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[1,2] and are the primary source of diamonds. Kimberlite volcanism involves the formation of diverging pipes or diatremes[3], which are the locus of high-intensity explosive eruptions[1,2]. A conspicuous and previously enigmatic feature of diatreme fills are 'pelletal lapilli'[1,3,4] — well-rounded clasts that consist of an inner 'seed' particle with a complex rim, thought to represent quenched juvenile melt[1,5,6]. Such clasts are widely documented in a range of pyroclastic successions on Earth[7,8,9]. 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[10,11,12]. 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[13] 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[11,12,14] or ejected by powerful gas flows[1,5]. This type of multi-stage intrusion will result in spatial and temporal variation in the structure and composition of pipe-fill, and consequently will influence local diamond grade and size distributions. A similar origin may apply to pelletal lapilli in other alkaline volcanic rocks including carbonatites, kamafugites and melilitites.

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Introduction

Kimberlite melts ascend from the Earth's mantle to the surface in a matter of hours to days[1,15]. Their diatreme-hosted deposits provide valuable insights into the dynamics of other volcanic conduits and represent the main source of diamonds on Earth. Kimberlite diatremes are subject to a wide range of volcanic and sedimentary processes and interactions[16,17], exceptional fossil preservation[18], and hydrothermal metamorphism[1,5]. Additionally, the xenoliths and xenocrysts they contain provide valuable information on the structure and composition of the deep subcontinental mantle[19,20].

Most volcaniclastic kimberlites contain ubiquitous yet poorly understood composite particles termed 'pelletal lapilli'[1,2,3,4,6,21]. These are defined as discrete sub-spherical clasts with a central fragment, mantled by a rim of probable juvenile origin[3]. Pelletal lapilli typically range in size from $<1-60$ mm, and occur both as accessory components of pipe-filling volcaniclastic kimberlites, and as the main pyroclast type in narrow, steep-sided 'pipes' within the diatreme. These clasts have previously been attributed to incorporation of particles into liquid spheres in the rising magma[2,8] and rapid unmixing of immiscible liquids[22]. 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[7], kamafugites[8,23], melilitites[7,22] and orangeites[9]. They have also been referred to as 'tuffisitic lapilli'[7], 'spherical lapilli'[24], 'spinning droplets'[8,25] and 'cored lapilli'[26,27]. Although pelletal lapilli are similar in appearance and structure to 'armoured' (or cored) lapilli[28], the latter are formed by accretion of moist fine-grained ash (as opposed to liquid melt) to the central fragment[29,30]. Pelletal lapilli share similar properties to particles formed during industrial fluidized granulation processes, but such processes have not previously been considered in a geological context.

Fluidized spray granulation

Fluidized spray granulation is widely used in industrial engineering to generate coated granules with specific size, density and physicochemical properties[14]. The mechanism involves continuous injection of atomizable liquids, solutions or melts into a powdery fluidized bed[11], which produces a dispersion of larger coated granules that are simultaneously dried by the hot fluidizing gas[11,12]. 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[31]. U_{mf} is defined according to the semi-empirical Ergun equation[32]:

$$\frac{-\Delta P}{h} = 150\, \frac{\mu_g . U_g}{x_p^2} \frac{(1-\epsilon)^2}{\epsilon^3} + 1.75\, \frac{\rho_g U_g^2}{x_p} \frac{(1-\epsilon)}{\epsilon^3} \qquad (1)$$

where $\Delta P/h$ is the pressure drop across a bed of height, h, ρ_g is the gas density, μ_g is the gas dynamic viscosity, ϵ is porosity and x_p is the diameter of spherical particles. Fluidized spray granulation is a characteristically steady growth process, producing uniform well-rounded particles with a concentric layered structure[12]. These physical features are diagnostic of pelletal lapilli in kimberlite deposits.

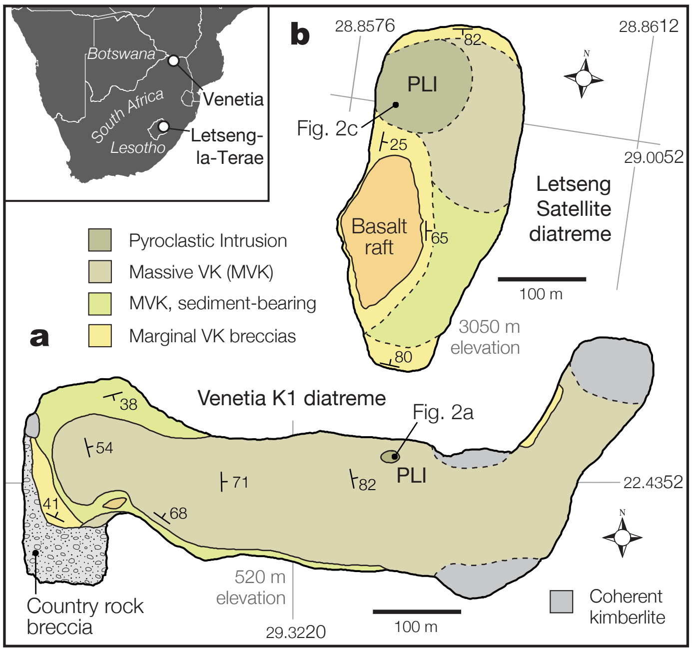

Fig. 1. **Simplified geological maps of the Venetia and Letseng kimberlite pipes. a.** Venetia K1 is dominated by massive volcaniclastic kimberlite (MVK, see text for details) with subordinate marginal breccias, sediment-bearing volcaniclastic kimberlite and coherent kimberlite lithofacies. The ∼15 m wide pelletal-lapilli intrusion (PLI) occurs near the north-central margin of the pipe, 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 ref.[33]). Inset depicts location of the deposits in southern Africa.

Field observations and results

Locality 1: Venetia K1 diatreme, South Africa

Pelletal lapilli occur prominently in two of the world's largest diamond mines, Venetia (South Africa) and Letšeng-la-Terae (Lesotho), both well-exposed, extensively surveyed[5,6,34] and economically significant localities. The Venetia K1 diatreme was emplaced during the late-Cambrian Period (c. 519 ± 6 Ma)[35] into metamorphic rocks of the neo-Archaean—Proterozoic Limpopo Mobile Belt (3.3–2.0 Ga). The diatreme (Fig. 1a) is dominated by massive volcaniclastic kimberlite, MVK (previously termed tuffisitic kimberlite breccia, TKB, sometimes abbreviated TK)[21], a characteristically well-mixed lithofacies comprising serpentinized olivine crystals and a polymict range of lithic clasts[34,36]. The formation of MVK has been attributed to fluidization[1,2,6], the scale and context of which is heavily debated[36,37]. Pelletal lapilli (Figs. 2a-b) are confined to a narrow (10–15 m diameter), discordant and lenticular body near the northern margin 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-sized, and very rare bomb-sized clasts (Fig. 2a-b), ranging in diameter from 0.2 – 100 mm (mean, $\bar{x} = 9.4$ 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.

Locality 2: Letšeng-la-Terae Satellite pipe, Lesotho

The Letšeng-la-Terae Satellite pipe erupted during the Late Cretaceous Period (c. 91 Ma[6,33]) through Lower Jurassic flood basalts of the Drakensberg Group. Pelletal lapilli occur within a steep-sided ($\sim80°$), 100 m-wide circular intrusion. This cross-cuts MVK and marginal inward-dipping volcaniclastic breccias (Fig. 1b)[33] defining a nested geometry[34]. Pelletal lapilli are characteristically well rounded (Fig. 2c-d), ranging in size from 60 μm to 61 mm ($\bar{x} = 3.5$ mm; Fig. 3a). Pelletal cores typically constitute mantle[33] and crustal xenoliths, the most abundant being basaltic lithic clasts (85%) of presumed Drakensberg origin[6]. 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[6].

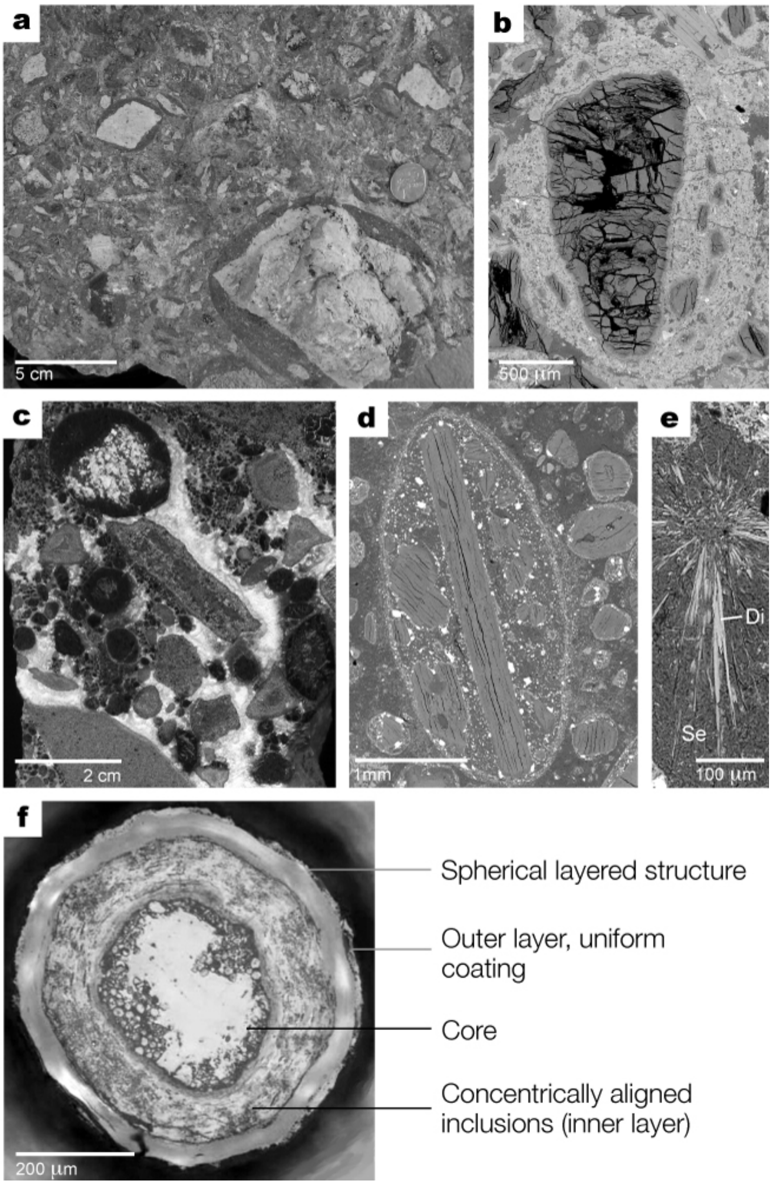

Fig. 2. **Photographs of pelletal lapilli from southern African kimberlites and a synthetic analogue. a.** Exposure from the Venetia K1 pyroclastic intrusion showing concentrations of pelletal lapilli, which are characteristically well rounded. **b.** SEM (backscattered-electron) image of a pelletal 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 void-filling serpentine (Se) and diopside (Di), an assemblage indicative of low-temperature hydrothermal alteration[5]. **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 (after ref.[38]).

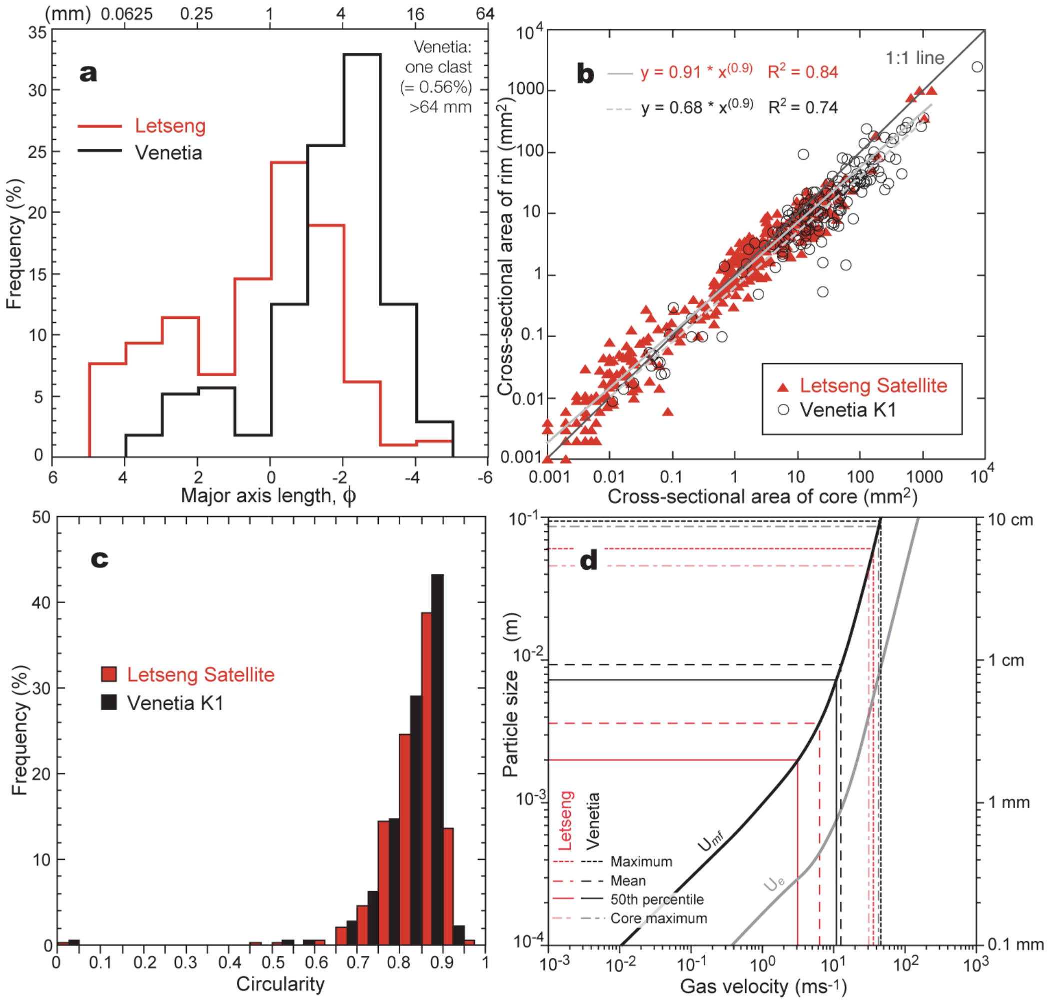

Fig. 3. **Particle size and shape properties of pelletal lapilli from Letseng and Venetia, and the relationship between size and gas velocity. a.** Step plot showing the frequency (%) of lapilli versus lapilli size in phi (ϕ) scale where ϕ = -Log$_2 d$, and d is the lapillus long-axis in millimetres. **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 (see methods). **d.** Variation in the minimum fluidization velocity (U_{mf}, equation 1) and escape velocity (U_e)[39] for crystals and lithic clasts, fluidized by CO_2 at 1000°C (modified after ref.[1]). Parameter values are ρs = 3300 kg m^{-3}, to represent olivine crystals and dense lithic clasts; voidage, ϵ_{mf} = 0.5 and viscosity, μ = 4.62×10^{-6} Pa s. The graph shows the gas velocities required to reach U_{mf} and U_e for a range of characteristic particle sizes (shown) for Letseng and Venetia. Note that U_{mf} of the maximum lapilli size $\simeq U_e$ of the mean lapilli size. The window between U_{mf} and U_e shows that a range of particle sizes can be supported (i.e. fluidized) but not ejected.

Discussion

Characteristics of pelletal lapilli

The characteristics of observed pelletal lapilli (Figs. 2-3) are indicative of fluidized spray granulation[11,12]. This process generates well-rounded composite particles[12], uniformly coated[40] with layered concentrically-aligned inclusions (Fig. 2f)[38]. 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[11], 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 circular-elliptical geometry exhibited by pelletal lapilli (Figs. 2 & 3c) suggests their formation is governed by surface tension[1], a major variable in fluidized spray granulation[40]. The presence of multiple rims and concentrically aligned phenocrysts in some pelletal lapilli (Fig. 2b)[22] is suggestive of a systematic multi-stage layering process[12].

Another key characteristic of spray granulation is the generation of a narrow particle size distribution[14], partly due to the agglomeration of fines[11,14]. 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 -9.7ϕ)[34,36]. Venetia MVK contains a high proportion of small olivine crystals (mode $\simeq$ 0.2 mm) with proportionally fewer larger lithic clasts (mode $\simeq$ 23 mm) resulting in a bimodal joint size distribution (Fig. 6 in ref.[34]). 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 -5ϕ) 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).

Constraints on gas velocity

To fluidize and coat 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[1,34,36]. 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[13,31]. Additionally, the tapered geometry gives rise to a circulating fluidized system[31] 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)[39] of the median-size 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[36].

Given the high volatile contents required to generate melts of kimberlite composition (5–10 wt.%)[41,42], we argue that gas flow-rates required to fluidize clasts are easily achievable during degassing of a kimberlite magma. Assuming a pyroclastic intrusion diameter of 10 m, and taking gas velocities of 12 ms^{-1} (for the mean of the Venetia distribution; see Fig. 3d) and 45 ms^{-1} (for the maximum size; see Fig. 3d), we would require gas flow-rates on the order of 942 m^3s^{-1} to 3.5×10^3 m^3s^{-1} respectively. Our previous calculations[36] show that degassing in kimberlite root zones could result in gas mass flow-rates as high as 3×10^6 m^3s^{-1}, so the above estimates seem conservative. Given these estimates, a hypothetical kimberlite dyke segment of conservative length, $h =$ 10 km, breadth, $b = 50$ m and width, $2w = 2$ m, containing 10% volatiles, could release sufficient gas volumes to fluidize the entire intrusion-fill for tens of seconds to several minutes. As such, a degassing dyke could sustain a gas jet for long enough to efficiently entrain a significant amount of recycled pyroclasts. These results are not surprising, as comparable basaltic systems (e.g., persistently active volcanoes) can release large volumes of gas with broadly equivalent mass fluxes over significant periods of time[43], without necessarily erupting any significant volume of degassed lava[44].

Formation of pelletal lapilli

We propose that fluidized spray granulation occurs when a new pulse of kimberlite magma intrudes into unconsolidated pyroclastic deposits within the diatreme (Fig. 4a). The magma is transported through a dyke or system of dykes in the deep feeding system, which at low to intermediate levels drive explosive volcanic flows[45] within the tapered pyroclastic intrusion. At the interface between the dyke and conduit, intensive volatile exsolution results in the formation of a gas jet[13] where velocities are sufficiently high (order of tens of metres per second)[1,36] to fluidize the majority of particles (Fig. 3d) and inhibit formation of liquid bridges between clasts[12]. Particles from MVK are entrained into the jet due to the drag force exerted by the fluidizing gas (Fig. 4a)[11]. Degassing is accompanied by a continuous spray of low-viscosity melt into the gas jet region[11]. Melt droplets are provided by fragmentation – the catastrophic bursting of bubbles to form a gassy spray[46]. The fragmentation level (Fig. 4a) will vary depending on the tensile strength of the magma[46], which will be influenced by the ambient pressure-temperature conditions, magma rheology[47] and magma water content[1,46]. As melt droplets are deposited on the hot particles, they produce a thin film governed by surface tension, which dries rapidly to form a solid uniform coating[11] (Fig. 4a-b). Most of the very fine ash (<500 μm) is either agglomerated to the pelletal coatings[11,12,14] or elutriated by powerful gas flows[1,5]. 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 (Fig. 4b). Fluidization may be promoted by a sudden drop in pressure and corresponding increase in gas exsolution accompanying fracture development[48]. The lack of segregation of large lithic clasts indicates a relatively rapid termination of gas supply[36].

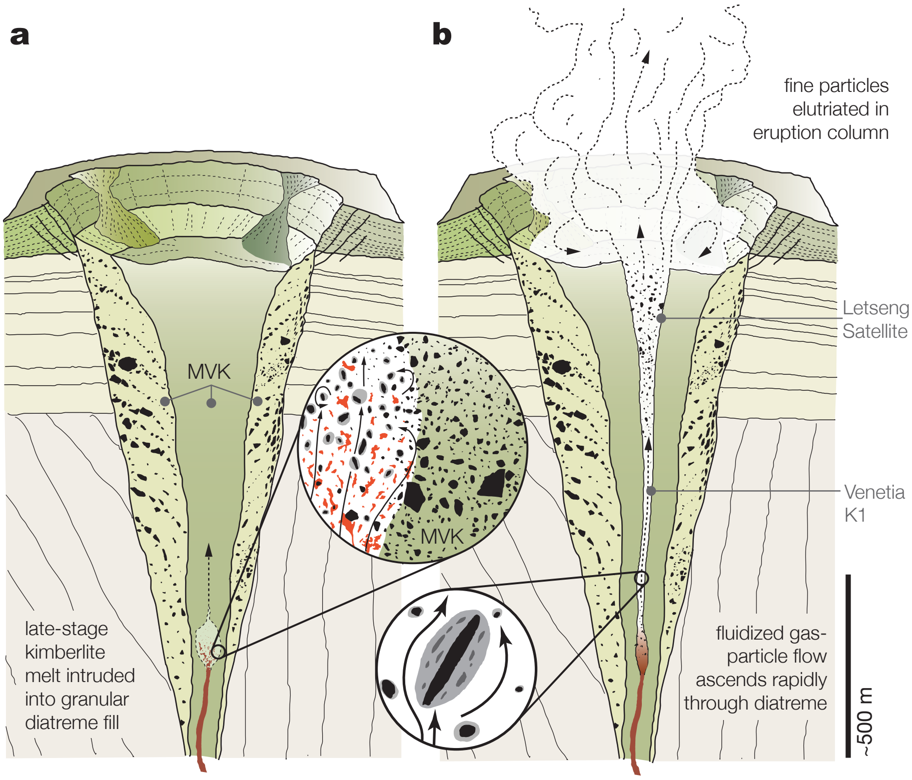

Fig. 4. **Schematic showing the formation of pelletal lapilli in kimberlite diatremes (see text for details). a.** A fluid, volatile-rich melt is intruded into loose diatreme fill; intensive volatile exsolution produces a gas-jet, opening up a fracture within MVK deposits; inset: particles from MVK are entrained and fluidized in the gas jet and uniformly coated by a spray of melt (red); fine particles are either agglomerated to pelletal coatings or elutriated by strong gas flows. **b.** Driven by gas expansion and exsolution in the jet region, gas-particle dispersion ascends rapidly (>20 m/s) and turbulently through the diatreme and the eruption is abruptly ended.

Our observations from Venetia and Letseng can be explained by dyke intrusion resulting in explosive flow processes within a narrow conduit. However, we recognise that this model will not explain all occurrences of pelletal lapilli globally, and that other important granule-forming processes may operate during eruptions. An example might include Hawaiian-style lava fountains at the surface, where it is common for melt and gas phases to coincide with crystals and entrained clasts[49]. It is not difficult to conceive situations in which such particles could be fully supported by the viscous drag of escaping volatiles, and simultaneously coated by a spray of fragmented low-viscosity melt. This process would provide an opportunity for recycling of previously generated pyroclasts. However, our model (see Fig. 4) provides a mechanism for pelletal lapilli to form at depth in pyroclastic intrusions within the vent, consistent with field relationships observed at Venetia and Letseng. In this model, pelletal lapilli are also expected to get erupted explosively and ejected during degassing (see Fig. 4b), producing deposits at the surface in which pelletal lapilli are volumetrically substantial.

Occurrence of pelletal lapilli in MVK

Several mechanisms could lead to incorporation of pelletal lapilli into more typical vent-filling MVK, as observed in other pipes such as Letseng (main pipe), Wesselton, Lemphane, Liqhobong, Kao and Premier[3,50]. For example, pelletal lapilli ejected at the surface will get deposited in marginal bedded regions, which are capable of subsiding to deep levels in the pipe during subsequent explosive bursts at depth[51,52,53,54], and gas-fluidization of the pipe-fill[36,55]. Such large-scale fluidization processes are thought to promote thorough mixing of pre-existing pyroclastic material[34,36] (including pelletal lapilli), as the vigorously fluidized dispersion effectively erodes and entrains loose material from the marginal subsided strata in the pipe[55]. It is very likely that successive eruptive phases would disrupt and disaggregate pre-existing pyroclastic intrusions, and thereby mix assemblages of pelletal lapilli together with several phases of MVK. This model is supported by the presence of steep internal contacts in kimberlite pipe-fills, separating distinct eruptive units with variable particle size distributions[34,36].

Fluidized spray granulation may also help explain the welding of pyroclasts from low-viscosity magmas[56], and complex 'transition zones' between hypabyssal and diatreme-facies kimberlites[21,57,58]. Within the Venetia K1 intrusion, occasional coalesced lapilli boundaries suggest that clasts either agglomerated during circulation or may have remained hot and partially molten during emplacement. Although sprayed kimberlite melt is likely to solidify rapidly upon contact with lithic lapilli, high magma supply rates may lead to sustained high temperatures and the system becoming dominantly viscous with particle sintering and agglutination[59]. This would explain observed gradations to non-welded deposits and overlaps in texture and composition with adjacent pyroclastic deposits[56].

Concluding remarks

The origin of pelletal lapilli is important for understanding how magmatic pyroclasts are transported to the surface during explosive eruptions. Observed differences in juvenile composition may signify a magma with a different mantle provenance, or one that had differentiated at depth prior to ascent[60]. Any resulting compositional differences may be significant in terms of diamond grade (carats per tonne), size and quality. Recognising the structurally variable nature of the pipe-fill is also important for economic forecasting. For example, the Letseng pelletal lapilli intrusion has yielded a relatively high number of large diamonds (106–215 cts), compared to the surrounding pipe-fill[33].

Spray granulation requires a strong fluidizing gas flow, so our model sheds new light on the role and magnitude of fluidization in kimberlite volcanic systems[37]. Our constraints on gas velocity provide important new inputs into thermodynamic models of kimberlite ascent and eruption, estimates of gas budget, and possibly even magma rheology. The ability to tightly constrain gas velocities is significant, as it enables estimation of the maximum diamond size transported in the flow. Gas-fluidization and magma coating processes are also likely to affect diamond surface properties.

Our observations also have important implications for understanding pyroclastic processes in conduits of active volcanoes (e.g. Ol Doinyo Lengai) where episodes of ash venting (commonly attributed to fluidization) have been related to changes in eruptive activity. In such settings, pressurised CO_2 will flow through volcaniclastic deposits in the vent and crater on its way to the surface, and is likely to fluidize some of the granular material whilst ejecting the finer particles. The gas source is different but gives rise to the same phenomena. Our results add support to the hypothesis that pelletal lapilli in other volcanic settings are formed within the diatreme as opposed to the eruption column[27].

Most diatremes worldwide contain minor hypabyssal intrusions that cross-cut pyroclastic lithofacies[1,3]. When such melts penetrate loose granular deposits in the presence of rapid gas flows, we envisage that some degree of spray granulation is inevitable. Based on the abundance of pelletal lapilli in volcanic deposits worldwide[1,3,4,7,8,22], fluidized spray granulation is likely a fundamental, but hitherto unrecognized physical process during volcanic conduit formation.

Methods

Hand specimens containing pelletal lapilli were collected from both pyroclastic intrusions (Fig. 1) and analysed petrographically using optical and scanning electron microscopy (HITACHI S-3500N). High-resolution digital photographs (scaled and oriented) were taken of polished slabs and bench exposures. Particle size distribution analysis was carried out following the technique outlined in ref.[34]. Because there is naturally a size limit to observable particles at any scale, samples were analysed at several overlapping scales[34]. Individual pelletal lapilli, lithic fragments and serpentinized olivine crystals were manually identified and digitized in the Adobe Illustrator (CS4) graphics package, and the resulting bitmap images were then processed in the image analysis software package, ImageJ (developed by the U.S. National Institute of Health; http://rsb.info.nih.gov/ij/download.html) following ref.[6]. This provided major and minor axis measurements, cross-sectional areas for cores and rims, and circularity values (defined as $4\pi \times$ area/perimeter^2; i.e. 1.0 indicates a perfect circle).

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Author contributions

T.G. directed the research; T.G., R.B. and M.T. carried out the fieldwork and sampling; T.G. and R.B. performed petrographic analysis and analyzed particle size distributions; T.G. and T.H. wrote the paper, T.G. drafted the figures and T.H. assisted with plotting the maps. All authors discussed the results and contributed to the final manuscript.

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