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Compressive behavior of high viscosity granular systems: Effect of particle size distribution

Melissa L. Sweat, Andrew S. Parker, Stephen P. Beaudoin *

School of Chemical Engineering, Purdue University, 480 Stadium Mall Drive, West Lafayette, IN 47907, United States

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ABSTRACT

Granular compressive behavior has been evaluated with respect to particle size distribution. All granules were prepared with a polydimethylsiloxane (PDMS) binder with viscosity ranging from 12 to 2413 Pa·s. All granules were prepared with 90% particulates and 10% binder by mass. Granules were prepared using two unimodal particle size distributions (420–595 μm silica and 105–420 μm silica) and two bimodal particle size distributions. The bimodal size distributions were comprised of 420–595 μm silica and 0–63 μm silica in 75%/25% and 50%/50% mass fractions, where the first percentage refers to the amount of 420–595 μm silica particles included. The granules were compressed with 1, 10, and 100 mm/s velocities. The granules prepared with 420–595 μm silica particles were found to exhibit greater granular strength than those prepared with 105–420 μm particles. These results are uniquely different from those of previous studies which indicate that a smaller size fraction of particles will result in a stronger granule. The addition of a secondary particle size distribution in conjunction with the 420–595 μm silica particles results in a granule with significantly increased strength over the granule prepared with a unimodal distribution. It is suggested that the bimodal size distribution allows an increased number of interparticle contacts per unit volume to increase the granular strength.

1. Introduction

Wet granulation is an essential aspect of many industries, including pharmaceuticals, foods, and agriculture, among others [1,2]. The ultimate goal of the work described here is to understand the impact of particle size distribution on the explosive Composition C-4. C-4 is a wet granular material comprised of ~90% cyclotrimethylenetrinitramine (RDX) mixed with ~10% polymeric binder [3–5]. The RDX used in the explosive composite is a bimodal powder with constituent powders ranging from <44 μm to <2000 μm [3,5]. For these reasons, the effect of particle size distributions on the behavior of wet granular material must be evaluated. As a first step, the effect of the size distribution is studied here using a model, benign material.

Typically, wet granulation involves a dry powder combined with a liquid binder to produce granular material [1,2,6]. Wet granulation is frequently considered to occur *via* three principle processes: nucleation and binder distribution, consolidation and growth, and attrition and breakage [1,2,6–15]. Of these, the attrition and breakage are arguably the least understood [1,2]. The work presented here is largely focused on the breakage of wet granules, where the two primary modes of failure are considered plastic and brittle [11]. Granules undergoing plastic deformation will deform largely without the presence of any principle cracking, while brittle failure is easily identified by the presence of a major crack or cracks [11,16]. In either case, there is no significant elastic recovery of the material.

Studies of dynamic wet granular failure have focused on binder viscosity, interfacial parameters (surface tension and contact angle between the binder and primary particles), and particle size and shape [1,2,11,14–43]. Much of this work indicates that an increase in binder viscosity will result in an increase to granular strength [1,2,11,15–39, 44]. Additionally, studies of spherical particles indicate that an increase in filler particle size is predicted to result in a decrease in particle strength due to the reduced number of interparticle contacts per unit volume [2,11,18,19,45,46]. However, very few studies exist to create a comprehensive model to describe the behavior.

Further, some use discrete element modeling (DEM) to describe the behavior of granular materials [1,6,47–49]. While strides have been made using DEM for the breakage of agglomerates on impact, these typically make a few assumptions that are not valid here. Because these studies are limited to impact testing, the deformation rates are on the order of 10 m/s or greater [1,6]. As will be discussed in the following sections, the compression testing in this work occurs between 1 mm/s and 100 mm/s – several orders of magnitude less than in the impact studies. Additionally, DEM studies have historically focused on granules comprised of smooth, rigid particles, and any binder considered is often of very low viscosity (on the order 0.1 Pa·s) [1,6,47–49]. The system examined in this study incorporates granules comprised of realistic

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* Corresponding author.
E-mail address: sbeaudoi@purdue.edu (S.P. Beaudoin).

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particle size distributions with particulate roughness and non-sphericity, as well as some granules comprised of bimodal particle size distributions. The binder in this work is also highly viscous, ranging from ~10 to 2500 Pa·s.

In lieu of a robust, computationally expensive model, studies have been completed to describe the compressive behavior of granules as a function of dimensionless groups, as shown in Eq. (1) [11,14,16,18–21]. Here, σ_p is defined as the peak flow stress; d_{32} is the specific surface mean particle size (or Sauter mean); γ_{lv} is the binder surface energy; θ is the liquid-vapor contact angle; μ is the binder viscosity; and $\dot{\varepsilon}$ represents the strain rate. μ_f is the coefficient of internal friction; S is the granule liquid saturation; and Φ is the granule packing fraction. Because the coefficient of internal friction, granule liquid saturation, and granule packing fraction are typically held constant in these studies, the authors commonly consider the granular compressive behavior to be a function of the left side of the equation (the normalized peak flow stress, Str^*) and the first term of the right side of the equation (the dimensionless capillary number, Ca, which is the ratio of the viscous and surface forces in the system). These are defined individually in Eqs. (2) and (3).

$$\frac{\sigma_p d_{32}}{\gamma_{lv}\cos\theta} = f\left(\frac{\mu\dot{\varepsilon}d_{32}}{\gamma_{lv}\cos\theta}, \mu_f, S, \phi\right) \tag{1}$$

$$Str^* = \frac{\sigma_p d_{32}}{\gamma_{lv}\cos\theta} \tag{2}$$

$$Ca = \frac{\mu\dot{\varepsilon}d_{32}}{\gamma_{lv}\ \cos\theta} \tag{3}$$

The normalized peak flow stress can be considered to be the ratio of the normal forces required to cause granule breakage to the interfacial forces resisting movement of the binder away from the solid. The capillary number is the ratio of viscous forces resisting shear within the binder to the solid-binder adhesion interactions resisting motion of the binder past the solids. At low Ca in these systems, failure is classified as semi-brittle and is considered to be independent of strain rate and binder viscosity, and the behavior transitions to plastic failure at high Ca. Here, catastrophic failure does not typically occur in that no clear plane of separation is observed. Instead, the compact deforms/flows under the applied load. Significant void creation occurs without the presence of observable "cracks" where the material has catastrophically deformed. In practice, the granular material may be held as a single deformed piece simply due to the behavior of the compact being dominated by viscous forces. Large cracks do not generally form, as the viscosity of the binder allows the applied load to be dissipated through the system with only catastrophic deformation – not failure. Prior studies showed that granules prepared with very high viscosity binders can often be removed as a single piece following compression experiments [44]. Any void creation appears at the exterior of the granule only. Accordingly, the designation "failure" seems misleading for these materials – rather, catastrophic deformation is a more apt description for the behavior observed.

Prior work largely focuses on granules containing powders with unimodal particle size distributions of small particles [1,2,11,15,17–39,50]. This study seeks to expand the previous work to include systems containing powders with unimodal distributions of large particles, as well as systems containing powders with bimodal size distributions. Generally, particles that are relatively uniformly packed will form stronger granules compared with granules exhibiting chaotic packing due to the increased number of interparticle contacts per unit volume resisting motion [6,48,51–55]. The change in behavior due to particle packing has been observed experimentally and through DEM simulations, where the addition of a second, smaller size fraction causes an increase to both the bulk powder density and the overall strength of the powder [56,57]. By incorporating a bimodal particle size distribution where the large particles are at least an order of magnitude larger than the small ones, the particle packing should be optimized as small particles fill the interstitial voids created by the packing of large particles. This is expected to result in a significant increase to the granular strength observed.

2. Experimental

Several studies exist to study the effects of size distribution on the compressive behavior of granular systems [1,11,14,16,18–21,36,40–42,58]. These have largely focused on granules comprised of relatively small spherical particles with a unimodal size distribution bound with relatively low viscosity binders. The overall effect of high viscosity binders has rarely been discussed [44]. The research presented here seeks to explore the effect of large particles (~225 and ~340 μm) and bimodal size distributions using mixtures of ~340 μm particles with ~40 μm particles.

2.1. Materials

Three size fractions of silica particles were used in this work (Fig. 1): 420–595 μm silica (large), 105–420 μm silica (small), and 0–63 μm silica (very small). The 420–595 μm silica was supplied by VWR, and Sigma Aldrich provided the 420–595 μm and 0–63 μm silica powders. These powder sizes were chosen based on their similarity to the particle sizes used in the creation of Composition C-4 [3,5]. A variety of powder size characteristics were evaluated *via* a Malvern Mastersizer 2000 Light Diffraction Particle Size Analyzer, including the surface weighted mean (Sauter mean diameter, x_{32}), the volume weighted mean (x_{43}), the 10th percentile diameter (x_{10}), the median diameter (x_{50}), the 90th percentile diameter (x_{90}), and the circularity. These data are shown in Table 1, along with the advertised sieve range and size range that will be used through the remainder of this work.

The circularity (C) may be calculated according to Eq. (4), where the area (A) and perimeter (P) have been obtained through image processing *via* ImageJ [59]. For comparison, spherical glass ballotini are often estimated to have circularity ~0.9–0.95, while lactose (a powder often studied due to its highly irregular shape) has a circularity of ~0.72 [11]. Accordingly, the 105–420 μm silica is considered highly irregular, while the 420–595 μm silica is significantly more uniform. The cumulative size distributions for the unimodal distributions are shown in Fig. 2. The cumulative and normalized distributions of the bimodal particle fractions are given in Fig. 3. The first bimodal distribution is comprised of 75% 420–595 μm silica mixed with 25% 0–63 μm silica by mass. The second bimodal distribution was prepared with 50% each of the 420–595 μm and 0–63 μm silica particles by mass. Granules were made by mixing each of the powders with each of the eight polydimethylsiloxane (also referred to as PDMS or silicone oil) binders. These binders had viscosities of 12, 29, 58, 97, 289, 579, 965, and 2413 Pa·s. Because PDMS is a Newtonian fluid, the viscosity of each binder was assumed to remain constant at room temperature (the temperature of the study) over the duration of the study. Additionally, the surface energy of silicone oil is 0.0228 N/m, and is assumed independent of viscosity [60]. The binder for Composition C-4 is non-Newtonian (as described

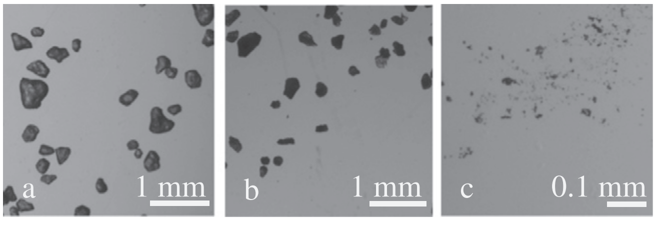

Fig. 1. Optical microscopy images of the 420–595 μm silica (a), 105–420 μm silica (b), and 0–63 μm silica (c) particles.