Powder Technology 287 (2016) 431–438

Contents lists available at ScienceDirect

Powder Technology

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

Experimental and theoretical studies on compressive deformation characteristics of particle aggregates in water

Masaki Kubo [a,*], Ryutaro Ishibashi [a], Ken-ichi Sugioka [b], Takao Tsukada [a], Osamu Koike [c], Masahiro Fujita [d]

[a] Department of Chemical Engineering, Tohoku University, 6-6-07, Aramaki, Aoba-ku, Sendai 980-8579, Japan
[b] Department of Mechanical Systems Engineering, Toyama Prefectural University, 5180 Kurokawa, Imizu-shi, Toyama 939-0398, Japan
[c] Department of Chemical System Engineering, The University of Tokyo, Yayoi, Bunkyo-ku, Tokyo 113-8656, Japan
[d] Department of Mathematics, Josai University, 2-3-20, Hirakawa-cho, Chiyoda-ku, Tokyo 102-0093, Japan

ARTICLE INFO

Article history:
Received 5 December 2014
Received in revised form 3 August 2015
Accepted 5 October 2015
Available online 13 October 2015

Keywords:
Particle aggregate
Compressive deformation
Confocal laser scanning microscope
Discrete element method
Frictional coefficient

ABSTRACT

The compressive deformation characteristics of a single aggregate of polystyrene particles were studied both experimentally and theoretically. Experimental apparatus to measure the mechanical characteristics of a single aggregate with in situ observation of the compression behavior of the aggregate was constructed. Then, the time variations of the compressive force and deformation behavior of the aggregate during simple compression were investigated. In addition, discrete element method (DEM) simulations were carried out to numerically demonstrate the compression behavior of an aggregate between two planes, where the time variation of the compressive force of the aggregate as well as the area strain of each particle in the aggregate can be predicted. The largest spike-like peak of the compressive force appeared at the stage where an aggregate with a hexagonal bilayer structure was formed, as observed experimentally. The compressive force of the aggregate increased as the number of particles in the aggregate increased or the diameter of the primary particles decreased. An increase in the frictional coefficient between primary particles caused an increase in the compressive force of the aggregate for the entire period of compression, but the frictional coefficient between a particle and a wall did not affect the compressive deformation behavior of the aggregate.

1. Introduction

Acquiring correct knowledge of the aggregation behaviors of fine particles in a liquid shear flow and of the mechanical characteristics of particle aggregates in the liquid phase is important for the understanding and control of product behaviors in several processes such as fine-particle wet manufacturing, polishing with abrasive slurries, and solid–liquid separation. In the chemical–mechanical polishing (CMP) process used to planarize metal and dielectric layers to achieve multilayer metallization in the microelectronics industry, for instance, it is believed that large particles, i.e., aggregates of primary CMP slurry particles, are one of the main reasons for the formation of scratches and pitting on polished wafer surfaces [1–3]. Therefore, the aggregates in CMP slurries should be disintegrated into monosized particles before the polishing process. In contrast, in solid–liquid separation processes, such as filtration and gravity settling, it is desirable that solid particles agglomerate to increase their sizes [4–6]. Therefore, it is necessary to inhibit the generation of mechanical forces that cause aggregates to disintegrate.

To determine whether the formed aggregates break under the process conditions, information on the mechanical characteristics and breakage strength of an aggregate itself in the liquid phase is crucial; thus, equipment to measure the deformation behaviors and mechanical properties of an aggregate itself should be established. However, there have been few studies in which the mechanical properties of a single aggregate in the liquid phase were investigated, although studies on the mechanical properties of a single aggregate or granulate performing compression tests in the gas phase have been reported [7–11]. Zhang et al. [12] developed an apparatus for the measurement of compression forces of a single aggregate based on a micromanipulation technique, and measured the breaking forces of single polystyrene latex aggregates formed at the isoelectric point using a vortex mixer and Brownian motion. They demonstrated from the experimental results that the method of bringing primary particles together, i.e., the method of preparing aggregates, affects the breakage strength of the aggregate through interparticle forces, such as van der Waals force, and the aggregation structure. However, their measurements were carried out using only one primary particle size and at relatively small size of the aggregates, where the mean diameter of the aggregates was approximately twice that of the primary particles. In addition, the compressive behaviors of aggregates have not been observed in situ.

On the other hand, numerical simulation is also one of the methods used to investigate the breakage behavior of an aggregate. Recently, discrete element method (DEM) simulations have been widely used to analyze the compression behavior of a single dry aggregate in the

* Corresponding author.
E-mail address: kubo@pcel.che.tohoku.ac.jp (M. Kubo).

http://dx.doi.org/10.1016/j.powtec.2015.10.010
0032-5910/

gas phase [13–16]. For instance, Golchert et al. [14] studied the effect of the aggregate shape and structure on the mechanisms and extent of breakage of dry aggregates under a compressive load using the DEM, where the structure of an actual granule characterized by X-ray microtomography was used as the structure of one of the aggregates in the simulation. However, all the above numerical studies were on compression tests of a dry aggregate with a relatively large diameter.

In this work, to investigate the compression behavior of a single aggregate in the liquid phase, experimental apparatus to measure the mechanical characteristics of a single aggregate with in situ monitoring of the compression behavior of the aggregate was first constructed. Then, using this apparatus, the compression behavior and breakage strength of an aggregate with a size of around 10 μm, which was composed of polystyrene latex particles with a diameter of 1 or 2 μm, in a water droplet on a glass substrate were investigated. However, because of a limitation in the measurement, i.e., the non-detection of extremely small compressive forces acting on the aggregate in the early stage of compression, not all the compression behavior up to the breakage of the aggregate could be measured. Also, it is difficult to visualize the displacement of each particle inside the aggregate during compression. Therefore, numerical simulations have been performed to investigate the compression behavior of an aggregate in detail, using DEM, a numerical technique to model the three-dimensional motion of particles in the liquid [17,18]. Here, the effects of the primary particle size, the aggregate size, the frictional coefficients between particles and between a particle and a wall on the compression behavior of a single aggregate between two planes were investigated.

2. Experimental section

2.1. Experimental apparatus and procedure

The mechanical characteristics of a single aggregate of fine particles during compression were measured using similar experimental apparatus to that developed by Zhang et al. [12]. Fig. 1 shows a schematic diagram of the apparatus constructed in this work. A suspension droplet containing aggregates was placed on a microscope cover glass mounted on an *XYZ*-axis motorized stage (Mark-204-MS, Sigma Koki Co., Ltd.), and a target aggregate in the droplet was chosen in the field of view of a confocal laser scanning microscope (CLSM) of 632.8 nm (1LM15H, Lasertec Co., Ltd.) with a 60× water immersion lens. Then, the single aggregate in the droplet on the cover glass was compressed using a cylindrical glass probe with a tip diameter of less than 100 μm, which was moved downward at a constant speed of 0.2 μm/s by a micromanipulation technique (FC-401, Sigma Tech. Co., Ltd.). The compressive force imposed on the aggregate was measured through a force transducer (406A, Aurora Scientific Inc.) to which the glass probe was attached. The voltage signal output by the transducer was recorded through a digital multimeter (R6871E, Advantest Co., Ltd.) attached to a personal computer at a sampling interval of 50 ms, in which the voltage signal was transformed in the compressive force following the voltage–force relationship measured in advance and the force versus time curve was drawn. In addition, the deformation behavior of the aggregate during compression was observed from below through the cover glass using the CLSM. The images of aggregates recorded using a CCD camera were captured on a personal computer, and then the size and projected area of the aggregates were analyzed using WinROOF (MITANI Co., Ltd.). All the experiments were carried out at room temperature.

To fabricate the glass probe for the compression of the aggregate, part of a glass rod with a diameter of approximately 500 μm was etched by hydrofluoric acid, as shown in Fig. 2(a), to obtain a tip diameter of less than 100 μm, and then the tip surface was optically polished. Fig. 2(b) shows polished tip surface, which appears to be sufficiently smooth to compress the aggregate. The tip diameter of the glass probe was between approximately 50 and 100 μm.

The buoyancy force, which was evaluated from the maximum length of the probe immersed in the water droplet in the experiment, was 4.16×10^{-4} μN, and the hydrodynamic drag force due to the motion of the probe was approximately 0.1 μN. Thus, both can be ignored owing to their small magnitude compared with the compressive force. In addition, the surface tension acting on the side wall surface of the probe was calibrated, i.e., removed as a background signal, because it was a fixed value during compression.

2.2. Sample preparation

A suspension of polystyrene latex particles of 1 or 2 μm (±1%) diameter (Polysciences., Inc.) was used in the experiments, where uniform

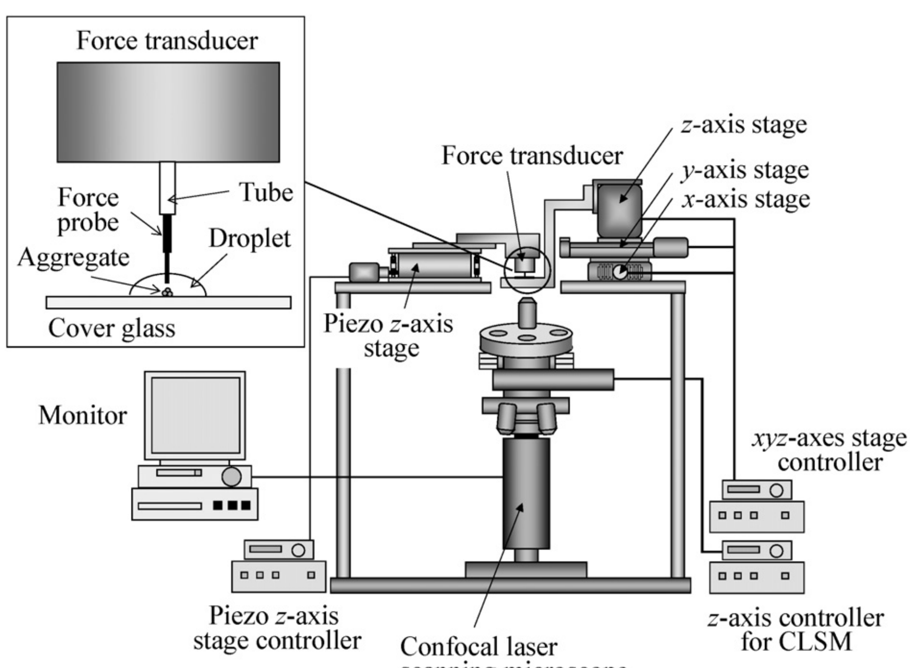

Fig. 1. Experimental apparatus.