Earth and Planetary Science Letters 433 (2016) 232–240

Contents lists available at ScienceDirect

Earth and Planetary Science Letters

www.elsevier.com/locate/epsl

# Experimental volcanic ash aggregation: Internal structuring of accretionary lapilli and the role of liquid bonding

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ARTICLE INFO

*Article history:*
Received 5 August 2015
Received in revised form 4 November 2015
Accepted 6 November 2015
Available online 1 December 2015
Editor: T.A. Mather

*Keywords:*
volcanic ash
aggregation
accretionary lapilli
experimental volcanology
fluidized bed technology

ABSTRACT

Explosive volcanic eruptions can release vast quantities of pyroclastic material into Earth's atmosphere, including volcanic ash, particles with diameters less than two millimeters. Ash particles can cluster together to form aggregates, in some cases reaching up to several centimeters in size. Aggregation alters ash transport and settling behavior compared to un-aggregated particles, influencing ash distribution and deposit stratigraphy. Accretionary lapilli, the most commonly preserved type of aggregates within the geologic record, can exhibit complex internal stratigraphy. The processes involved in the formation and preservation of these aggregates remain poorly constrained quantitatively. In this study, we simulate the variable gas-particle flow conditions which may be encountered within eruption plumes and pyroclastic density currents via laboratory experiments using the *ProCell Lab System*® of Glatt Ingenieurtechnik GmbH. In this apparatus, solid particles are set into motion in a fluidized bed over a range of well-controlled boundary conditions (particle concentration, air flow rate, gas temperature, humidity, liquid composition). Experiments were conducted with soda-lime glass beads and natural volcanic ash particles under a range of experimental conditions. Both glass beads and volcanic ash exhibited the capacity for aggregation, but stable aggregates could only be produced when materials were coated with high but volcanically-relevant concentrations of NaCl. The growth and structure of aggregates was dependent on the initial granulometry, while the rate of aggregate formation increased exponentially with increasing relative humidity (12–45% RH), before overwetting promoted mud droplet formation. Notably, by use of a broad granulometry, we generated spherical, internally structured aggregates similar to some accretionary pellets found in volcanic deposits. Adaptation of a powder-technology model offers an explanation for the origin of natural accretionary pellets, suggesting them to be the result of a particular granulometry and fast-acting selective aggregation processes. For such aggregates to survive deposition and be preserved in the deposits of eruption plumes and pyroclastic density currents likely requires a significant pre-existing salt load on ash surfaces, and rapid aggregate drying prior to deposition or interaction with a more energetic environment. Our results carry clear benefits for future efforts to parameterize models of ash transport and deposition in the field.



## 1. Introduction

Volcanic ash, fragments of juvenile lava, crystals and/or older rock units less than two millimeters in diameter (Fisher, 1961), is produced in large quantities during explosive volcanic eruptions. Fine volcanic ash (particle diameter, $(p_d) < 63$ µm) can be distributed far away from the eruptive vent by ash transport processes, where it may cause a plethora of hazards and impacts (Dingwell et al., 2012). Under certain conditions within eruption plumes and pyroclastic density currents, ash particles can cluster together to form ash aggregates that range from micrometers to centimeters in size. Aggregates exhibit different aerodynamic properties than the individual ash grains which comprise them, promoting their 'premature' sedimentation from the plume and decreasing their residence time within the atmosphere, relative to the individual particles in isolation (Le Roux, 2014). Thus, ash aggregation may influence ash distribution (e.g., Folch et al., 2010) and deposit stratigraphy (e.g., Durant et al., 2009).

Ash aggregates can be characterized using the terminology of Brown et al. (2012); this prior study defined two classes of ash

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http://dx.doi.org/10.1016/j.epsl.2015.11.007

aggregates, particle clusters (*PC*) and accretionary pellets (*AP*). Particle clusters are sub-divided into ash clusters (*PC1*) and coated particles (*PC2*), while *APs* can be divided into poorly-structured pellets (*AP1*), accretionary pellets (*AP2*) and liquid pellets (*AP3*). However, although field observations have evidenced the formation of all of these subtypes, the aggregates most commonly preserved within the geologic record tend to be *AP1* or *AP2*. The *AP1* aggregates are normally spherical or sub-spherical, ranging in diameter from several 100 µm to a few mm, and generally show poor internal structure. The *AP2* aggregates are similarly shaped, and can also grow to sizes of several mm; they consist of a relatively coarse grained core and (possibly several) fine grained rims.

The presence of the *AP1* and *AP2* aggregates in deposits evidences their comparative resilience to the highly energetic processes related to eruption dynamics and sedimentation processes. This preservation is not universal, as field studies have also documented shattered fragments of accretionary lapilli in deposits (Brown et al., 2010). However, our capacity to interpret the presence, absence and particular features (e.g., internal stratigraphy or granulometry) of aggregates within deposits as being indicative of particular eruption features or characteristics is limited. This stems from the absence of a quantitative understanding of the physical and/or chemical properties and in-plume boundary conditions which drive aggregate formation, disaggregation and the capacity of aggregates to survive deposition and be preserved in ash deposits.

The drivers on aggregate formation may relate to the mechanisms of both ash adhesion and aggregate growth. Particle adhesion is likely a product of three major forces, 1) electrostatic bonding, 2) Van der Waals forces and 3) liquid bonding. Although electrostatic charging of volcanic plumes is a well-known phenomenon (Lane and Gilbert, 1992; Cimarelli et al., 2013) that can lead to dry aggregation (e.g. Taddeucci et al., 2011; Del Bello et al., 2015), it is neglected in this study. With maximum surface charges of $\pm 10^{-5}$ C m$^{-2}$ observed for particles in volcanic plumes (Gilbert et al., 1991), the electrostatic attraction and binding potential of ash particles is very low and even negligible in the presence of a liquid binder, as implied by experiments in fluidized beds with other particles (Randolph, 1988; Liu and Cameron, 2001; Saleh and Guigon, 2006). In order to build large ash aggregates stable enough to be preserved in the geologic record, we consider that liquid bonding may be the most important aggregation mechanism.

Previous in-field (Trusdell et al., 2005; Branney et al., 2008; Bonadonna et al., 2011), numerical (Costa et al., 2010; Folch et al., 2010) and laboratory studies (Gilbert and Lane, 1994; Schumacher and Schmincke, 1995; Van Eaton et al., 2012) have investigated the role of liquid bonding. Vibratory pan aggregation techniques, (Schumacher and Schmincke, 1995; Van Eaton et al., 2012) have successfully reproduced ash aggregates with characteristics (bulk diameter, density or granulometry) linearly dependent on humidity or wetting, until an 'overwetting' threshold was reached, whereafter, liquid 'mud' droplets (*AP3*) were formed (e.g. Gilbert and Lane, 1994). However, the influence of liquid bonding on ash aggregate formation remains only partially constrained. For example, although laboratory experiments have generated particle clusters (*PC*), the complex internal structures typical for *AP2* aggregates have yet to be reproduced. Furthermore, the effect of additional variables such as the surface tension and viscosity of the binding liquid (e.g. Kueppers et al., 2011), which in volcanic systems may be commonly comprised of condensates/solutions of co-erupted gases (e.g., $H_2O_{(g)}$, $SO_{2(g)}$, $HCl_{(g)}$, $HF_{(g)}$), have yet to be investigated.

The preservation of ash aggregates within ash deposits may be contingent on the establishment of strong interparticle binding mechanisms within the aggregate (e.g., cementation of solid bridges). Cementation may be driven by the interstitial precipitation of various sulphate and halide salts during the evaporation of the binding liquid. This process of cementation has been invoked following investigations of several field deposits (Tomita et al., 1985; Gilbert and Lane, 1994; Brown et al., 2010). The specific chemistry and abundance of these salts may depend both on the pH of the binding liquid and its capacity to dissolve or corrode the ash surface, and on the presence of pre-existing salts emplaced by higher temperature gas-ash interactions (cf. Witham et al., 2004; Ayris and Delmelle, 2012).

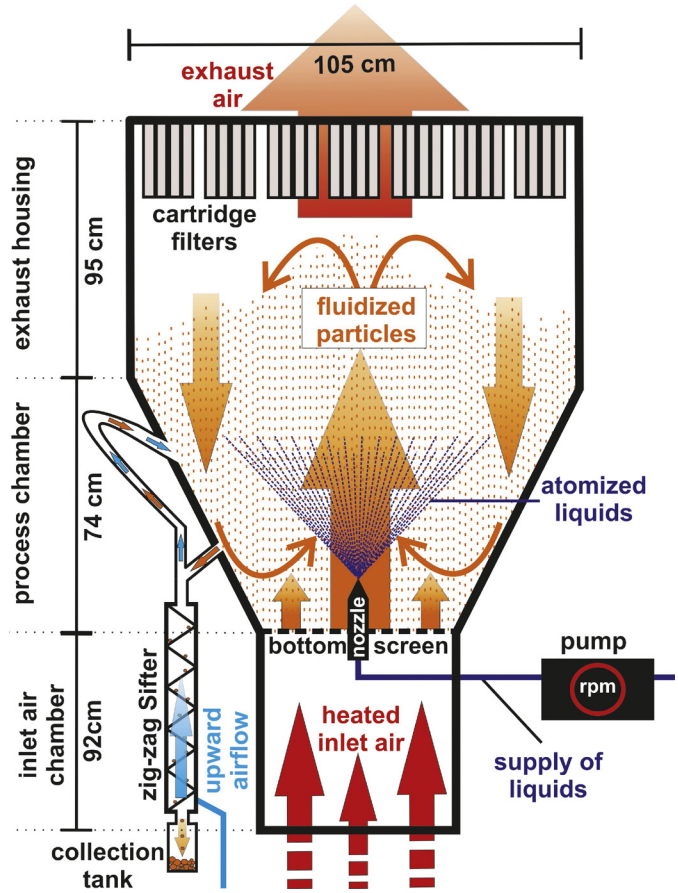


**Fig. 1.** Glatt ProCell® Lab System; solids are deposited on the bottom screen. Heated air flows from the inlet air chamber through the screen into the process chamber and drags particles upwards (fluidization). A nozzle in the center of the bottom screen sprays liquid into the fluidized particles. Liquid droplet size and spray rate are controlled manually. Topside cartridge filters separate fine particles from the exhaust air (exhaust chamber). Aggregates fall into the sifter through a contraflow air stream into a collection tank.

In this mechanistic study we present results of laboratory investigations on the formation and recovery of *AP1* and *AP2*-type ash aggregates. We investigated ash aggregation within fluidized beds, commonly utilized in industrial sectors, in particular in the food, animal feed, pharmaceutical, fertilizer, detergent and mineral processing industries, for investigation or generation of aggregates. Fluidized bed systems transform a granular material from a static (i.e. solid-like) to a dynamic (i.e. fluid-like) state, promoting aggregate formation under precisely-controlled conditions such as humidity, granulometry, air flow and temperature (Salman et al., 2006). In this study, we pioneer the use of this technology for our mechanistic investigation of volcanic ash aggregate formation, and provide new insights into the variables which permit the formation and survival of ash aggregates.

## 2. Methodology

For this study, we use the ProCell® Lab System (Fig. 1) by Glatt Ingenieurtechnik GmbH, Weimar, Germany. The ProCell® Lab