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# THE ORIGIN OF PELLETAL LAPILLI IN EXPLOSIVE KIMBERLITE ERUPTIONS

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#### Abstract

Kimberlites are volatile-rich magmas from mantle depths in excess of 150 km and are the primary source of diamonds. Kimberlite volcanism involves the formation of diverging pipes or diatremes, which are the locus of high-intensity explosive eruptions. A conspicuous and previously enigmatic feature of diatreme fills are 'pelletal lapilli' well-rounded clasts that consist of an inner 'seed' particle with a complex rim, thought to represent quenched juvenile melt. Such clasts are widely documented in a range of pyroclastic successions on Earth. New observations of pelletal lapilli show they coincide with a transition from magmatic to pyroclastic behaviour, thus offering fundamental insights into eruption dynamics and constraints on vent conditions. We provide strong evidence that pelletal lapilli form by fluidized spray granulation - a coating process used widely in industrial applications. We propose that pelletal lapilli are formed when fluid volatile-rich melts intrude into earlier volcaniclastic infill close to the diatreme root zone. Intensive degassing produces a gas jet in which locally-scavenged particles are simultaneously fluidized and coated by a spray of low-viscosity melt. Most fine particles are either agglomerated to pelletal coatings or ejected by powerful gas flows. This mechanism has significant implications for diamond exploration, as multi-stage intrusions result in variable diamond grade and size distributions. A similar origin may apply to pelletal lapilli in other alkaline volcanic rocks including carbonatites, kamafugites and melilitites.

#### Introduction

Most volcaniclastic kimberlites contain ubiquitous yet poorly understood composite particles termed 'pelletal lapilli' (Clement and Skinner, 1985; Mitchell, 1986; Sparks et al., 2006; Wilson and Head, 2007; Gernon et al., 2008). These are defined as discrete sub-spherical clasts with a central fragment, mantled by a rim of probable juvenile origin (Mitchell, 1986). These clasts have previously been attributed to incorporation of particles into liquid spheres in the rising magma (Junqueira-Brod et al., 1999; Wilson and Head, 2007) and rapid unmixing of immiscible liquids (Lloyd and Stoppa, 2003). However, these models fail to explain many aspects of their internal structure, composition and abundance in pyroclastic intrusions. Pelletal lapilli have been identified globally in a wide range of other alkaline volcanic rocks including carbonatites (Stoppa, 1996), kamafugites (Junqueira-Brod et al., 1999; Stoppa et al., 2002), melilitites (Stoppa, 1996; Lloyd and Stoppa, 2003) and orangeites (Mitchell, 1995). They have also been referred to as 'tuffisitic lapilli' (Stoppa, 1996), 'spherical lapilli' (Keller, 1981) and 'spinning droplets' (Junqueira-Brod et al., 1999, 2005). Pelletal lapilli share similar properties to particles formed during industrial granulation processes, but such processes have not previously been considered in a geological context.

Fluidized spray granulation is widely used in industrial engineering to generate coated granules with specific size, density and physicochemical properties (Rajniak et al., 2007). The mechanism involves continuous injection of atomizable liquids, solutions or melts into a powdery fluidized bed (Zank et al., 2001), which produces a dispersion of larger coated granules that are simultaneously dried by the hot fluidizing gas (Christoph-Link and Schlünder, 1997; Zank et al., 2001). When gas flows upwards through particles, the point of minimum fluidization  $(U_{mf})$  occurs when the flow velocity (U) is sufficiently high to support the weight of particles without transporting them out of the system (Davidson and Harrison, 1963). Fluidized spray granulation is a characteristically slow and steady growth process, producing uniform well-rounded particles with a concentric lavered structure (Christoph-Link and Schlünder, 1997).



#### **Field Observations**

Pelletal lapilli occur prominently in two of the world's largest diamond mines, Venetia (South Africa) and Letsengla-Terae (Lesotho), both well-exposed, extensively surveyed (Stripp et al., 2006; Walters et al., 2006; Gernon et al., 2008) and economically significant localities. The Venetia K1 diatreme (Fig. 1a) is dominated by massive volcaniclastic kimberlite, MVK, a characteristically well-mixed lithofacies comprising serpentinized olivine crystals and a polymict range of lithic clasts (Walters et al., 2006; Gernon et al., 2009). The formation of MVK has been attributed to fluidization (Sparks et al., 2006; Wilson and Head, 2007; Gernon et al., 2008), the scale and context of which is heavily debated (Brown et al., 2008; Gernon et al., 2009).



Fig. 1: Simplified geological maps of the Venetia and Letseng kimberlite pipes. **a.** Venetia K1 is dominated by massive volcaniclastic kimberlite (MVK) with subordinate marginal breccias, sediment-bearing volcaniclastic kimberlite and coherent kimberlite lithofacies. The ~15 m wide pelletal-lapilli intrusion (PLI) occurs near the diatreme centre, where it cross-cuts MVK and is closely associated with numerous minor late-stage dykes. **b.** The Letseng Satellite pipe is also dominated by MVK; pelletal lapilli are confined to a northern circular pipe ~100 m wide (modified after Palmer et al., 2008).

Pelletal lapilli (Figs. 2a-b) are confined to a narrow (10 - 15 m diameter), discordant and lenticular body in the centre of K1 (Fig. 1a). Although pipe-like, we refer to these features as pyroclastic intrusions to avoid confusion with the large-scale (0.5 - 1 km diameter) pipes or diatremes in which they occur. Field and drill-core data suggest that the intrusion is a steep-sided tapering cone, associated with numerous late phlogopite-rich dykes. The intrusion is characteristically

structureless, clast- to matrix-supported and poorly to moderately sorted. It contains abundant (90 vol.%) coated lapilli- to bomb-sized clasts (Fig. 2a-b), ranging in diameter from 0.2 - 100 mm (mean = 9.4 mm; Fig. 3a). These pelletal lapilli comprise a sub-angular lithic clast or olivine macrocryst as their core surrounded by a variably thick coating (generally <1 cm); typically this coating comprises olivine-phlogopite-spinel bearing kimberlite with a heavily altered groundmass containing amorphous serpentine and talc. A concentric alignment of crystals is commonly developed in the coating around the core (Fig. 2b). In some cases, the pelletal coatings appear to have partially coalesced.



Fig. 2: Photographs of pelletal lapilli from southern African kimberlites and a synthetic analogue. a. Exposure from the Venetia K1 pyroclastic intrusion showing concentrations of pelletal lapilli, which are characteristically well rounded. b. SEM (backscattered-electron) image of a pelletal lapillus from (a), comprising a serpentinised olivine core and fine-grained rim comprising talc, spinel and numerous concentrically aligned micro-phenocrysts. c. Hand specimen from Letseng showing circular-elliptical pelletal lapilli and crystals. d. SEM image of an elliptical pelletal lapillus from (c); note the incorporation of smaller crystals into the



rim. e. SEM image of the matrix of (d) showing the inter-growth of voidfilling serpentine (Se) and diopside (Di), an assemblage indicative of lowtemperature hydrothermal alteration (Stripp et al., 2006). f. For comparison, a synthetic pharmaceutical granule produced by several stages of fluidized granulation; crystalline sugar core surrounded by layers of glucose, talc, polymers and cellulose.

The Letseng-la-Terae Satellite pipe contains pelletal lapilli within a steep-sided ( $\sim$ 80°), 100 m-wide circular intrusion. This cross-cuts MVK and marginal inward-dipping volcaniclastic breccias (Fig. 1b) defining a nested geometry. Pelletal lapilli are characteristically well rounded (Fig. 2c-d), ranging in size from 60 µm to 61 mm (mean = 3.5 mm;

Fig. 3a). Pelletal cores typically constitute mantle (Palmer et al., 2008) and crustal xenoliths, the most abundant being basaltic lithic clasts (85%) of presumed Drakensberg origin (Gernon et al., 2008). The rims to serpentinized olivines (Fig. 2d) typically consist of euhedral to subhedral olivine phenocrysts, very fine-grained chrome spinel, perovskite and titanite. The pore space is infilled by a secondary serpentine-diopside assemblage (Fig. 2e), which further from olivine clusters gives way to calcite (Gernon et al., 2008).



Fig. 3: Particle size and shape properties of pelletal lapilli from Letseng and Venetia, and the relationship between size and gas velocity. a. Step plot showing the frequency (%) of lapilli versus lapilli size in phi ( $\phi$ ) scale where  $\phi = -Log_2d$ , and d is the lapillus long-axis in millimetres. b. The area of the rim is plotted against the area of the core for pelletal lapilli from both intrusions. c. Histograms showing circularity for pelletal lapilli distributions from Letseng and Venetia. d. Variation in the minimum fluidisation velocity (Umf) and escape velocity (Ue) for crystals and lithic clasts, fluidised by CO2 at 1000°C (modified after Sparks et al., 2006). Parameter values are  $\rho_s = 3300 \text{ kg m}^3$ , to represent olivine crystals and dense lithic clasts; voidage,  $\varepsilon_{mf} = 0.5$  and viscosity,  $\mu$ =  $4.62 \times 10^{-6}$  Pa s. The graph shows the gas velocities required to reach U<sub>mf</sub> and U<sub>e</sub> for a range of characteristic particle sizes (shown) for Letseng and Venetia. Note that  $U_{mf}$  of the maximum lapilli size  $\approx U_e$ of the mean lapilli size.

### Results

The characteristics of observed pelletal lapilli (Figs. 2-3) are indicative of fluidized spray granulation (Christoph-Link and Schlünder, 1997; Zank et al., 2001). This process generates well-rounded composite particles (Christoph-Link and Schlünder, 1997), uniformly coated (Panda et al., 2001) with layered concentrically-aligned inclusions (Fig. 2f; Jacob et al., 2009). For both deposits, data show a moderate to strong positive correlation between the cross-sectional area of the seed particle and that of the coating (Fig. 3b), suggesting a uniform coating process and underlying scale invariance. Particle growth rate generally increases with increasing particle diameter (Zank et al., 2001), due to their greater surface area. However, in this instance larger clasts have proportionally less rim material (gradient < 1; Fig. 3b).



Larger clasts have higher inertia, requiring higher sustained velocities for fluidization, and experiencing increased abrasion at lower velocities (U <  $U_{mf}$ ). The circularelliptical geometry exhibited by pelletal lapilli (Figs. 2 & 3c) suggests their formation is governed by surface tension (Sparks et al., 2006), a major variable in fluidized spray granulation (Panda et al., 2001). The presence of multiple rims and concentrically aligned phenocrysts to some pelletal lapilli (Fig. 2b; Lloyd and Stoppa, 2003) is suggestive of a systematic multi-stage layering process (Christoph-Link and Schlünder, 1997).

Another key characteristic of spray granulation is the generation of a narrow particle size distribution (Rajniak et al., 2007), partly due to the agglomeration of fines (Zank et al., 2001; Rajniak et al., 2007). This is evidenced by the incorporation of small discrete rimmed crystals within larger pelletal rims (Fig. 2b & d). Although the Venetia and Letseng size distributions are not strictly narrow (Fig. 3a), the host and proposed source material (i.e. MVK, see Fig. 1) has a remarkably wide size distribution with observed crystal and lithic inclusions ranging from 0.015-~800 mm (6 to -9.7¢; Walters et al., 2006; Gernon et al., 2009). Venetia MVK contains a high density of small olivine crystals (mode = 0.2 mm) with proportionally fewer larger lithic clasts (mode = 23 mm) resulting in a bimodal joint size distribution (Fig. 6 in Walters et al., 2006). Lapilli sizes at Letseng and Venetia also show slight bimodality (Fig. 3a), but the size range is more restricted (0.03-32 mm; 5 to - $5\phi$ ) with a higher proportion of larger lapilli (Venetia mode = 5.7 mm) and a relative paucity of fine-grained particles (<0.5 mm; Fig. 3a).

#### Discussion

To fluidize the largest observed pelletal lapilli in the intrusions, gas velocities must have reached ~45 m/s (Fig. 3d), broadly consistent with other estimates for MVK (Sparks et al., 2006; Walters et al., 2006; Gernon et al., 2009). We emphasize, however, that the local velocity due to gas bubbles and jets is normally several times greater than the characteristic velocity of the bed (Davidson and Harrison, 1963; Roach, 1993). Additionally, the tapered geometry gives rise to a circulating fluidized system (Davidson and Harrison, 1963) enabling a wide range of pelletal lapilli sizes to coexist in equilibrium.

For Venetia,  $U_{mf}$  of the maximum-size lapilli is approximately equal to the escape velocity  $U_e$  (the velocity at which particles escape from the system) of the mediansize lapilli (Fig. 3d). This implies there must be significant local variation in gas velocity to sustain fluidization across the range of particle sizes observed whilst retaining the smaller size fraction. For Letseng, median particle size is considerably lower (~2 mm,  $U_{mf} = 3$  m/s) suggesting greater variation in gas velocity, which can be explained by the wider vent diameter and more pronounced tapering. Clasts too large to become fluidized will behave as dispersed objects (Gernon et al., 2009).

We propose that fluidized spray granulation occurs when a late-stage kimberlite dyke intrudes into unconsolidated pyroclastic deposits within the diatreme. Intensive volatile exsolution results in formation of a gas jet where velocities are sufficiently high (order of tens of metres per second; Sparks et al., 2006; Gernon et al., 2009) to fluidize the majority of particles (Fig. 3d) and inhibit formation of liquid bridges between clasts (Christoph-Link and Schlünder, 1997). Particles from MVK are entrained into the jet due to the drag force exerted by the fluidizing gas (Zank et al., 2001). Degassing is accompanied by a continuous spray of low-viscosity melt into the gas jet region (Zank et al., 2001), which uniformly coats the fluidized particles. Most of the very fine ash (<500 µm) is either agglomerated to the pelletal coatings (Christoph-Link and Schlünder, 1997; Zank et al., 2001; Rajniak et al., 2007) or elutriated by powerful gas flows (Sparks et al., 2006; Stripp et al., 2006). Due to a combination of cohesion, high gas velocities and high fluid pressures, a fracture develops and the fluidized dispersion ascends turbulently through the diatreme fill with limited attrition and breakage. Fluidization may be promoted by a sudden drop in pressure and corresponding increase in gas exsolution accompanying fracture development (Kokelaar, 1982). The lack of segregation of large lithic clasts indicates a relatively rapid termination of gas supply (Gernon et al., 2009).

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