**Discussion Contribution: Volume fraction variations and dilation in colloids and granulars**

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**Abstract**
We discuss the importance of spatial and temporal variations in particle volume fraction in understanding the force response of concentrated colloidal suspensions and granular materials.

**Keywords**
Colloid, granular, dilation, volume fraction, glass, rheology

Two major themes of the Discussion Meeting, arising particularly from the papers presented by Pouliquen (2009), Menon (2009), and Bonn (2009), were how colloidal systems and granulars differed, especially with regard to the importance of dilation in granular response; and the role and use of system volume fraction Φ. In this contribution I wish to further consider the role of the volume fraction and how it is discussed and used, with particular relevance to the dilation response of concentrated colloidal suspensions and how this compares with dilation in dry and wet granulars.

Considering measurements/control of Φ, Poon (2009) pointed out that experimental volume fractions quoted for colloidal systems were unavoidably prone to imprecision: care must be taken to avoid ascribing particular numerical values greater importance than warranted. This is particularly important near the apparent colloidal glass transition, where rheological and dynamic properties become extremely sensitive to exact values of Φ.

The key point of this contribution is that *spatial heterogeneity* in volume fraction is an important and usually neglected aspect of concentrated particulates. Most discussion of Φ treats this parameter as a single global characteristic of a system. When discussing the regime of very high concentration, *ie* the colloidal glass transition, granular jamming, and close-packing limits, this use of Φ is too simplistic. All systems exhibit spatial and temporal variations of *local* volume fraction (Haw 2006) (a truly close-packed frozen system will still exhibit spatial but not temporal variation). Particularly in concentrated systems a single global value of volume fraction does not adequately characterise a particulate system since, due to the rheological and dynamical sensitivity to Φ already mentioned, small variations in local Φ imply potentially large variations in local response. Variations and fluctuations in local Φ cannot be ignored (Haw 2006). Of course, global variations in Φ may also be important, *eg* in shear banding phenomena (Moller *et al.* 2008).

In the Discussion Meeting Cates (2009) raised the question of whether one difference between colloidal and granular systems might be the impossibility of a fixed-volume colloidal fluid responding by dilation: given an incompressible solvating fluid, and no free surfaces *eg* fluid interfaces (Cates *et al.* 2005), total volume must be conserved. Recent experiments demonstrate that rheological jamming in cornstarch suspensions (not strictly colloidal) is directly associated with global dilation (Fall *et al.* 2008). Consideration of local variations in volume fraction illustrates that dilation *can* happen in colloidal suspensions, albeit *local* dilation rather than global, even in a confined geometry such as a channel flow (Haw 2004). Local regions of the system may dilate in order to deform in response to stress, while other regions become more concentrated (compress) to conserve total volume. There is evidence of this effect in converging channel flow experiments (Haw 2004), although to my knowledge direct microscopic measurements are lacking. These are challenging because exact resolution of (usually polydisperse) particle volumes is required to give precise values of local Φ. Such resolution at sub-micron scale in optical microscopy is difficult and in practice most confocal techniques deliver particle centre coordinates only.

One might argue that the experiments in Haw (2004) involve extensional strain and an essentially inhomogeneous stress, therefore encouraging inhomogeneous response, *ie* local dilation: the heterogeneity of the geometry and stress generate heterogeneous response. A homogeneous stress such as in simple shear might seem less likely to create local variations in response, *ie* local dilations and compressions. Recent shear-banding experiments show that even a homogeneous stress field can lead to variations in response (Moller *et al.* 2008), *ie* shear-banding phenomena are intrinsic to the fluid, not to the geometry or stress field. Furthermore, recall that variations in *local* volume fraction are present even in an unstressed, globally homogeneous system, and hence variations in response even to a homogeneous stress may certainly be expected.

Local dilation in a colloid or wet granular has a further physical consequence in contrast to a dry granular system: the necessity for interstitial fluid flow from the compressed region into the dilated region. This is effectively a porous-medium flow which introduces a new timescale into the problem, determined by the fluid viscosity and the local volume fractions/configurations of compressed and dilated regions between which the fluid flows. Additionally there is the possibility of temporary *dilation hardening*. In the geophysical literature, for example, dilation hardening has been suggested as important in earthquake response (*eg* Whitcomb *et al.* 1973, Rudnicki and Rice 1975, Rice 1975, Rudnicki and Chen 1988). Highly shear-stressed regions of porous rocks or soil must first dilate to allow shear deformation. Interstitial fluid must therefore flow from compressed to dilated zones. Rather than immediately suffer shear deformation, however, dilated zones can be temporarily 'hardened' by the temporary imbalance of pore pressure between their dilated insides and surrounding compressed regions: there is an effective compressive stress acting on the dilated region, temporarily strengthening it against shear deformation and delaying the shear strain response. (This is somewhat reminiscent of the capillary-pressure stabilised granulation mechanism proposed in Cates *et al.*, 2005.) However, once pore pressure is equilibrated by interstitial fluid transfer, the hardening stress returns to zero and the dilated region is no longer strengthened against shear deformation: this is when the actual earthquake shear strain occurs. Measurements of variation in sound speed prior to earthquake strain support this temporary dilation hardening picture for wet granulars/porous rocks. Clearly the presence of fluid, combined with local dilation, may have significant physical effects not present in dry granulars, while even in dry granulars the presence of interstitial air has important effects, though here the fluid is compressible in contrast to the usual situation in colloids and wet granulars (*eg* Burtally *et al.* 2002, Muite *et al.* 2004, LePennec *et al.* 1998).

Is this relevant to colloidal suspensions? The experiments in Cates *et al.* (2005), Fall *et al.* (2008) and Haw (2004), as well as a venerable rheological literature (*eg* Metzner and Whitlock 1958) demonstrate that concentrated suspensions are 'naturally' prone to respond by dilation. Even in a fixed-volume geometry, local dilations/ compressions are possible, and will be encouraged even in a homogeneous flow by unavoidable spatial variations in local volume fraction, to which response and dynamics will be particularly susceptible around the 'divergence' of the glass transition. It seems reasonable therefore that dilation/fluid effects important in wet granulars will also be important in concentrated colloids. Experiments are underway to test for the presence of dilation hardening in model colloidal systems (Haw, Campbell and Thomson, unpublished).

In conclusion, the high concentration response of colloidal suspensions may not be comprehensible if we insist that the volume fraction be treated as a single global parameter. Accepting that this parameter varies spatially and temporally may help us understand response and dynamics (indeed may even have relevance in explaining the link between the bulk dynamic glass transition and local crystal nucleation in hard spheres [Haw 2006, Pusey 2009]), and also indicates the importance of considering the role of the solvating fluid in colloids and wet granulars.

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**Short title**

Volume fraction variations in colloids and grains