled (Antonyuk et al., 2010; Müller & Tomas, 2014; Schilde, Burmeister, & Kwade, 2014), and is often measured by compressing a single granule (Cheong, Adams, Hounslow, & Salman, 2009; Müller et al., 2015; Rahmanian et al., 2009; Rahmanian & Ghadiri, 2013), such as in Fig. 1(a). However, measuring a statistically representative amount of single granules may be prohibitive due to large variations in their size, strength, and shape. Very small granules, below 0.5 mm, are particularly difficult to measure even with special tools (Pitchumani, Zhupanska, Meesters, & Scarlett, 2004) or nanoindenters (Raichman, Kazakevich, Rabkin, & Tsur, 2006; Schilde et al., 2014), so these materials are more conveniently measured in packed beds, as shown in Fig. 1(b).

Measurement of a bed instead of individual granules also gives a better understanding of the uniformity of industrially produced granules. The bed strength represents the overall strength of an average granule, although the result is affected by the often-uneven force distribution in the bed, which in turn results from the granule strength, size distribution, and the uniaxial nature of the compression. Still, the same principles apply in bed and single granule compression: size, porosity, and roughness of the granules affects the strength (Antonyuk et al., 2005) together with the granule shape (Pitchumani et al., 2004) and internal imperfections (Subero-Couroyer et al., 2003). In bed compression, the result is also affected by the granule size distribution and granule friction (Kwan et al., 2015; McDowell & de Bono, 2013). Granule strength can be affected by cyclic loading (Salman & Tomas, 2014) in a fatigue mechanism where a repeated force below the granule's strength induces microcracks (lowering its strength (Pitchumani et al., 2004)), or causes hardening (increasing its strength (Antonyuk et al., 2010)); the atmosphere also affects granule strength (Cheong et al., 2009; Müller et al., 2015).

Both measuring methods (bed and single compression) for granule strength are justified. In single-granule compression, a statistically reliable number of granules is needed to measure the average granule strength (Subero-Couroyer et al., 2003); even with easily measured large granules, it is laborious to achieve consistent compressive strength values, although some applicable approaches are now available (Pitchumani et al., 2004; Subero-Couroyer, Ghadiri, Brunard, & Kolenda, 2005). In bed compression, a large number of granules is measured simultaneously, providing the appropriate statistical characteristics on the first experiment; however, predicting the average properties of a single granule from these results is not straightforward due in part to the non-uniform strain that results from uneven load distribution of a randomly packed granule bed and a uniaxial compression tool.

In bed compression, the type of granule also has an effect on the yielding behavior of the bed. Some authors (e.g., Antonyuk et al., 2005, 2010; Aryanpour & Farzaneh, 2015; Briscoe & Özkan, 1997; Meyer & Faber, 1997; Subero-Couroyer et al., 2005) have studied large (over 1 mm) granules or granules that behave elastically or plastically, such as metals (Thyagarajan, Cantin, Kashyap, & Bettles, 2015), where the shear stresses have an important role in bed compression. Even otherwise brittle ceramic powders often include a binder (Carneim & Messing, 2001) in the granules that causes plastic deformation. Given the above, there is a lack of published information on brittle granules, which would break instantaneously without a significant plastic deformation. Here we show, through a case analysis, that bed compression curve-analysis is applicable for brittle granules with a diameter below 100 μm, and we present a systematic approach to interpreting the compression testing results.

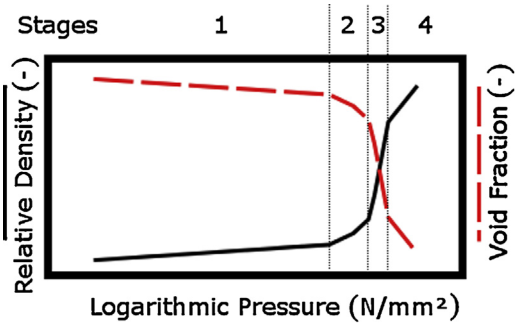

Fig. 2. Schematic illustration of a compression curve showing the four stages.

Theoretical background

Compression stages

The behavior of granules during compression of a bed, including deformation and breakage, exhibits four stages (Briscoe & Özkan, 1997; Mort, Sabia, Niesz, & Riman, 1994) that are shown in Fig. 2. The stages appear in steady-state compression due to the increase in pressure that can be identified using a semi-logarithmic plot; presenting the compression behavior either by the relative density or the void fraction versus the logarithmic pressure, the effect of the pressure increase can be distinguished. The density change is very small until the onset of breakage in stage three, when it becomes logarithmic, enabling a linear fitting on a half-logarithmic scale (Song & Evans, 1994).

During stage 1, the granules deform elastically, pack, and rearrange. The packing density is governed by the shape and size distribution of the granules and influenced by different effects (Kwan et al., 2015), which are beyond the scope of this article. When the rearrangement takes place, the relative density increases as the granules achieve more dense packing.

During stage 2, the compressive strength of the granules is exceeded and they fracture. Some researchers (see especially

Please cite this article in press as: Järveläinen, M., et al. Compression curve analysis and compressive strength measurement of brittle granule beds in lieu of individual granule measurements. *Particuology* (2016), http://dx.doi.org/10.1016/j.partic.2015.10.006