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# Computational Analysis of Intratool Interactions in Electrochemical Micromachining with Multitip Tool Electrodes

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We have investigated the use of complex tool electrodes (templates) in electrochemical machining with ultrashort voltage pulses using a three-dimensional computational model. Through a systematic study of profile evolution varying pulse durations and separations between features, we have quantified the degree to which the performance of these templates is decreased relative to that of a simple tool electrode. We have found both the discrepancies and the separation at which the maxima occur are reduced with decreasing pulse duration. We explain these discrepancies through examining for the first time the effects of intratool electrode interactions on the overpotential and dissolution current at the substrate electrode. These studies have revealed that overpotential is largely additive among the individual components of a tool electrode above a critical separation between components, while the exponential nature of the dissolution current with respect to overpotential leads to increased dissolution when using a complex tool electrode.
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The machining of electrically active substrates on the micrometer scale and smaller is an active research area with numerous competing technologies.[1-3] Electrochemical machining has typically been considered ill-suited for such precision work due to an inability to confine the location of the dissolution, but recently the use of ultrashort voltage pulses[4-8] has largely overcome this limitation. While this technique offers improvements over other methods, particularly with respect to the production of 3D and high aspect ratio features and the limiting of surface damage, it at present suffers from limited processing speeds.[5] This is exacerbated by the nature of the tool electrode and how it is used to create patterns in the substrate surface in many experimental systems. At present, the majority of experimental studies employ a simple tool electrode (such as an STM tip) which is used to trace out the entire design in the substrate in series. For many patterns, however, this process could be parallelized with the appropriately-shaped tool template. This approach is made more viable by the lack of degradation of the tool electrode during electrochemical processing but may be hindered by reduced fidelity in the communication of the tool shape to the substrate relative to that of the simple tool electrode.

In this article, we investigate the use of tool templates in electrochemical machining with ultrashort voltage pulses using a three-dimensional computational model consisting of simulations of the potential evolution and profile evolution. The substrate morphologies formed by a simple tool electrode are compared with those of the equivalent tool template for a variety of pulse durations. The differences between the two types of etching are then explained by the natures of the potential evolution and dissolution current evolution. For the first time, we examine the effects of communication between different portions of a complex tool electrode on these quantities.

The simulation of the evolution of the potential during the voltage pulse is based upon a continuum-level model of the electrochemical double layers and the electrolyte, depicted in Fig. 1a. The tool and substrate are considered as idealized polarizable electrodes, and thus the double layer at each surface is represented by capacitors in parallel. The resistance of the electrolyte is modeled as a mesh of interconnected resistors, which also connects to the capacitors at each surface. Upon application of a voltage pulse, the initial potentials at nodes within the electrolyte are calculated, with reflective boundary conditions employed as appropriate. Then, at set intervals, the capacitors are charged by the average local current during the interval, with the updated potentials of nodes within the electrolyte solved simultaneously. Details of the technique for a two-dimensional system may be found elsewhere.[9-11]

The transient overpotential information obtained from the above potential evolution is used to determine the local dissolution current density at the substrate surface. This is modeled according to Tafel behavior,[12] as large overpotentials are expected during the latter stages of the voltage pulse, when the dissolution current will be at its maximum. In this case, the dissolution current density is proportional to an exponential function of the overpotential, $j = j_0 \exp(\alpha F\eta/RT)$, where $j_0$ is the exchange current density, $\alpha$ is the transfer coefficient, and $F$ is the Faraday constant. Integrating this quantity during the length of the pulse and normalizing by the combined length of the pulse and pause periods gives the spatially-varying average dissolution current density, which is proportional to the local etch rate. This information can then be coupled with an interface tracking method (here, the level set method[13]) to predict

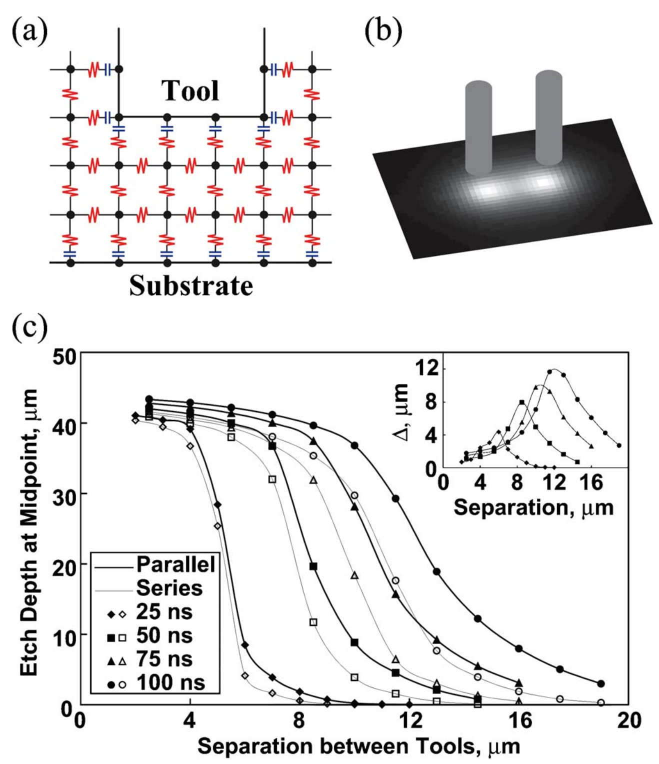


**Figure 1.** (a) Schematic of the simulation domain. (b) Schematic of the parallel etching system. (c) Midpoint etch depth vs tool separation for the parallel and series cases at the indicated pulse durations. Inset: Difference in midpoint etch depth (parallel case−series case).

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substrate modifications due to the dissolution process and to obtain the spatially-varying, transient behaviors of the dissolution current density.

In order to investigate the use of complex templates for tool electrodes, we consider the etching of two high aspect ratio holes in close proximity to one another, as depicted in Fig. 1b. These holes may be etched by a tool consisting of a single cylinder with two vertical etches in series or in a single etch by a more complex tool consisting of two cylinders with the appropriate spacing, generating the two holes in parallel. Upon simulation of these two methods, however, we note significant differences in the substrate surface morphologies, particularly when comparing the region lying between the holes. For the series case, little etching occurs in this region unless the holes themselves overlap—that is, a single hole generated by the tool extends beyond the midpoint between the holes. In contrast, the parallel process often gives rise to significant etching in this region, even when the resolution of a single hole would not contain the midpoint.

To quantify the difference between the parallel and series etch behaviors, the etch depth at the midpoint between the hole centers was compared for a variety of pulse durations and tool separations, as shown in Fig. 1c. Here, tools consisting of one or two 5 μm diameter cylinders are lowered 40 μm into a copper substrate under conditions matching a recent experimental study:[6] a velocity of 1.5 μm/min, −2.3 V pulses ranging from 25 to 100 ns, a 1:10 pulse:pause ratio, and a 0.1 M $CuSO_4$/0.075 M $H_2SO_4$ electrolyte solution. In the case of one cylinder, the tool is removed after the initial etch and moved laterally across the substrate surface (both without voltage pulsing), then lowered to a depth of 40 μm a second time under etch conditions. The separation between tools is defined as the minimum distance between the outer circumferences of the tools for the parallel etching case or the analogous minimum distance between the outer circumferences of the initial tool positions for the series cases. Note that, for both parallel and series etching, the trend in midpoint etch depth vs tool separation is the same for all pulse durations, while for small separations, the etch depth is roughly equal to the tool etch depth. As the separations get larger, a critical separation is reached at which the etch depth declines rapidly, ultimately leading to no significant etching in the midpoint region. The disparities in the behaviors become clearer when looking at the difference in etch depth between the parallel and series cases at the same tool separation, as illustrated in the inset in Fig. 1c. Here we see that, for each pulse duration, there is a critical separation between tools at which a maximum disparity is found between the series and parallel etching cases, with both the critical separation and disparity increasing with increasing pulse duration. Thus, as the pulse duration is decreased to allow a smaller resolution and better communication of the tool shape to the substrate surface, the advantage of the series case over the parallel case shifts to tool features in closer proximity to each other.

To further understand the different etch behaviors for the two cases, we examined the overpotential on the substrate surface throughout the duration of the pulse, as described in Fig. 2. For this analysis, we chose a 50 ns pulse and a separation of 8 μm between tools, near the critical separation for this pulse duration where any discrepancies between the behaviors should be at a maximum. Two etch conditions were considered: an early stage when the tools had just reached the initial substrate surface and when the tools were 40 μm below the initial surface (and etching was steady-state). For both conditions, the substrate formed during the parallel etch process was used as a basis and the evolution of the overpotential was calculated for both the parallel and series cases (with the calculations for the two series locations made individually). The overpotentials at the end of the pulse for the parallel case were then compared with the addition of the overpotentials for the series cases, for both the initial etch (Fig. 2a) and steady-state condition (Fig. 2b). Discrepancies reached a maximum of 0.10 V between the two cases, less than one-sixth of the maximum overpotential for the parallel case, with all differences revealing higher values for the combined series case. Thus, adding the overpotentials of individual tools from the series cases gave approximately the overpotential of the parallel case, with any interaction between the tools resulting in a decreased rather than increased overpotential.

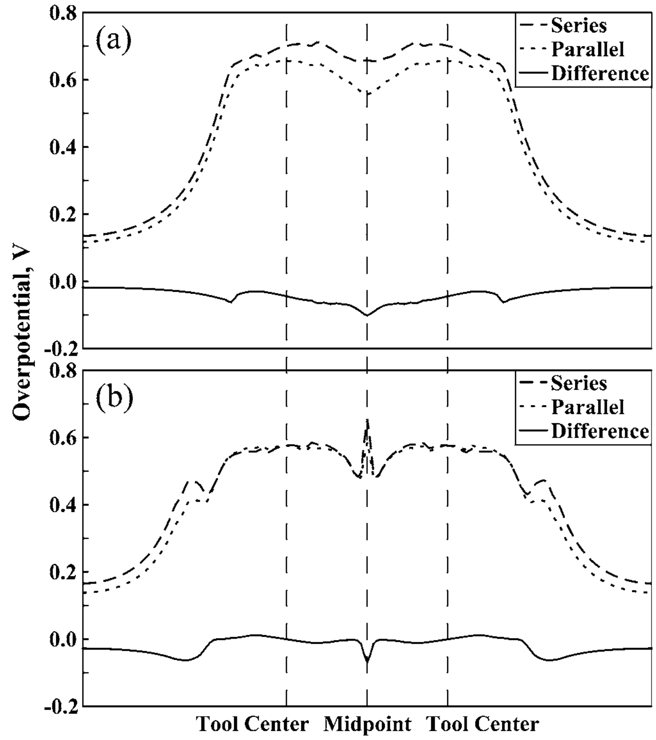


**Figure 2.** Cross sections showing the overpotential at the end of a 50 ns pulse at the substrate surface for tool located at (a) the initial substrate surface and (b) 40 μm below the initial substrate surface. Tool separation is 8 μm. The series overpotential is the sum of the individual tool contributions. The difference is defined as parallel−series.

A similar analysis for the dissolution current gives markedly different results, as shown in Fig. 3. Here, the dissolution currents at the end of the pulse for the parallel cases are greater than the addition of their respective series cases in almost all regions. For the initial etch stage (Fig. 3a), this difference is largest at the regions of the substrate shadowed by the tool but is also significant in the region lying between the tools. Once steady-state etching has begun, however, the difference is largely confined to the area of the substrate between the tools (Fig. 3b). This behavior can be explained by the exponential nature of the dissolution current with respect to the overpotential, following the Tafel model. Assuming that the overpotential for the parallel case is essentially additive of the series cases in all regions, the comparison for the dissolution current is between quantities of the form $\exp(\eta_{Tool1} + \eta_{Tool2})$ and $\exp(\eta_{Tool1}) + \exp(\eta_{Tool2})$ for the parallel and series cases, respectively. Thus, the dissolution current can be significantly larger for the parallel case, particularly in those regions having the largest overpotentials, such as those opposite the cylindrical tools and lying between them. As the vertical etching process begins, the contribution of one tool to the hole formed by the other is decreased due to the obstruction of the substrate itself, leading to the bottoms of the holes for the parallel case generally matching those of the series case. The region lying between the holes, however, is still affected by phenomenon described above, leading to the increased etching seen for the parallel case.

As larger arrays of tools were considered, the combination of overpotential contributions from individual tools was no longer a good approximation of that generated by the tools in parallel, as illustrated in Fig. 4a. Here the overpotential at the end of a 50 ns pulse from an array of 5 μm diameter tools separated by 8 μm in

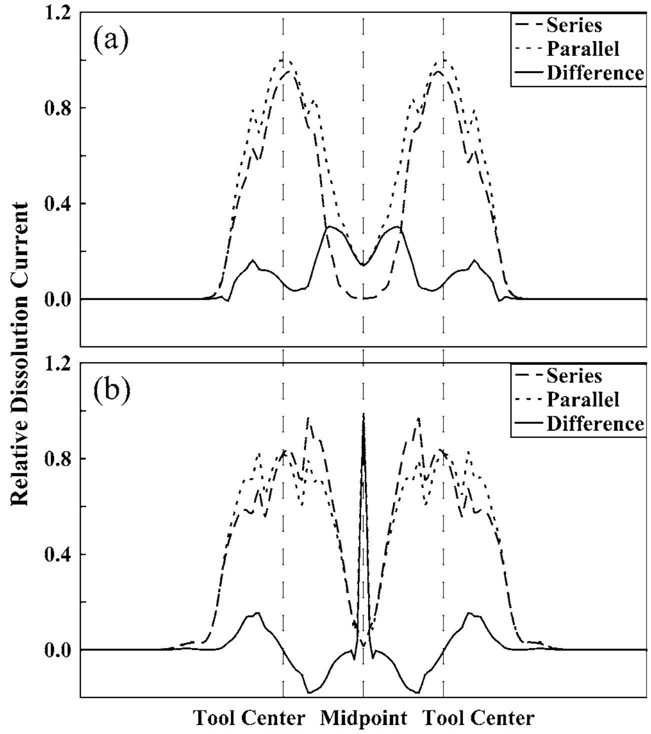


**Figure 3.** Cross sections showing the relative dissolution current at the end of a 50 ns pulse at the substrate surface for tool located at (a) the initial substrate surface and (b) 40 μm below the initial substrate surface. Tool separation is 8 μm. The series dissolution current is the sum of the individual tool contributions. The difference is defined as parallel−series.

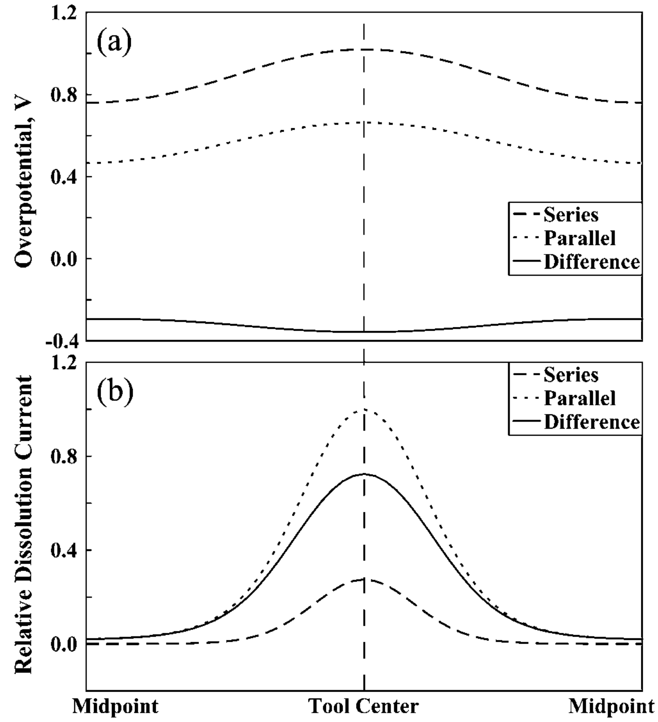


**Figure 4.** Cross sections showing the (a) overpotential and (b) relative dissolution current at the end of a 50 ns pulse at the substrate surface for the unit cell of an array of tools separated by 8 μm in both *x*- and *y*-dimensions. Tools were held 3 μm above an unmodified substrate. The series quantities are the sum of the individual tool contributions to the central unit cell of a 5 × 5 cell region. The difference is defined as parallel−series.

both the *x*- and *y*-dimensions (simulation domain of size 13 × 13 μm) was compared with the combined overpotential of 25 individual tools, each using a simulation domain of size 65 × 65 μm. For these calculations, an unmodified substrate a distance of 3 μm from the tool tip(s) was used, and the 25 individual tools were located to match the locations of the tools of a 65 × 65 μm portion of the tool array. In all regions, the combined overpotential was larger than that of the parallel case. Further analysis revealed the discrepancies decreased with increasing tool separation, with the system reaching the limiting case of approximately additive behavior at a separation of 16 μm in both *x*- and *y*-dimensions. A similar pattern of behavior was observed for a variety of pulse durations and separations between tool(s) and substrate: At small separations between tools in the array, the combined overpotential of individual tools exceeded that of the parallel tool in all regions, while at larger separations the system became additive, and thus $\eta_{parallel} \leqslant \Sigma\eta_{series}$.

This behavior of the dissolution current was as before, however, as shown in Fig. 4b. Discrepancies in all regions of the simulation domain were in favor of the parallel case over that of the combined series cases. Similar to the overpotential case, further studies indicated these differences decreased as the tool separation increased. This was once again a function of the exponential nature of the dissolution current with regards to the overpotential but was now not so simply deconstructed as the earlier case with two tools, as $\exp(\eta_{parallel})$ is no longer approximately equal to $\exp(\Sigma\eta_{series})$ at small separations.

## Conclusion

We have shown that the use of tool templates can lead to a degradation in the communication of features to the substrate surface relative to that of a simple tool, depending on the proximity of those features to one another and the pulse duration used during the etch process. This phenomenon can be explained by the exponential nature of the dissolution current with respect to the overpotential. While the overpotential generated by the tool template is often roughly equivalent to that of the sum of its parts considered individually, the corresponding dissolution currents can vary widely, as the comparison is between a summing of exponents (thus, multiplicative) and the sum after individual exponentiation (additive). Therefore, care must be taken when designing tool templates to account for the pulse duration to be used, insuring that features in close proximity to each other do not merge.

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