Mode transition from adsorption removal to bombardment removal induced by nanoparticle–surface collisions in fluid jet polishing

Xuechu ZHAO¹, Liran MA², Xuefeng XU¹,∗
¹ School of Technology, Beijing Forestry University, Beijing 100083, China
² State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China
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Abstract: The effects of impacting particles from a jet of liquid on the removal of a surface material (on the impacted workpiece) were investigated. Experimental observations show that the cross section of the material removed changed from ‘W’-shaped to ‘U’-shaped as the size of abrasive particles was increased. Comparisons between removed material profiles and particle collision distributions indicate that the particle–surface collisions are the main reason for the material removal. The deduced number of atoms removed by a single collision implies that a transition occurs in the removal mode. For nanoscale particles, the polished surface is likely to be removed in an atom-by-atom manner, possibly due to the chemisorption of the impacting particles on the impacted surface. Contrarily, for the case of microscale particles, bulk material removal produced by particle bombardment is more likely to occur. The present mechanism of material removal for particle–surface collisions is further corroborated experimentally.

Keywords: ultra-smooth surface; fluid jet polishing; nanoparticle–surface collision; material removal

1 Introduction

Ultra-smooth surface refers to the perfect surface with atomic-level roughness and without defects [1–3]. Owing to its high reflectivity and low scattering loss, there is an increasing demand for ultra-smooth surfaces in a wide range of applications, including high-energy laser optics, extreme ultraviolet lithography, and X-ray imaging [1–5]. With the development of modern optics, information industry, and functional materials, the demand for ultra-smooth surfaces has increased. Conventional ultraprecision machining techniques, such as nano grinding, are unsuitable for such high-quality surfaces because a damaged layer may form on the machined surfaces [6–8]. Therefore, innovative processing technologies have been developed, including chemical mechanical polishing [9–11], elastic emission machining [12, 13], and fluid jet polishing (FJP) [14–16].

One of the most promising technologies for ultra-smooth surfaces is FJP, a non-contact machining method in which a mixture of water and abrasive particles is sprayed onto the polished surface through a nozzle (see Fig. 1(a)) [14]. FJP can produce surfaces with atomic smoothness and has received a lot of research attention in the past decades [17–20]. To improve the quality of the polished surfaces in FJP, understanding the underlying material removal mechanism is essential. Experiments showed that the amount of material removal has a linear relationship with the abrasive concentration, and that there is no material removal (even under pressure as high as 120 MPa) by using pure water without abrasive particles [16, 21, 22]. This means that the collision
of the abrasive particles on the workpiece surface must have a decisive effect on the material removal by FJP.

The mechanism by which the particle–surface collisions in a liquid jet remove materials from a workpiece surface remains elusive. Two basic mechanisms of material removal have been proposed: (1) bombardment removal; (2) adsorption removal. In the bombardment model, material removal is associated with scratches, cracks, and craters on the sample surface generated by rubbing, plowing, and cutting, which are in turn due to the continuous bombardments of the high-velocity particles [21–25]. Contrarily, the adsorption model believes that the ultra-smooth surface is produced probably not by the erosion of the particles, but by the chemical reactions between the colliding surfaces. When the impacting particles come into contact with the workpiece surface, there is some probability of chemical reactions and bonds forming between the contacting surfaces. The bonding atoms of the workpiece surface may be removed from the surface when the particles move away under the hydrodynamic effect of the surrounding fluid [3, 4, 15, 26–33].

The speculation that material removal is caused by chemical adsorption is based on several reasons in various studies, i.e., the ultra-smooth surface is not likely generated by mechanical interaction [3], the results from molecular dynamic (MD) simulations [4, 15, 29], the impact force acting on the surface by the particles [26], the sharp decrease in material removal rate with increasing concentration of H$_2$O$_2$ in the slurry [27], the change in the Fourier transform infrared attenuated total reflection (FT-IR-ATR) spectrum of the surface before and after polishing [28], and no material removal when using chemically inactive nanoparticles [33]. Besides these, knowing the events that take place when a single particle collides with a solid surface should be helpful to clarify the mechanisms of material removal induced by particle–surface collisions. To this end, the impact of a cylindrical liquid jet containing deionized water and SiO$_2$ nanoparticles on single crystal silicon surfaces has been experimentally investigated, and an atomic scale deformation, which is to some extent in favor of the bombardment model of the material removal, was found on the impacted surfaces [25]. Using fluorescent nanoparticles as tracing particles, collisions between the impacting nanoparticles and the impacted surface in a liquid jet were directly observed [34, 35]. Adsorption of the impacting particles on the solid surface and the consequent desorption of the particles were also observed, implying the possibility of the adsorption model of material removal [34]. Because nanoparticle–surface collision and collision-induced material removal are difficult to be directly observed, molecular dynamic simulations are often utilized. But most of the simulations have been performed on the nanoparticle–surface collisions in vacuum or air, not in liquids [36–40].

This study aims to clarify in which manner, bombardment or adsorption, a workpiece surface material is removed by the collision with a single particle from a liquid jet. We focus on the effects of the particle size on material removal. To this end, polishing experiments on silicon surfaces using slurry jets containing abrasive particles of different sizes were performed. The profiles of the material (to be removed) on the silicon surfaces were measured and then compared with the distributions of the particle–surface collisions along the impacted surfaces. The consistency between removed material profiles and particle collision distributions confirms that the particle–surface collision is likely the main cause of material removal during polishing. The number of the workpiece atoms removed by a single particle–surface collision were analyzed. The results indicate that a transition occurs in the material removal mode of particle–surface collision when the size of the abrasive particles increases.

2 Material and methods

To explore the role of particle–surface collision on material removal, polishing experiments on silicon surfaces using abrasive fluid jet were performed. In the experiments, a colloidal slurry was delivered by a pump to a converging nozzle with a diameter of about 1 mm and then impacted vertically on the silicon surfaces. The standoff distance, i.e., the distance between the nozzle exit and the workpiece surface, is 10 mm (see Fig. 1(a)). The impacting speed of the jets used here ranges from about 41 to
about 63 m·s⁻¹, which are the typical jet speeds used in FJP [15, 26]. The abrasive particles used here are SiO₂ particles with diameters of about 100 nm, 500 nm, 2 µm, and 5 µm (see Fig. 2). The abrasive concentration of the liquid jet in all the experiments is about 5 wt%. After the impact, the morphology of the polished surfaces was measured by white light interference.

Knowledge of the particle–surface collision is also needed for revealing the relationship between the collision and material removal. Because direct observation and measurement of the fluid field and the nanoparticle–surface collision are of great difficulty, the computational fluid dynamics (CFD) method has been used here. The Reynolds number of the jet flow is defined as \( Re = \frac{\rho U d}{\mu} \), where \( \rho \) is the fluid density, \( U \) is the mean velocity of the fluid at the nozzle exit, \( d \) is the diameter of the nozzle, and \( \mu \) is the fluid dynamic viscosity [41, 42]. Here, \( Re \approx 50,000 \), which is much higher than the criterion for the inception of turbulence. In this study, the turbulent jet flow is simulated by solving the Reynolds-averaged Navier–Stokes (RANS) equations with ANSYS Fluent, and renormalization group (RNG) \( k-\varepsilon \) model has been chosen as a closure for RANS equations owing to its successful applications in the simulation of impinging jet flow [17, 41–45]. The workpiece surface is treated as a no-slip wall and the standard wall function is used with appropriate treatment of the near-wall region in terms of mesh refinement.

The geometry of the computational domain including the nozzle structure and the boundary conditions used here is shown in Fig. 1(b). In our CFD model, the diameter of the nozzle entrance \( D \) is 6 mm, the nozzle diameter \( d \) is 1 mm, the length to diameter ratio of the nozzle \( L_1/d \) is 4, the contraction angle of the nozzle \( \alpha \) is 30°, and the distance between the nozzle and the workpiece surface \( L_2 \) is 10 mm. Owing to the axisymmetric configuration of the system, the cylindrical coordinates \((r, z)\) are chosen. The entire computational domain begins with the entrance of the nozzle which is set as the pressure-inlet and ends up with the pressure-outlet whose pressure is set as 0.1 MPa which is equal to the atmospheric pressure. The simulation in this paper aims to investigate the particle–surface collision in the fluid jet, not the material removal on the polished surfaces. The distribution of material removal on the polished surfaces will be measured by experiments. Because the dimensions of material removal on the surface during the polishing are very tiny compared to the dimensions of the computational domain, the influences of the material removal on the flow field and the particle motion can be neglected. Thus, the geometry of the simulation does not change in the simulation.

In the simulations, the flow field of a pure liquid jet was first calculated by the volume of fluid (VOF) method in which air is the primary phase and water is the second phase. After a converged solution for the liquid jet was obtained, the particles were
added into the fluid across the nozzle entrance. Initially, the particles are distributed uniformly along the entrance and have the same velocity as the surrounding fluid. Then, the motion of the particles in the fluid was computed by the discrete phase model (DPM). Owing to the low particle concentration used here, both the particle-particle interactions and the influences of the particles on the flow field were neglected.

3 Results and discussions

3.1 Relationship between material removal and particle–surface collision

The profiles of the material removal on the polished surface measured by the white light interference are shown in Fig. 3. It can be seen that owing to the axisymmetric experimental setup, the material removal on the polished surface is also axially symmetric to the jet axis. In the experiments, two different types of cross-sections of the material removed were observed, i.e., 'W'-shaped and 'U'-shaped (see Fig. 3). The experiment results show that 'W'-shaped material was removed when the abrasive particle is small, i.e., when the diameter of the particles is 100 nm or 500 nm. Whereas, for the case of larger abrasive particles with diameters of about 2 µm and 5 µm, 'U'-shaped material is more likely to be removed.

The relationship between the profiles of material removed and the size of abrasive particles implies that the particles play an important role in polishing. Investigations into the relationship between the distribution of collision density and the profiles of materials removed would be helpful in the understanding of the material removal mechanism. Here, the collision density is defined as the number of particle–surface collisions per unit time per unit area of the impacted surface. From the radial position \( r \) of each particle–surface collision obtained by numerical simulations, a function \( N(r) \), which represents the total number of particle–surface collisions in the region \((-r, r)\) of the impacted surface (see Fig.

![Fig. 3](https://mc03.manuscriptcentral.com/friction)
1(b)), can be deduced. Considering that, initially, the particles are distributed uniformly at the entrance of the nozzle, the collision frequency, which is defined as the total collision number per unit time on a circular area of the polished surface with the center at the origin and with a diameter of $r$, can be calculated as $F_c(r) = \frac{\pi \rho_s U d^2}{4N_0^2} N(r)^2$, where $\rho_s$ is the number density of the particles in the liquid, $U$ is the mean velocity of the fluid at the nozzle exit, $d$ is the diameter of the nozzle, and $N_0$ is the total number of particles added into the fluid across the nozzle entrance in the simulations. Further, the collision density at the radial position $r$ can be expressed as

$$\rho_c(r) = \frac{dF_c(r)}{2\pi r dr} = \frac{\rho_s U N(r)d^2}{4N_0^2} \frac{dN(r)}{dr}$$

The calculated distributions of the collision density on the polished surface were compared with the profiles of the removed material (Fig. 4). It can be seen from the figure that the variation of the collision density shows the same trend as that of the removed material, i.e., when the size of the abrasive particles is small, both of them increase first and then decrease with the increase in radial distance $r$. They both have a minimum at the position $r = 0$ and show a peak at almost the same radial position. The numerical simulations indicate that as the diameter of the impacting particle increases, the collision density at $r = 0$ also increases. For the case of particles with diameters of about 5 $\mu$m, the collision density can even reach its maximum value at $r = 0$. Accordingly, the material removal at $r = 0$ increases, and as a result, the shape of removed material changes from 'W'-shaped to 'U'-shaped.

The variation of the collision density with the particle size can be attributed to the synergy between the hydrodynamic effect and the centrifugal effect.

**Fig. 4** Distributions of the observed material removal (solid lines in the pictures) and the calculated collision density (dashed lines in the pictures) on the impacted surface as a function of the radial position $r$. The abrasive particles are SiO$_2$ particles with a diameter of (a) 100 nm, (b) 500 nm, (c) 2 $\mu$m, and (d) 5 $\mu$m.
on the particles. Driven by the hydrodynamic force which is proportional to the particle diameter \( d_P \), the particle will move with the streamline of the fluid. Meanwhile, owing to the inertia force which is proportional to the third power of \( d_P \), the particle will deviate from the fluid streamline and may collide with the impacted surface. For the small particles, the hydrodynamic force will dominate and almost no collision occurs in the region near the origin (i.e., \( r=0 \)), thus forming an inverse parabolic collision density curve. Contrarily, if the particle is large enough, the inertia force may dominate and the collision number can reach its maximum in the region near the origin. In Fig. 4(c), the collision density near the origin of the abscissa is low but the depth of the dimple reaches its maximum. This contradiction can be explained by the agglomeration of the abrasive particles in the liquid causing more collisions in the region near the origin than in the calculations.

3.2 Material removal by a single particle–surface collision

The consistency between the distribution of collision density and that of the removed material indicates that the particle–surface collision is possibly the main reason for the material removal in the polishing. Exploring in which way the material is removed by a single particle–surface collision will be helpful to understand the material removal mechanism. From the measured volumes of the removed material \( V \) on the polished surfaces in the polishing experiments, the number of atoms removed from the workpiece surfaces per unit time can be obtained as

\[
N_{\text{atom}} = \frac{\rho_w V}{M_{\text{atom}} T},
\]

where \( \rho_w \) is the density of the workpiece, \( M_{\text{atom}} \) is the mass of an atom of the workpiece, and \( T \) is the operation time. Then, the number of atoms removed by a single particle–surface collision can be expressed as

\[
\text{NPC} = \frac{N_{\text{atom}}}{F_{1}(\infty)},
\]

where \( F_{1}(\infty) \) is the total number of particle–surface collisions per unit time on the whole polished surface.

The material removal rate MRR, the total number of particle–surface collisions per unit time CPS, and the number of atoms removed per single collision NPC are shown in Fig. 5 as functions of the particle diameter on a double logarithmic plot. The figure shows that the material removal rate increases with both the particle diameter and the jet speed. From the figure, it can be seen that the variation of the atom number removed by a single collision with the jet speed is not monotonous. This may be attributed to the instabilities in the experimental conditions and the inaccuracies in the measurements. However, the dependence of NPC on the particle
size is very obvious. The figure clearly shows that NPC grows almost as a power law with increasing particle diameter at all the jet speeds. For the abrasive particles with a diameter of about 100 nm, the number of removed atoms by one single collision is much lower than 1, with an average of about 0.1. This means that when small nanoparticles are used as abrasive particles, approximately 10 collisions are needed to remove just one atom from the workpiece surface. Contrarily, one single collision by larger particles with a diameter of about 5 µm can remove up to millions of atoms from the impacted surfaces.

3.3 Mode transition of material removal generated by the particle–surface collision

The huge difference in the number of removed atoms by a single collision implies that the mechanisms of material removal may be different for different size particles. The results indicate that a transition occurs in the mode of material removal induced by particle–surface collisions. Based on the values of NPC of impacting particles with different sizes, the mechanism by which particles remove the surface material can be speculated.

It can be reasonably conjectured that, when the diameter of the abrasive particle is about 100 nm, the surface material is likely to be removed in an atom-by-atom manner because the average number of removed atoms by the collisions is much lower than one. In the aqueous solution, surfaces of both particles and workpieces are easily hydroxylated and then terminated by hydroxyl groups. In the present case of SiO₂ particles and Si surfaces, hydroxide species (i.e., Si–OH) should be formed on both surfaces. When the particles hit the workpiece surface, dehydration reactions (i.e., Si–OH + Si–OH → Si–O–Si + H₂O) may be activated in which siloxane bonds Si–O–Si are formed between the particle and the workpiece accompanied by the release of water molecules, where P represents the atom of the particle, O the oxygen atom, and M the atom of the workpiece. Owing to this selective removal in the polishing, the workpiece surface is expected to be flattened on an atomic scale.

As for larger particles with microscale diameters, the average number of removed atoms is much larger than one, and therefore we can reasonably speculate that bulk removal of material due to the bombardment of particles is more likely to occur (see Fig. 6(b)). Assuming that the shape of the crater generated on the impacted surface by a single collision of a spherical particle is a spherical cap whose diameter is equal to the diameter of the impacting particle, the height $h$ and the base radius $R$ of the crater can be easily calculated and are 0.658 nm and 51.284 nm for 2 µm particle, and 2.3 nm and 151.69 nm for 5 µm particle.

3.4 Experimental corroborations

Different material removal modes will result not only in different material removal rates but also in different surface qualities. If the material is removed
atom-by-atom, an ultra-smooth surface with atomic-level roughness without defects can be achieved [4, 15, 26, 46, 47]. Contrarily, in the bombardment removal regime, the material removal rate can be higher, but the surface quality could be deficient.

Assuming that the spherical cap-shaped craters are located uniformly and closely on the impacted surfaces, the surface roughness of the finished surfaces can be estimated from the calculated dimensions of the craters and are about 0.223 nm (root-mean-square, rms), 0.208 nm (roughness average, Ra), and 0.658 nm (peak-to-valley height, PV) for 2-µm abrasive particles, and 0.791 nm (rms), 0.745 nm (Ra), and 2.3 nm (PV) for 5-µm abrasive particles. The results are reasonably consistent with the previous experimental results (rms = 0.518 nm for 2-µm SiO₂ particle [43], PV = 1.25 nm for 2-µm SiO₂ particle [3], Ra = 0.987 nm for 3-µm CeO₂ particle [26], rms = 0.64 nm and PV 2.97 nm for 1–10-µm SiO₂ particle [48]).

To confirm the proposed mechanism for the material removal, polishing experiments on silicon surfaces using different size abrasive particles were performed, and the morphology of the polished surfaces was measured by using an atomic force microscope (AFM) (see Fig. 7). The measured surface roughness of both the original surface and the polished surfaces are illustrated in Fig. 8. The results show that the surface roughnesses of silicon surfaces polished by nanoparticles (i.e., particles with a diameter of 100 nm or 500 nm) is about 0.33 nm (Ra), which is even lower than that of the original silicon surface (Ra ≈ 0.38 nm). This means that the surface quality is improved after polishing and may imply that the nanoparticles remove the material in an atom-by-atom manner.

The results also indicate that the quality of the surfaces polished by microscale particles is much lower than that by nanoparticles. This implies that, for the case of microscale particles, the workpiece surface material was more likely removed by bombardment. The measured surface roughness generated by the microscale particles was compared with the predictions calculated by the bombardment removal model. The results show that the measurements reveal the essence of the predictions: the surface roughness increases dramatically when the diameter of the abrasive particles increases from 2 to 5 µm.

Fig. 7 AFM images of (a) original silicon surface and that of the surfaces polished by SiO₂ particles with a diameter of (b) 100 nm, (c) 500 nm, (d) 2 µm, and (e) 5 µm.

From the figure, it can be seen that there are some deviations between the experimental measurements and theoretical predictions. The surface roughness obtained by nanoparticles is higher than the predicted value in the bombardment case with 2 µm particles. One possible explanation for the deviations is the idealized distribution of the craters on the polished surfaces assumed in the calculations of the surface roughness. Another explanation for the deviations between the experimental measurements and theoretical predictions is the possible
Fig. 8 Surface roughness of original silicon surface and silicon surfaces polished by SiO₂ particles with a diameter of 100 nm, 500 nm, 2 µm, and 5 µm.

agglomeration of the abrasive particles in the experiments.

4 Conclusions

In summary, polishing experiments and numerical simulations on particle–surface collisions were performed, and the effects of the particle size on material removal were analyzed. The consistency between the measured profiles of the removed material and the computed distributions of the collisions confirms that the particle–surface collision is the main means by which materials are removed in FJP. The number of removed atoms by a single collision was deduced as a function of particle size and the results imply that the mode of the collision-induced material removal changes as the size of the abrasive particles increases. For smaller nanoparticles, the workpiece surface is likely to be removed in an atom-by-atom manner through the chemical adsorption of the impacting particles on the impacted surface, resulting in a lower material removal rate and a higher surface quality. For the microscale particles, bulk removal induced by the bombardment of particles may occur, accompanied by higher material removal rate and deficient surface quality.

Our proposed material removal mechanisms by the particle–surface collisions are corroborated by the measurements on the roughness of the polished surfaces. However, more direct evidence such as the remains of reaction products on the polished surfaces would be helpful to validate the chemical adsorption model of material removal in FJP. There is an energy barrier that needs to be overcome for the dehydration reactions that take place the contacting surfaces [4, 15, 33]. Material removal by chemical reaction should be closely related to; not only the number of collisions but also the density of the hydroxyl groups on the surfaces, the size of the contact area, the impacting velocity of the particles, the rise of particles, etc. For simplicity, only the collision number is considered in this study. However, despite these limitations, we give a rough estimation of which manner material is removed by particle–surface collision. Our study can be used as the basis for devising more complicated models, for example, models considering the quantitative connection between the removal mode and the particle size.

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**Xuechu ZHAO.** She is a Ph.D. student of mechanical engineering at Beijing Forestry University, China. She received her B.S. degree in mechanical engineering from Beijing Forestry University in 2015. Her current research interests are the fabrication of ultra-smooth surfaces and the material removal mechanism of fluid jet polishing.

**Xuefeng XU.** He is a professor of mechanical engineering at Beijing Forestry University, China. He received his B.S. and M.S. degrees in mechanical engineering in 1994 and 1997, respectively, all from Tianjin University, China, and received his Ph.D. degree in mechanical engineering in 2008 from Tsinghua University, China. His current research interests include the material removal mechanism of fluid jet polishing, contact electrification, triboluminescence, and droplet evaporation.