Glass optics with ultra-low roughness surfaces (<2 Å rms) are strongly desired for high-end optical applications (e.g., lasers, spectroscopy, etc.). The complex microscopic interactions that occur between slurry particles and the glass workpiece during optical polishing ultimately determine the removal rate and resulting surface roughness of the workpiece. In this study, a comprehensive set of 100 mm diameter glass samples (fused silica, phosphate, and borosilicate) were polished using various slurry particle size distributions (PSD), slurry concentrations, and pad treatments. The removal rate and surface roughness of the glasses were characterized using white light interferometry and atomic force microscopy. The material removal mechanism for a given slurry particle is proposed to occur via nano-plastic deformation (plastic removal) or via chemical reaction (molecular removal) depending on the slurry particle load on the glass surface. Using an expanded Hertzian contact model, called the Ensemble Hertzian Multi-gap (EHMG) model, a platform has been developed to understand the microscopic interface interactions and to predict trends of the removal rate and surface roughness for a variety of polishing parameters. The EHMG model is based on multiple Hertzian contacts of slurry particles at the workpiece-pad interface in which the pad deflection and the effective interface gap at each pad asperity height are determined. Using this, the interface contact area and each particle’s penetration, load, and contact zone are determined which are used to calculate the material removal rate and simulate the surface roughness. Each of the key polishing variables investigated is shown to affect the material removal rate, whose changes are dominated by very different microscopic interactions. Slurry PSD impacts the load per particle distribution and the fraction of particles removing material by plastic removal. The slurry concentration impacts the areal number density of particles and fraction of load on particles versus pad. The pad topography impacts the fraction of pad area making contact with the workpiece. The glass composition predominantly impacts the depth of plastic removal. Also, the results show that the dominant factor controlling surface roughness is the slurry PSD followed by the glass material’s removal function and the pad topography. The model compares well with the experimental data over a variety of polishing conditions for both removal rate and roughness and can be extended to provide insights and strategies to develop practical, economic processes for obtaining ultra-low roughness surfaces while simultaneously maintaining high material removal rates.

**Keywords:** mechanochemical; polishing; borosilicate glass; fused silica; roughness
strongly influence the roughness during polishing of fused silica glass. Using a model, called the Ensemble Hertzian Gap (EHG) model, which accounts for multiple Hertzian contacts of slurry particles on the workpiece, the load per particle was determined. Combining with a bimodal removal function to account for both chemical and plastic removal, the roughness of the polished surfaces was simulated. In the following study, the EHG polishing model is expanded (now called the Ensemble Hertzian Multi-gap (EHMG) model) to account for a more comprehensive set of polishing parameters including glass type, slurry PSD, slurry concentration, and pad topography. A representative set of experimentally polished surfaces was measured for material removal rate and surface roughness to demonstrate the validity of the model and to illustrate the nature of the microscopic interactions controlling both removal rate and roughness.

II. Experimental Procedure

Various flat (100 mm diameter × 10 mm thick) glass workpieces [fused silica glass (Corning 7980; Corning Incorporated, Corning, NY); borosilicate glass (BK; Schott Glass Technologies, Duryea, PA); and phosphate glass (LHG-8 laser glass; Hoya Corporation, Fremont, CA)] were polished using the convergent polishing method. The baseline polishing conditions were the same for all samples (MHN polyurethane polishing pad (MHB15A 0.50 HII; Eminess Technologies, Scottsdale, AZ), 20 rpm lap rotation, 20 rpm optic rotation, 100 mm lap center—optic center separation, 0.6 psi applied pressure, 0.6 mL/min slurry feed rate, 1 cm stroke, and 4 h polishing run). The key polishing variables investigated were slurry PSD and composition, slurry concentration, pad treatment (or topography), and the glass type. Table I summarizes the polishing conditions for each of the samples. Each polishing condition was performed on a minimum of three samples to check for repeatability.

For the pad treatment, diamond conditioning (DC) was performed using a diamond conditioner (Diamonex 250355FT; Morgan Technical Ceramics, Allentown, PA) in the presence of water. The various slurries used in this study include NanoArc® 6752 cerium oxide (Nanophase Nanoengineered Products, Romeoville, IL), NanoArc® 6750 cerium oxide (Nanophase Nanoengineered Products), Unstabilized Hastilite PO cerium oxide (Universal Photonics, Hicksville, NY), Stabilized Hastilite PO cerium oxide, NanoArc® 6450 cerium oxide (Nanophase Nanoengineered Products), UltraSol™ 2EX colloidal silica (Eminess Technologies), and UltraSol™ S27 colloidal silica (Eminess Technologies). Some key characteristics of the PSD are listed in Table I; PSD measurement method is described elsewhere. Note that NanoArc® 6752 and 6750 have the same PSD, but 6750 has a higher viscosity. Also, Stabilized Hastilite PO is the same as Unstabilized Hastilite PO except that it has been chemically modified to improve the tail end of the PSD (details described elsewhere).

The material removal rates of the samples were measured gravimetrically. The surface roughness was measured at two spatial scale lengths: (1) \( \mu \)-roughness (800 \( \mu \)m × 1600 \( \mu \)m with of pixel size 1.5 \( \mu \)m) using a white light interferometer (WYKO NT3300, Bruker Corporation, CA); and (2) AFM roughness (5 \( \mu \)m × 5 \( \mu \)m with pixel size of 9.8 nm) using an atomic force microscope (Digital Instruments Dimension 3100, Bruker Corporation, CA). The topography of the pad surface was measured using a 3D laser scanning confocal microscope (Keyence VK-X110, Itasca, IL).

III. Results

The average measured removal rates and rms \( \mu \)-roughness over the two spatial bands (800 \( \mu \)m × 1600 \( \mu \)m and 5 \( \mu \)m × 5 \( \mu \)m) for each of the polishing experiments are summarized in Table I. The polishing experiments are grouped into four major series: slurry PSD (S), slurry concentration (N), pad topography (T), and a mixed glass type-slurry PSD (F,B, or P). The baseline polishing experiment (Sample C) on fused silica glass (using Stabilized Hastilite ceria slurry, MHN pad treated with 5 min diamond conditioning) intersects many of the polishing series and is listed multiple times in the Table to aid in interpretation of each of

![Fig. 1. Schematic illustrating proposed parameters affecting material removal and resulting surface roughness during polishing.](image-url)
<table>
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<th>Series</th>
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<th>Slurry type</th>
<th>Slurry type</th>
<th>Slurry PS (nm)</th>
<th>Exp. slope of PSD (μm)</th>
<th>Slurry conc (Baume)</th>
<th>Pad treatment</th>
<th>Removal rate (μm/hr)</th>
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the series. Also, listed in the Table are the major characteristics of the polishing slurry including the average particle size and exponential slope ($d_{PSD}$) in the PSD of the largest particles at the tail end of the distribution. The measurements of PSD, these parameters, and their correlation with surface roughness are described in our previous study.$^9$ $d_{PSD}$ is determined from a single exponential fit to the tail end of the measured PSD of the slurry $[f(r)]$ in the form:

$$f(r) = Ae^{-\frac{r}{d_{PSD}}}$$

where $r$ is the particle radius and $A$ is a pre-exponential constant. Note the slurry PSD series samples are generally listed in order of decreasing $d_{PSD}$ (i.e., narrowing PSD). For the other series, each polishing experiment is listed in order of increasing slurry concentration (i.e., Baume) for the Slurry Concentration series, increasing treatment or use of the pad for the pad topography series, and increasing removal function of the glass or increasing $d_{PSD}$ of the slurries in the glass type–slurry series.

Figure 2 shows a cross plot of the removal rate and AFM roughness for each of the polished samples in the first three series. The baseline polishing experiment (Sample C) has been circled to illustrate the crosscut between many of the series. For the Slurry PSD (S) series, the general trend is that higher removal rates result in higher roughness. These results are consistent with the general observation that long polishing times at low removal rates are required to achieve low roughness surfaces. In contrast, for the Slurry Concentration (N) and Pad Topography (T) series, the opposite effect was observed where higher removal rates corresponded to lower rms AFM roughness. This trend was most dramatic with the Pad Topography series by comparing Samples T1 and T3; the removal rate increased from 0.08 to 2.10 µm/h, while the corresponding AFM roughness decreased from 0.67 to 0.26 nm. Note, the µ-roughness (800 µm × 1600 µm) showed similar trends as the AFM roughness (5 µm × 5 µm) for most of the series. The exception was the Slurry Concentration series which showed a significant increase in µ-roughness with increase in slurry concentration and removal rate (see Table I).

The results for the glass type–slurry PSD series are shown in Fig. 3. Regardless of the slurry used, the removal rate had the following general trend: phosphate glass > borosilicate glass > fused silica. This is qualitatively consistent with the removal function trend measured for single particles in our previous study of nanoscraping on these glasses (see Fig. 4).$^{19}$ In addition, the magnitude of the difference in removal rates between the three glasses increased with increase in the width of the PSD (i.e., increase in $d_{PSD}$). Also, in general, the rms µ-roughness followed a similar trend. In contrast, the AFM roughness was largely unchanged for the different glasses, with the exception that Unstabilized Hastilite slurry followed the same glass trend described above.

![Fig. 2. Cross plot of measured removal rate and AFM Roughness of fused silica glass samples polished under different conditions described in Table I.](image1)

![Fig. 3. Effect of Slurry PSD and glass type on removal rate, µ-roughness and AFM roughness.](image2)

![Fig. 4. Measured removal function (shaded regions) as a function of load for individual slurry particles (after $^{19}$); and calculated load per particle distribution (lines) for three selected slurries using the EHMG model (see Appendix).](image3)
Figure 5 compares the pad topographies of the various pad treatments (T) as measured by laser confocal microscopy over a maximum spatial scale length of 1.4 mm. The as-fabricated pores in the pad are easily observable. The new MHN pad has a significant number of tall (100’s of μm) asperities which are removed with polishing time and diamond conditioning treatment. The results in Fig. 5 are summarized in a more convenient form as cumulative height histograms in Fig. 6, which are used directly as inputs to the EHMG model discussed below. A more uniform pad topography correlated with an increase in the observed removal rate on the polished workpiece. The pad topography over a larger (20 mm × 20 mm) region is shown in Fig. 7(a) for Sample T3 illustrating the overall longer range flatness of the pad. As shown in Fig. 7(b), the longer range flatness is also increased with diamond conditioning treatment as illustrated by several lineouts from Sample T1 (untreated, new MHN pad) and T3 (45 min DC MHN pad). Finally, some secondary, but important effects on removal rate and roughness are also evident from the data summarized in Table I. First, S1 and S2 were polished using the same slurry material and PSD, but with very different slurry viscosities. The increase in slurry viscosity resulted in significantly lower removal rate and lower roughness which is likely caused by increased hydrodynamic forces reducing the overall load per particle distribution. This is qualitatively consistent with the trend expected with the EHMG model (see Discussion Section). Second, even though S4–S6 were polished using ceria and silica slurries with similar PSD, they still had noticeable differences in removal rate and roughness. Possible causes for this include interface chemistries, surface charge effects, and inherent reactivity differences between silica and ceria. Such effects to be thoroughly evaluated require additional experimentation and characterization and are left for future study.

IV. Discussion

(1) Single Particle Removal Function and the Load per Particle Distribution

The amount and shape of removal by each individual particle, combined as an ensemble of multiple Hertzian contacts of slurry particles, forms the basis to model the overall material removal rate and roughness. In our previous study, we proposed that removal can occur both by molecular level chemical reaction at the Angstrom level (via condensation–hydrolysis reactions) and by nano-plastic deformation at the nanometer level. Note the former is governed more by the chemistry of the slurry (composition of the slurry particle, pH, viscosity, colloidal charge, etc.) and glass type, while the latter is dominated more by mechanics. As described in the Introduction, the quantitative removal function was experi-
mentally determined in the nano-plastic regime for various glasses as a function of load using a unique nanoscratching technique on an atomic force microscope. Figure 4 shows the determined removal function on a log-log plot of removal depth versus load per particle for fused silica, borosilicate, and phosphate glass. At very low loads (<10^{-6} N), removal occurs by molecular interactions with the workpiece surface and no removal occurs. At intermediate loads (10^{-8} to 10^{-6} N, depending on the glass type), material removal occurs at the molecular level with an average removal depth in the Angstrom level. Finally at higher loads (>10^{-6} N), removal occurs by nano-plastic deformation in the nanometer regime whose removal depth can change with load and glass type. If particles are removing material in the plastic regime, this would result in higher roughness than if particles are removing material in the molecular regime. Finally, notice the fairly significant difference in the removal depth of the various glasses in the plastic regime (0.55 nm for fused silica, 0.85 nm for borosilicate glass, and 3.0 nm for phosphate glass). The next key step is to determine the load on each particle at the interface during polishing. Here, we formulate the EHMG model to determine the load per particle distribution (see Appendix for detailed description). The schematic setup of the EHMG model is shown in Fig. 8 where an ensemble of spherical polishing particles is embedded into the pad at the interface between the workpiece and polishing pad. The combination of the measured polishing slurry PSD, pad topography and mechanical properties of the particles, pad, and workpiece are used in two sets of load balances to calculate the load per particle distribution, the fraction of pad area making contact with the workpiece, and the fraction of applied load carried by the particles versus the pad. The first load balance is for the pad deformation which determines the overall compression of the pad (h_f) and hence fraction of pad area making contact with the workpiece (see top of Fig. 8 showing compression of all of the pad asperities). The second load balance is for determining the interface gap, g_i for each individual pad asperity i (see bottom of Fig. 8). The calculated load per particle distributions are shown in Fig. 4 for three slurries used in this study overlaid with the removal function described above. Large differences in the load per particle distributions are determined due to changes in the tail end of the distribution. The narrow PSD slurry (NanoArc 6450; d_{PSD} = 0.15 μm) leads to a lower load per particle distribution than the wider PSD slurry (Unstabilized Hastilite; d_{PSD} = 0.92 μm). The effects of pad topography, glass type, and slurry concentration have much weaker effects on the load per particle distributions (results not shown).

(2) Removal Rate
At the macroscopic level, material removal has been historically described by the widely used Preston equation.\textsuperscript{4,22}
\[
\frac{dh}{dt} = k_p \sigma_0 V_r 
\]

where \( \frac{dh}{dt} \) is the average thickness removal rate, \( \sigma_0 \) is the applied pressure, and \( V_r \) is the average relative velocity of the polishing particle relative to the workpiece. As generally observed, removal rate increases with an increase in applied pressure and lap-optic rotation rates. Using the EHMG model, a more fundamental expression of the removal rate is derived based on the summation of the removal of all the individual particles [see Eq. (A6) in the Appendix]. Rewriting in a more convenient form, the macroscopic material removal rate can be estimated using microscopic material removal parameters in the form:

\[
\frac{dh}{dt} \approx N_f f_x f_L V_r (f_p (d_p) - f_m (d_m) + f_m (d_m)) 
\]

where \( N \) is the areal number density of particles at the work-piece-pad interface, \( f_i \) is the fraction of active particles (i.e., particles resulting in removal) in the PSD, \( f_x \) is the fraction of pad area making contact with workpiece, \( f_m \) is the fraction of applied load carried by particles (rather than carried by the pad directly), \( f_p \) is the fraction of active particles that result in plastic removal, \( <d_p> \) is the average depth of particles that result in plastic removal, \( <d_m> \) is the average contact zone diameter of particles that result in plastic removal, \( f_m \) is the fraction of active particles that result in molecular removal, \( <d_m> \) is the average depth of particles that result in molecular removal, and \( <d_m> \) is the average contact zone diameter of particles that result in molecular removal. The terms in the parentheses in Eq. (3) represent the cross-sectional area of material removed from the workpiece surface; the first four terms before the parentheses represent the number of loaded particles per unit area resulting in removal.

Equation (3) provides significant insights as to the type of microscopic interactions that can influence the material removal rate during polishing. Hence, Eq. (3) can be utilized as a general tool for optimizing polishing conditions, complementing the standard practice of simply increasing load and velocity as suggested by the Preston Equation [Eq. (2)]. To test this, the EHMG model was used to calculate the critical parameters [as outlined in Eq. (3) and the Appendix] for a representative subset of polishing experiments described in Table I. The results are summarized in Fig. 9. The top of Fig. 9 shows the measured material removal rate for four experimental series. In each series, the removal rate increases with a different manipulated polishing variable: widening PSD, increasing slurry concentration, flattening pad topography, and change in glass type. For each series, the bottom row of Fig. 9 shows which parameter(s) in Eq. (3) (as calculated by the EHMG model) increases the most in response to changes in the manipulated variable. Note that the other calculated parameters in Eq. (3) were comparatively slowly varying and are not shown on the plot for simplicity of presentation. For each series, the model predicts a different parameter from Eq. (3) dominates the observed trends. Hence, the reason for the increase in removal rate is significantly different for each of the series at the microscopic scale.

Starting with the Slurry PSD series (column 1 of Fig. 9), the change in PSD has the biggest impact to the load per particle distribution as shown in Fig. 4. A change in the tail end of the PSD of the slurry influences the number of particles loaded (active particles) and the fraction of particles that remove material in the molecular versus plastic regime (\( f_p \)).

The increase in the removal rate is then dominated by the greater number of particles removing material in the plastic regime which has a larger removal depth per particle.

For the Slurry Concentration series (column 2 of Fig. 9), the removal rate initially increases and then plateaus at high slurry concentrations. The EHMG model predicts this is dominated by two factors: (1) the number of active particles (\( N \times f_x \)) and (2) the fraction of load carried by the pad (\( f_m \)). At low concentrations, both \( N \times f_x \) and \( f_m \) increase, resulting in an increase in removal rate. However, at higher concentrations, the number of active particles starts to decrease due to a slight increase in the load per particle from sampling more, larger particles in the distribution. Also, at higher concentration the fraction of load carried by the particles approaches and plateaus at 1 (i.e., there are so many particles at the interface that all the load is carried by particles and not any by the pad). Both the experimental and calculated data show a similar plateau effect.

For the Pad Topography series (column 3 of Fig. 9), the increase in removal rate with the diamond conditioning treatment (hence pad flattening) is dominated by the increase in the fraction of pad area making contact with the workpiece (\( f_x \)). Using the measured height histograms in Fig. 5, the calculated load pad balance [using Eq. (A2)] determines \( f_x \). The pad with tall asperities (new MHN pad) results in less overall contact between the pad and the workpiece (smaller \( f_x \)) and hence fewer particles at the interface contributing to the removal. As a result, flatter pad topologies lead to larger \( f_x \) and higher calculated removal rates, correlating well with the experimental data. Note, the rate of increase in \( f_x \) from 5 to 45 min diamond conditioning to 45 min diamond condition was proportionally smaller than the proportional increase in the measured removal rate. This is likely due to the fact that the spatial scale length for the pad topography used as input into the model was limited to 21.4 mm. Between the 5 and 45 min diamond conditioning, the flatness of the pad at longer spatial scale lengths also increases, influencing the overall fraction of the pad area making contact which is not currently captured in the EHMG model at these length scales. However, the experimental measurement of pad flatness at larger spatial scale lengths supports this trend [see Fig. 7(b)].

Finally for the Glass Type series (column 4 of Fig. 9), using the wider PSD slurry (Unstabilized Hastilite), the removal rate increase is dominated by the change in the single particle removal function, namely depth of plastic removal (\( d_p \)). The measured removal rate and calculated load pad balance [using Eq. (A2)] determines \( f_x \) and \( d_p \) increase by approximately the same amount going from silica to borosilicate to phosphate glass. Note that changes in the mechanical properties of the glass cause very slight changes in the load per particle distribution. The relationship between the removal function differences with the observed removal rates is consistent with the rest of the Glass Type—Slurry series samples shown in Fig. 3. For example, the narrow PSD slurries (such as NanoArc 6450 with the load per particle distribution as shown in Fig. 4) have essentially all particles operating in the molecular removal regime. The measured removal rate difference between the glass types with this slurry is much smaller due to the smaller difference in depth of molecular removal (\( d_m \)).

In summary, the experimental data are consistent with the relative removal rate calculated using the EHMG model. However, we imposed a number of simplifying assumptions and/or inferred values of some parameters that have yet to be measured experimentally, which prevent from a true quantitative prediction of the removal rate. First, the EHMG model assumes a simple correlation between the slurry volumetric concentration and the areal concentration of particles at the interface (\( N \)). In a real polishing system, this correlation is likely affected by colloidal chemistry, pad charge, pad topography, etc. which are not accounted for in the model. Second, for simplicity, the particles at the interface are treated as a monolayer of particles (see Fig. 8). In reality, the particles stack up in multiple layers (as evidenced by data in a previous study\(^{16}\)). Third, the larger particles in the PSD are assumed to have the same mechanical properties as the smaller particles. However, many of the larger particles are likely
agglomerates of the smaller particles, hence having an effective modulus that is lower than the smaller particles. Also, these larger particles may have a propensity to be broken into smaller particles or just prevented from entering into the workpiece–pad interface gap. Fourth, all particles are assumed to be stationary at the pad–particle interface (needed to achieve relative velocity between workpiece and particle), and the removal length due to the particle is assumed to be infinitely long. In other words, the effects of rolling, sliding, dislodging, and loss of reactivity of the particles are not accounted for in the model. Fifth, the removal depth due to molecular removal \( (d_m) \) has not been measured directly. To the authors’ best knowledge, a technique to quantitatively and directly measure this parameter has not been described. Instead, this value has been inferred using polishing removal rate data. Finally, \( d_m \) is clearly a function of chemistry of reaction which can be altered with various parameters which are not accounted for directly in the current EHMG model. Despite these assumptions and simplifications in the current EHMG model, it successfully predicts the removal rate trends over a broad range of the polishing parameters (slurry type, slurry concentration, pad topography, and workpiece glass composition).

### (3) Surface Roughness

Using the EHMG model and subsequent Monte Carlo simulations, simulated polished surfaces were created due to the summation of removal by individual particles for selected polishing experiments in the AFM roughness spatial scale length (5 μm × 5 μm) (see Appendix for description). Figures 10(a)–(c) compare the measured roughness (top) with that of the corresponding simulated surface (bottom) with changes in polishing parameters for a given polishing series (left to right). Figure 11 shows the same data summarized as rms roughness for quantitative comparison between the model and the experiment.

Figure 10(a) compares the effect of slurry PSD where it is apparent from the experimental results that widening the PSD resulted in an increase in surface roughness. The corresponding simulations show a similar trend. As also observed with the removal rate trend for this series, the increase in the fraction of particles removing material by nano-plastic mechanism \( (f_p) \) is the dominant reason for the increase in roughness with increase in width of the PSD \( (d_{PSD}) \). Since \( d_p > d_m \), the roughness increases.

Figure 10(b) compares the effect of pad topography. Experimentally, the roughness was observed to decrease with a more pad conditioning time (hence flatter pad topography); the simulated surfaces showed a weaker reduction in roughness with increase in flatness of the pad. As also observed with the removal rate trend for this series, this discrepancy is likely due to the small spatial length of the pad topography used in the model. Intuitively, the observed experimental trend is expected. A flatter pad would increase \( f_A \) (fraction of pad area making contact) which results in a greater number of particles making contact and a decrease in the average load per particle. Therefore, one would expect an increase in material removal rate, due to the greater number of particles making contact, and a drop in surface roughness, due to the decrease in average load per particle.

Finally, Fig. 10(c) compares the effects of glass type (using Unstabilized Hastilite slurry). Experimentally the roughness increased with glass type (silica < borosilicate < phosphate) as discussed earlier, matching the trend in simulation results. Due to the same reason for the removal rate increase, the roughness increase is attributed to increasing \( d_p \) (depth of plastic removal) with change in glass type. As can be seen in Fig. 11, the simulation results systematically underestimate the roughness compared to the experiments for the glass type.
This may be due to error in the value of $d_p$ determined or some other effect not yet captured in the EHMG model.

**Strategy for Optimized Polishing: Simultaneous High Removal Rate and Low Roughness**

The goals for an optical polishing process is to simultaneously and economically achieve: (1) the desired surface figure (overall wavefront), (2) high surface quality (low scratch/dig), and (3) low surface roughness. Cost reductions can be realized with short process times (i.e., high removal rates) and few process iterations and metrology cycles. Recently, a new method for full aperture polishing called convergent polishing was developed which directly addresses the first two goals.\(^{15,17}\) This pad polishing technique allows for finishing flat and spherical glass optics in which a workpiece, independent of its initial shape (i.e., its surface figure), will converge to a final surface figure with excellent surface quality under a fixed, unchanging set of polishing parameters in a single polishing iteration. In contrast, conventional full aperture polishing methods require multiple, often long, iterative cycles involving polishing, metrology, and process changes to achieve the desired surface figure. Using the EHMG model insights from this study, the convergent polishing method can be improved to also rapidly achieve low surface roughness without sacrificing surface figure or surface quality. Using narrow PSD slurries does decrease roughness but at the expense of a lower removal rate. However, exploring strategies to increase the fraction of pad area making contact ($f_A$) (resulting in more particles simultaneously removing material) may significantly increase the removal rate and simultaneously decrease the roughness. An example of this, demonstrated in this study, is to increase the flatness of the pad topography. Another strategy is exploring methods to increase $d_m$ (effect depth of molecular removal) through slurry chemistry to increase removal rate with an expected weak increase in roughness.

![Fig. 10. Comparison of measured AFM roughness (5 µm x 5 µm) with that simulated with the EHMG model for selected samples in the (a) slurry PSD series, (b) pad topography series, and (c) glass type series.](image-url)
face gap in the EHG model, here the interface consists of many interface gaps ($g_i$) due to the different asperity heights on the pad surface (see Fig. 8). Also, in contrast to the EHG model, the EHMG model determines the fraction of pad area making contact ($f_A$) using the height histogram of the pad, rather than relying on user defined input. In addition, with the EHMG model, the pad, in addition to the particles, can absorb some of the applied load in cases where the interface gap goes to zero.

The EHMG model description below is divided into the following steps: A.1) determination of pad deflection ($h_0$) and fraction of pad area making contact ($f_A$); A.2) determination of asperity stress distribution ($\sigma_i$), interface gap distribution ($g_i$), load per particle distribution [$P(r,g_i)$], and fraction of active particles ($f_i$); A.3) determination of the global material removal rate ($dh/dt$); and A.4) determination of the workpiece roughness using a Monte Carlo polishing simulation.

**A1. Pad Deflection and Fraction of Pad Area Making Contact**

Using a simple load balance, the applied pressure ($\sigma_o$) on the workpiece is balanced by the elastic response of the pad given by:

$$\sigma_o = E_2 \int_0^{h_i} \frac{h_i - h_t}{t_p} \, dh$$  \hspace{1cm} (A1)

where $E_2$ is the elastic modulus of the pad, $h$ is the asperity height, $h_t$ is the equilibrium pad deflection, and $t_p$ is the initial unloaded pad thickness (see Fig. 8). Equation (A1) can be solved numerically to determine $h_t$, which can be used to determine the fraction of pad making contact ($f_A$) using:

$$f_A = \frac{\int_{h_i}^{h_t} f_{pad}(h) \, dh}{\int_0^{h_i} f_{pad}(h) \, dh}$$  \hspace{1cm} (A2)

where $F_{pad}(h)$ is the cumulative height distribution of the pad topography at $h_t$.

**A2. Asperity Stress, Interface Gap, Load/Particle Distribution, and Fraction of Active Particles**

The stress on each pad asperity height $h_i$ at the equilibrium pad compression ($h_t$) is:

$$\sigma_i = \frac{h_t - h_i}{t_p} E_2$$  \hspace{1cm} (A3)

In addition, the stress on each pad asperity height carried by slurry particles is determined as:

$$\sigma_i = N_i \int_0^\infty 2f(r) P(r,g_i) \, dr$$  \hspace{1cm} (A4)

where $P(r,g_i)$ is the load on a given particle on given asperity of gap ($g_i$) and particle of radius ($r$) determined as:

$$P(r,g_i) = \frac{4}{3} E_{eff} \pi \sqrt{(2r - g_i)^3}$$  \hspace{1cm} (A5)

where $E_{eff}$ is the effective modulus (defined in Ref. [9]). Substituting Eqs. (A3–A5), the gap at each asperity height can be determined. Then it is straight forward to calculate the
stress and the load per particle distribution at each asperity height.

Two key extensions of the earlier EHG model to the present EHMG model can be seen in Eqs. (A3–A4). First, pad asperities of different initial heights are compressed with different amounts leading to different asperity loads as given by Eq. (A3) (see Fig. 8). Second, the interfacial gap is determined by the load carried by the particles as given in Eq. (A4), the gap becoming smaller as the load increases. It is evident from Eq. (A4) that the maximum load that can be carried by the particles is when they are completely embedded into the pad (i.e., when the gap goes to zero). If the total load on the asperity is larger than this maximum, the remainder of the load is carried directly by the pad and does not contribute to material removal. This is a source of deviation from the macroscopic Preston equation [Eq. (2)] behavior at high applied loads. The pad topography for Samples T1 and T3 has the same slurry PSD and thus the same interfacial gap determination. However, Sample T1 (polished on a new MHN pad with lots of tall asperities) has a greater fraction of active contact area with zero gap compared to Sample T3 (polished on highly DC MHN with much flatter pad surface). Hence, Sample T1 also has smaller fraction of the load being carried by the particles (see Discussion for further description).

A3. Material Removal Rate

Using a similar approach to that utilized in the EHG model,9 the macroscopic material removal rate can be estimated using average microscopic parameters. Here, the thickness removal rate is the summation of the removal due all the particles in the interface, which can be described as:

\[
\frac{dh}{dt} = \frac{V_r}{A_p} \sum_{j=1}^{N_t} d_j a_j
\]  

(A6)

where \(V_r\) is the relative velocity of the particle, \(A_p\) is the contact area between workpiece and pad, \(d_j\) is the depth of removal (defined by the removal function in Fig. 7) for a given particle \(j\), and \(a_j\) is the contact radius for a given particle \(j\). Depending on the load per particle, the removal can be 0 (no contact), \(d_m\) (molecular removal), or \(d_p\) (plastic removal) (see Fig. 4). The contact radius for each particle size and interface gap is given by:

\[
a(r, g) = \sqrt{r} \left( \frac{3 P(r, g)}{4 E_{13} r^2} \right)^{1/3}
\]  

(A7)

The average contact zone due to plastic removal \(<a_p>\) is determined by averaging overall asperities and active particles with \(P > P_{\text{crit}}\) and due to molecular removal \(<a_m>\) for \(P < P_{\text{crit}}\). By the same token, the fraction of particles that occur by plastic removal \((f_p)\) or by molecular removal \((f_m)\) are determined by summing up all the particles in each removal regime.

A4. Workpiece Roughness

With an established formalism for determining the contact characteristics and the material removal function for each slurry particle, the next step of creating a simulated polished surface can now be performed. Using a Monte Carlo simulation similar to that described for the EHG model in Ref. [9], workpiece topography and roughness were simulated now using the EHMG model. Slurry particles were selected randomly from the PSD [\(f(0)\)], and then randomly placed on a line direction on the surface of an initially flat workpiece within a defined microscopic window. The linear density of particles is given by \(N_1\), where \(N_1\) is the areal density of particles at the interface. Then, the particles made a line trench with a depth defined by the material removal function \((f(x))\) and a constant diameter of 2\(a\) [see Eq. (A7)]. The process was repeated for a large number of particles until the surface roughness stabilized.

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