



## Research paper

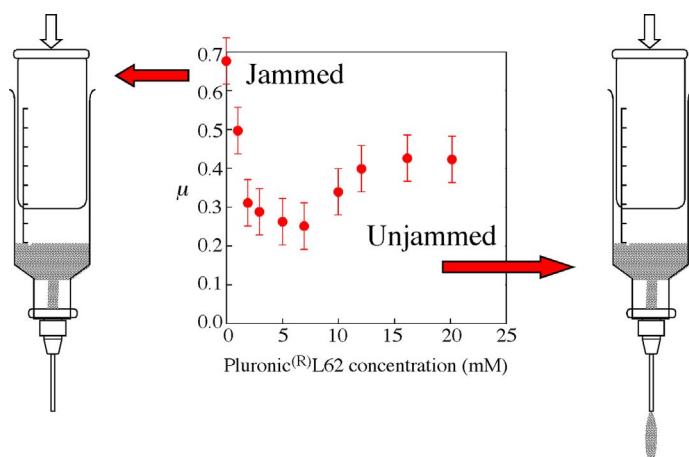
# The use of boundary lubricants for the reduction of shear thickening and jamming in abrasive particle slurries



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## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Keywords:

Abrasive slurries  
Lateral force microscopy  
Boundary lubrication  
Shear thickening  
Particle jamming

## ABSTRACT

Dense abrasive slurries are widely used in a variety of processes, but have problems in their application due to strong shear thickening and jamming. For particles in the supra-colloidal or non-Brownian, size range ( $d > 10 \mu\text{m}$ ), these are due primarily to the formation of hydroclusters at high shear rates and to direct particle-particle contacts. The latter appear to be a particular problem in the development of abrasive slurry jets (ASJ) used for abrasive water jet cutting. This study explores the use of two adsorbed surfactants and a covalently bonded silane as boundary lubricants for abrasive garnet particles of irregular shape with sharp asperities and average effective diameter  $50 \mu\text{m}$  to reduce shear thickening and jamming of their slurries. Particle-particle friction is determined using lateral force microscopy (LFM), and it is found that adsorbed layers of either a cationic surfactant, hexadecyltrimethylammonium bromide (HTAB) or a nonionic surfactant, Pluronic® L62, or of a grafted octadecyltrimethoxy silane (OTS) reduced the lateral friction by a factor of as much as two. Vane tool rheometry suggested similar reductions in shear thickening of dense garnet particle slurries, and an *ad hoc* study of jamming slurries through a flow constriction provided by a syringe yielded similar results.

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Received 24 July 2017; Received in revised form 11 September 2017; Accepted 15 September 2017

Available online 18 September 2017

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## 1. Introduction

Abrasive slurries are used in a variety of applications, including ultrasonic machining and polishing [1], chemical mechanical planarization (CMP) of microelectronic materials [2], and potentially in jet cutting [3,4]. Particle sizes vary from sub-micrometer upwards, and in most applications, the slurries are dense and handled at high shear rates. These conditions often lead to problems with shear thickening and even jamming (discontinuous shear thickening). For slurries of particles in the colloidal size range, shear thickening may result from inter-particle van der Waals forces, polymer bridging or depletion effects, etc. leading to aggregation, whereas for larger particles ( $> 10 \mu\text{m}$ ), other factors are more important [5]. These include the formation of “hydroclusters” of particles bound together by hydrodynamic lubrication forces [6] and direct particle-particle contacts [7]. The present study concerns aqueous slurries of polydisperse large-size (average linear dimension approximately  $50 \mu\text{m}$ ) angular, multifaceted garnet abrasive particles, as shown in Fig. 1, proposed for use in jet cutting applications.

Current abrasive water jets function by creating a low-velocity flow of ultra-high pressure water, which then passes through a gem orifice that accelerates the water into a mixing chamber [8]. Dry abrasive particles are introduced into the mixing chamber, entrained into the jet using the Venturi effect, and then accelerated out of the nozzle to strike the work piece. This process results in only approximately 10% momentum transfer to the particles, i.e., not all the kinetic energy of the stream is used to treat the work piece. In addition, the axial particle spacing in the jet is too large, roughly an order of magnitude larger than the particles themselves. Preliminary experiments have suggested that the inefficiencies of power transfer to abrasive particles and the sparse particle spacing within the jets might be solved by the development and implementation of dense abrasive slurry jets (ASJ) [3]. In contrast to the method of entrainment of dry abrasive particles into a high-pressure jet, as is current practice, an ASJ would employ direct pumping of a premixed abrasive suspension [3,9]. These results have shown the possibility of almost 100% efficient momentum transfer to the abrasive particles and to controllable particle spacing by using a high concentration of abrasive particles in the slurry. To this point, however, ASJ technology has not been commercialized due to issues in formulating stable slurries (with respect to sedimentation) and to problems associated with severe shear thickening. Various polymeric gellators have been found to address the problem of particle sedimentation, whereas the present work addresses the problem of

shear thickening and particle jamming during flow.

The surfaces of the proposed abrasive particles have sharp asperities, as seen in Fig. 1, so it is likely that densely-packed slurries of this type would be subject to strong shear thickening and even jamming at high shear rates as a result, at least in part, of direct point-to-surface particle contacts. Such effects might be mitigated by the use of appropriate bulk lubricants or boundary lubricants applied to the particle surfaces, reducing the coefficient of friction [3,10,11]. The present work seeks to assess the effectiveness of such surface treatments using lateral atomic force microscopy (LFM). This study complements the recent theoretical investigations of Seto et al. [12], Mari et al. [13], and Wyart and Cates [14], whose simulations show that as shear rate increases in such systems, the importance of particle-particle friction increases, leading ultimately to discontinuous shear thickening. These predictions have been corroborated by experiments of Lin et al. [15], at least for particles in the micron size range. The present work also complements that of Fernandez et al. [5], who obtained rheology results for suspensions of  $12 \mu\text{m}$  diameter ground quartz particle slurries at various volume fractions in alkali polymer solutions. In that study, adsorbed polymers were used to vary the friction coefficient, which in turn determined the critical particle volume fraction at which jamming occurred. Their slurry viscosity results were described using contact-dynamics simulations, which were also able to predict the transition from continuous to discontinuous shear thickening (jamming).

In the present study, we seek to investigate experimentally the effect of adsorbed surfactants, viz. the cationic hexadecyltrimethylammonium bromide (HTAB) and the nonionic polyethylene oxide (PEO)-polypropylene oxide (PPO) triblock copolymer Pluronic<sup>®</sup> L62, and surface functionalization with octadecyltrichlorosilane (OTS) on the lubricity of the abrasive garnet particles shown in Fig. 1 as characterized by LFM and on the rheological properties of dense slurries of these particles.

## 2. Materials and methods

### 2.1. Materials

The solid materials used in this study were garnet, an orthosilicate mineral consisting of  $[\text{SiO}_4]^{4-}$  tetrahedra connected by interstitial cations, commonly used as gemstones and abrasives. Flat-surfaced  $0.5 \text{ cm}$  square garnet pieces for LFM measurements were purchased from Allstarco.com. The garnet abrasives used, shown in Fig. 1, were 220 mesh size (average effective diameter  $50 \mu\text{m}$ ) with a specific gravity of 4.0, obtained through Alibaba.com. The surfactants used for surface treatments were hexadecyl-trimethylammonium bromide (HTAB) obtained from Sigma Aldrich (St. Louis, MO), and Pluronic<sup>®</sup> L62, a polyethylene oxide (PEO)<sub>6</sub>-polypropylene oxide (PPO)<sub>35</sub>-polyethylene oxide (PEO)<sub>6</sub> triblock oligomer obtained from BASF Corp. (Florham Park, NJ). The HTAB, a cationic surfactant, was presumed to adsorb head down to the negatively charged garnet surface, while the Pluronic<sup>®</sup> L62 was presumed to adsorb by hydrogen bonding of the PEO groups to protonated silanol groups on the garnet, as proposed by Malmsten et al. [16]. Both would produce a hydrophobic, lubricating outer surface. The organo-functional silane used was octadecyl-trichlorosilane (OTS) purchased from Gelest, Inc. (Morrisville, PA).

Both the large garnet pieces and abrasive particles were first cleaned with isopropyl alcohol followed by air-drying. The large garnet pieces used for the LFM measurements and abrasive particles used in the rheology tests were surface-treated in the same way. The garnet was placed in the surfactant solution of interest for 15 min for adsorption of surfactant to the surface. Concentrations of 0.1, 0.3, 0.6, 0.7, 0.8, 1.2, 1.5, 2, 3, 4, and 5 mM were used for the HTAB and 1, 2, 3, 5, 7, 12, 16, and 20 mM for the Pluronic<sup>®</sup> L62. Chemical modification of the garnet surface with OTS was carried out in a 95% (v/v) isopropyl alcohol in water mixture prepared in advance. Silane was added at 1% (v/v), and

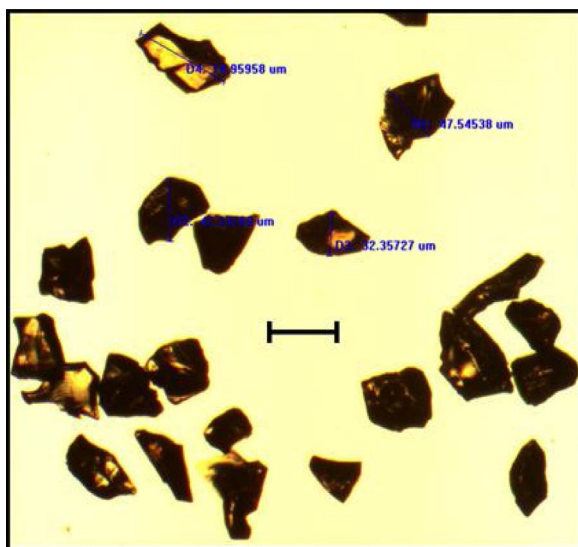


Fig. 1. Micrograph of garnet abrasive particles. Scale bar =  $50 \mu\text{m}$ .

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