Plasma-assisted electrochemical machining of microtools and microstructures

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Abstract

A novel hybrid electrochemical machining (ECM) approach combining a pulsed cathodic plasma and an electrochemical machining process, namely, plasma-assisted electrochemical machining (PA-ECM), is proposed in this study to strengthen the capacity of ECM, which entails both high efficiency and precision. The plasma characteristics, material removal behavior, surface topography and machining precision of PA-ECM for microtool fabrication are experimentally investigated under various conditions. The results show that PA-ECM can be realized under optimized electrical potentials with a vapor gaseous skin and electrolytic plasma layer formed around the cathode tool, of which the kinetic and thermal energies can enhance both the kinetics of the electrochemical reaction and mass transport during the ECM process. Through the design of the pulse voltage waveform, the formation and transportation of gaseous bubbles and plasmas can be well controlled. It has been shown that PA-ECM is effective and efficient for improving both the material removal rate and form accuracy in machining microtools. In the presence of plasma, a microrod tool with a high aspect ratio of 55:1 is successfully machined by PA-ECM in 5 s from its original diameter of 200 μm to approximately 18 μm, which seems to be the highest machining rate achieved so far. Additionally, in comparison to traditional ECM under the same conditions, PA-ECM provides a noticeable improvement in the microrod tool straightness error from 66.8 μm to 14.6 μm owing to the side surface insulation effect of the gaseous skin. The resulting surface roughness Ra is drastically reduced from 1096 nm to 46 nm, demonstrating that PA-ECM provides an innovative way to considerably improve the ECM efficiency without compromising the surface finish. Furthermore, the PA-ECM of microholes and microstructures are exhibited with improved precision, demonstrating the capacity of PA-ECM for micromachining.

1. Introduction

Electrochemical machining (ECM) is an established metal shaping process based on anodic dissolution, ideal for creating complex macro- and microstructures in extremely hard or exotic materials that are difficult to machine with conventional methods, such as superalloys, titanium alloys, and powdered metals [1–3]. In ECM, a low DC voltage of 5–20 V is applied across the interelectrode gap between the pre-formed cathode tool and the anode workpiece, while the electrolyte flows through the gap at a high speed of 10–60 m/s [3]. Anodic dissolution occurs, and the workpiece material is removed atom by atom obeying Faraday’s laws of electrolysis. Featuring a no-contact process, ECM can eliminate the mechanical force effects in the process, resulting in a burr-free part without thermal or mechanical stresses, microcracks, white layers or tool electrode wear, which finds wide industrial applications in turbines, dies and molds and medical implants [4,5]. However, the side gap between an electrode and the workpiece in conventional ECM is large due to unwanted stray machining, which renders ECM unsuitable for micromachining until 2000. An improvement in the ECM process involves techniques such as the use of an insulated sidewall of the tool [6], a low concentration electrolyte [7], smaller working gaps [8], and pulsed voltages [9] to improve the precision. In 2000, Schuster et al. [10] demonstrated that by applying nanosecond ultrashort voltage pulses, electrolysis dissolution can be confined to electrode regions in close proximity to achieve micrometer or submicrometer precision machining [10,11]. Han et al. 
used a wire electrochemical grinding method to fabricate a microrod (a diameter of 35 μm and a length of 163 μm), which is close to the micromachining ability of micro-EDM [12]. However, ultrashort pulse technology considerably reduces the machining efficiency due to the low duty ratio, which significantly limits its application. For example, in micro-ECM, the tool feed rate is only a few microns per second, which is much slower than that of micro-electrical discharge machining (EDM) [13]. To meet the industry requirements, through-mask electrochemical micromachining has been extensively used to enable mass-produced microcomponents, in which the substrate is covered with an insulating mask and dissolution proceeds only through the mask openings [14]. The preparation of the mask, however, complicates the ECM process.

To enhance the machining efficiency, another major ECM development is its combination with other machining processes, namely, hybrid processes. There have been several attempts to hybridize ECM with different physical or chemical mechanisms to achieve better process performance. For example, since the micro-ECM machine is similar to that of micro-EDM, a hybrid processing method of electrochemical discharge machining (EDCM) that combines both EDM and ECM has been developed for micromachining, in which EDM is used for roughing cut and ECM is used for finishing [15]. The process can reduce the machining time while increasing the machining accuracy and surface quality. As a further improvement, low-resistance deionized water is exploited to concurrently facilitate the electrical discharge and electrochemical reaction in a unique process with appropriate process control, which provides improved surface integrity and dimensional accuracy [16,17]. Nevertheless, the inevitable tool wear problem and thermally induced subsurface damage layer that is left on the workpiece is a drawback for precision machining. Pajak et al. developed a hybrid process of laser-assisted jet electrochemical machining (LAJECM), which combines a laser beam with an electrolyte jet for parallel processing [18,19]. The laser does not remove any material but assists electrochemical dissolution by thermal activation, as thermal energy enhances the kinetics of electrochemical reactions, providing faster dissolution. However, via this method, it is difficult to fabricate high aspect ratio complex shapes because an electrolyte jet is used as the tool. Another hybrid ECM process includes ultrasonically assisted electrochemical machining to improve the process efficiency by enhancing the interelectrode gap conditions [20]. All the hybrid methods described above require an auxiliary machining system, which results in increased process complexity and cost. On the other hand, electrolytic plasma processing (EPP) techniques are a hybrid of conventional electrolysis and atmospheric plasma processes, in which the application of an electrical potential is significantly greater than that of conventional electrolysis and leads to the formation of a continuous gas envelope around either the cathode or the anode accompanied by a plasma discharge [21,22]. EPP utilizes the basic principle of an electrolytic plasma to enable different chemical, electrical, mechanical and thermal interactions to take place on the electrode surface, which provides unique characteristics. To date, EPP has been widely used to engineer metal surfaces, such as plasma-assisted electrolytic cleaning [23], plasma electrolytic polishing (PEP) [24] and plasma electrolytic oxidation (coating growth) [25]. Nevertheless, most studies have concentrated on surface engineering only. Targeting engraving nonconductive materials, the spark-assisted chemical engraving (SACE) method has been developed to chemically etch glasses, utilizing the local high temperature caused by spark discharge [26–28].

In this study, intending to strengthen the capability of ECM for microfabrication, which entails high process efficiency as well as a good surface finish with high dimensional accuracy, an original hybrid machining process combining ECM with a cathodic plasma, namely, plasma-assisted electrochemical machining (PA-ECM), is proposed. In this method, material removal phenomena under high electrical potentials are exploited in such a way that the electrochemical dissolution is enhanced by the induced electrolytic plasma discharges. To fulfill this objective, an original approach, which is conceptually different compared to the aforesaid and other previous studies, is proposed. By doing so, ECM with enhanced machining efficiency and dimensional accuracy can be achieved.

2. Principle of PA-ECM

The proposed PA-ECM method for machining a microrod tool and a microstructure is schematically depicted in Fig. 1. The scheme of ECM is also included for comparative purposes. In both methods, the same salt aqueous solutions with a high conductivity serve as the electrolyte to enable high current densities and removal rates. However, unlike ECM, which generally utilizes pulsed DC voltages of 10–20 V with an anodically polarized workpiece, a pulse train that includes a relatively higher voltage, as shown in Fig. 1(a) and (b), is applied in PA-ECM. The substantial phenomenon of the PA-ECM process is the induced formation of a cathodic plasma envelope around the working electrode throughout the high voltage duration. With an increase in the voltage, hydrogen gas evolution which surrounds or interacts with the electrode material [29] can be significantly promoted at the cathodic electrode interface. At a critical current density value, a thin vapor gaseous skin that surrounds the working electrode can be formed, which behaves like a dielectric medium and an insulator between the electrode and the electrolyte [30]. The insulation of the tool electrode almost ceases the current flow and develops a high electric field of 10^6 V cm^-1 [21], resulting in plasma discharge across the vapor gaseous skin. The plasma can facilitate the impingement of a large number of electrons on the working electrode surface, which leads to rapid ohmic heating and the vaporization of the electrolyte surrounding the working electrode. The use of plasma energy, similar to that in laser-assisted jet ECM [31], promotes the subsequent anodic dissolution in the positive pulse duration by enhancing the kinetics of the electrochemical reactions. First, a localized increase in electrolyte temperature causes a higher local electrolyte conductivity and a higher local current density. Second, a higher temperature in the localized zone leads to a lower reaction activation energy [32,33]. Furthermore, the reaction product transportation processes at the interelectrode gap can be enhanced by the hydrodynamic flow resulting from the pulsating plasma discharge, which allows intense surface cleaning for polishing [23]. In addition, the plasma discharge is helpful for removing the oxidation skin for machining highly passivating materials. On the other hand, the formed vapor gaseous skin in PA-ECM can insulate the tool electrode and suppress unwanted side surface stray machining, leading to enhanced machining accuracy. By controlling the process conditions, the plasma discharge energy can be kept low to render little impact on the working electrode.

Table 1 shows a comparison of the similar processes of PA-ECM, all involving an electrolytic plasma. While SACE [26–28] focuses on chemically engraving nonconductive materials and PEP is applied for surface engineering, PA-ECM aims at metal shaping with both high efficiency and surface integrity. From the perspective of the material removal mechanism, while SACE involves discharge melting, vaporizing, and chemical etching, PA-ECM is primarily determined by electrochemical reactions such as anodic dissolution/oxidation combined with plasma-chemical reactions.

3. Experimental method and setup

To validate and characterize the proposed method, the PA-ECM experiments of microrod fabrication were carried out. Classical ECM experiments were also conducted for comparison purposes. A microrod was set as the machining target to simplify the experiments. A thin and hard microrod with a high aspect ratio and high surface integrity has been widely utilized as an essential tool for micromachining, scanning probe microscopy and medical measuring instrument applications [37, 38]. Considerable research has been undertaken to electrochemically fabricate microrod tools, for example, electrolyte jet turning [39,40], wire electrochemical grinding [12], micro-ECM [41], and
To facilitate the investigation, the ECM system shown in Fig. 2 was implemented in the study considering the ease of operation. A tungsten microrod workpiece was dipped in an electrolytic cell. A bipolar voltage, as shown in Fig. 2 (b), was applied between the workpiece and counter electrode, which were separated by a distance of a few centimeters. When a positive pulse was applied to the microrod, anodic dissolution occurred, resulting in the material removal from the microrod, which is the same as that during conventional ECM. In the negative pulse time, a relatively high voltage was applied to promote cathodic hydrogen gas evolution and induced a plasma discharge, which enhanced the subsequent anodic dissolution in the following positive pulse. A microrod can thus be machined by repeating the bipolar pulse train. The electrochemical reactions involved at the working electrode surface can be expressed as follows:

In the positive pulse (anodic):

\[
W + 8\text{OH}^- - 6e^- \rightarrow \text{WO}_4^{2-} + 4\text{H}_2\text{O} \quad (1)
\]

\[
4\text{OH}^- - 4e^- \rightarrow 2\text{H}_2\text{O} + \text{O}_2 \uparrow \quad (2)
\]

In a negative pulse (cathodic):

\[
2\text{H}_2\text{O} + 2e^- \rightarrow \text{H}_2\uparrow + 2\text{OH}^- \quad (3)
\]

Fig. 3 shows the experimental setup. A numerically controlled motion platform (V-731, Physik Instrumente) was employed to implement the experiments. A bipolar amplifier (HSA4014, NF) was used to supply bipolar voltage pulses, of which the output could be arbitrarily controlled by a function generator (33612A, Keysight). A tungsten rod with an initial diameter of 200 μm was immersed in the electrolyte to a depth of 1 mm and positioned using the motion stage to achieve optimal process conditions. The electrolyte consisted of a 5 mol/L aqueous solution of NaOH. A graphite plate of 50*30*5 mm in size was employed as the counter electrode. During machining, the voltage and current transient values were monitored in real time using an oscilloscope (MDO34, Tektronix) with an appropriate voltage (TPP0500B, Tektronix) and current (TCP0030A, Tektronix) probes.

For comparison purposes, both conventional ECM and PA-ECM experiments were carried out to investigate the difference in machining performance. Table 2 shows the specific experimental conditions, and the pulse waveforms used in ECM and PA-ECM are shown in Fig. 4. The main difference between ECM and PA-ECM was the negative-pulse voltage. In contrast to 0 V in ECM, a high negative voltage of -32 V was applied in the negative pulse of PA-ECM to generate plasma discharges. In the experiments, the amplitude and duty ratio of the positive

![Fabrication of microtool](image1.png)

![Machining of microstructure](image2.png)

**Fig. 1.** Schematic diagram of plasma-assisted electrochemical machining (PA-ECM) for microtool and microstructure fabrication, with the conventional ECM method [34–36] included here for comparison.

### Table 1

<table>
<thead>
<tr>
<th>Electrochemical Plasma Processes with PA-ECM [21,23,24,26–28]</th>
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<tbody>
<tr>
<td>Workpiece polarity</td>
</tr>
<tr>
<td>Plasma generation</td>
</tr>
<tr>
<td>Current density ( J )</td>
</tr>
<tr>
<td>Target</td>
</tr>
<tr>
<td>Applicable material</td>
</tr>
<tr>
<td>Applied voltage</td>
</tr>
</tbody>
</table>

electrochemical etching [34–36].

To facilitate the investigation, the ECM system shown in Fig. 2 was implemented in the study considering the ease of operation. A tungsten microrod workpiece was dipped in an electrolytic cell. A bipolar voltage, as shown in Fig. 2(b), was applied between the workpiece and counter electrode, which were separated by a distance of a few centimeters. When a positive pulse was applied to the microrod, anodic dissolution occurred, resulting in the material removal from the microrod, which is the same as that during conventional ECM. In the negative pulse time, a relatively high voltage was applied to promote cathodic hydrogen gas evolution and induced a plasma discharge, which enhanced the subsequent anodic dissolution in the following positive pulse. A microrod can thus be machined by repeating the bipolar pulse train. The electrochemical reactions involved at the working electrode surface can be expressed as follows:
voltage were varied for a comprehensive study. The duty ratio in this study is defined as follows:

\[ \text{Duty ratio} = \frac{\text{Positive pulse time}}{\text{Pulse period}}. \]

The microrod tool shape and surface morphology before and after the process were examined by a scanning electron microscope (TM4000Plus, Hitachi). The surface roughness of the resulting microrod was measured by a white light interferometer (CCI HD, Taylor Hobson). In-process observation of the machining phenomena was conducted using a high-speed video camera (Dimax HS1, PCO). Meanwhile, a thermal camera (T660, FLIR Systems) was employed to detect the temperature change at the electrode surface during machining. The resulting workpiece profile was measured using a confocal laser scanning microscope (VK-X1000, Keyence). The process mechanism was discussed in-depth based on the experimental results.

4. Necking phenomenon and evaluation of machined microrod tool precision

In this study, to investigate the effects of the induced plasmas and bubbles, no rotation or vibration was applied to the rod. Typically, a neck-shaped rod, as illustrated in Fig. 5, results from traditional ECM. Electrochemical machining takes place both at the frontal area and side surface of the rod. The necking is caused by a viscous layer that is formed by the etching products of oxides, metallic cations, and anions on the working electrode surface. The thicker viscous layer moves downward along the tungsten rod by gravity and accumulates at the bottom, which causes a serious concentration polarization and results in a higher etching rate at the shank of the electrode. Nevertheless, the phenomena can be eliminated by applying a vibration to the rod or using a linearly decreasing voltage or duty ratio [35,36], which, however, is not the

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**Fig. 2.** Schematic diagram of plasma-assisted electrochemical machining of a microrod tool with a bipolar pulse train. *(U_p is the positive pulse voltage, U_n is the negative pulse voltage, t_p is the positive pulse time, and t_n is the negative pulse time).*

**Fig. 3.** (a) Picture of experimental setup for PA-ECM. (b) Electrolytic cell for machining microrods with electrodes separated by several centimeters. (c) The intentionally induced cathodic plasma on the microrod electrode surface.

<table>
<thead>
<tr>
<th>Table 2</th>
<th>Experimental conditions for ECM and PA-ECM of microrod.</th>
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<tbody>
<tr>
<td>Item</td>
<td>ECM</td>
</tr>
<tr>
<td>Positive pulse voltage, ( U_p ) [V]</td>
<td>1–5, 30</td>
</tr>
<tr>
<td>Negative pulse voltage, ( U_n ) [V]</td>
<td>0</td>
</tr>
<tr>
<td>Duty ratio of the positive pulse, ( D ) [%]</td>
<td>50, 70, 90</td>
</tr>
<tr>
<td>Pulse frequency, ( f ) [kHz]</td>
<td>1</td>
</tr>
<tr>
<td>Workpiece electrode</td>
<td>Tungsten rod, 200 μm in diameter</td>
</tr>
<tr>
<td>Immersion depth, ( H ) [mm]</td>
<td>1</td>
</tr>
<tr>
<td>Processing time, ( t ) [s]</td>
<td>5, 12</td>
</tr>
<tr>
<td>Electrolyte</td>
<td>5 mol/L NaOH aqueous solution (36 S/m)</td>
</tr>
</tbody>
</table>
To evaluate the resulting microrod form accuracy, the microrod diameter and rod straightness error are investigated, which are defined as follows:

Rod diameter = \( \frac{\text{Maximum diameter} + \text{Minimum diameter}}{2} \)

Rod straightness error = \( \text{Maximum diameter} - \text{Minimum diameter} \).

In addition, the material removal rate (MRR), which was defined as the volume of material removal per unit time, was introduced to quantitatively evaluate the process efficiency. The rod volume change was calculated based on the measured profile of the rod before and after etching, as illustrated in Fig. 5.

5. Results and discussion

5.1. Comparison of machining efficiency between PA-ECM and ECM

In the initial experiments, relatively low positive voltages of 1–5 V were applied to achieve stable etching. Fig. 6(a) and (b) shows the captured electrode surface phenomena of the microrod during ECM and PA-ECM, respectively. According to the experimental observations, mild etching was confirmed in the ECM accompanied by a small quantity of oxygen bubble evolution due to the anodic reaction. In contrast, PA-ECM presented an evidential plasma discharge, as shown in Fig. 6(b), together with a mass of bubbles. The SEM images of the resulting microrod are shown in Fig. 7. In the ECM method, as shown in Fig. 7(a)–(e), the material removal can hardly be confirmed, and the microrod diameter shows almost no change even under 5 V. In the PA-ECM method, however, according to Fig. 7(f)–(j), considerable material removal is observed. According to the diameter and surface roughness data shown in Fig. 8, it is shown that the microrod is hardly machined by ECM, while in PA-ECM, the reduction in the rod diameter was several tens of time that of ECM, and the diameter decreased linearly with increasing positive voltage, indicating an increased dissolution rate. Notably, a minimum rod diameter of 70 \( \mu \text{m} \) was obtained under 5 V within 12 s by PA-ECM. The rod straightness error, as shown by the dotted line in Fig. 8(a), increases slightly with the voltage increase.

Meanwhile, as shown in Fig. 7, a smooth and defect-free surface topography, the same as that in ECM, is obtained by PA-ECM. According to previous literature, as a cathodic electrolytic plasma, the maximum temperature achievable during cathodic polarity can be higher than 500 \(^\circ\text{C} \) [32, 33]. However, in the present results, no direct influence of the temperature on the machined surface is found, indicating that the cathodic plasma exerts little impact on the machined surface and that the material removal is primarily determined by anodic dissolution and plasma-chemical reactions. On the other hand, the surface roughness of the resulting microrod in Fig. 8(b) shows a clear reduction with increasing positive voltage in both methods. The rod roughness resulting from ECM is slightly smaller than that by PA-ECM, which indicates that the ECM method has a better electropolishing effect than PA-ECM. Generally, electropolishing should operate under a diffusion-limited constant current plateau, achieved by the formation of a viscous layer, to realize the anodic leveling of the workpiece surface. In PA-ECM, however, the pulsating gaseous skin and plasma discharge disturb the viscous layer formation, thereby resulting in a slightly larger surface roughness compared to ECM.

In PA-ECM, the MRR increases linearly with increasing positive voltage due to the improved anodic current density (Fig. 9). In addition, it is found that the microrod length is shortened under higher voltages. As shown in Fig. 9, at low voltages below 3 V, the resulting microrod length is the same as the preset length of 1 mm. At higher voltages of 4 V and 5 V, however, the rod length is shortened to 955 \( \mu \text{m} \) and 742 \( \mu \text{m} \), respectively. It is considered that the current density is concentrated on the tip and causes excessive etching. Therefore, the pulse voltage and
duration must be appropriately controlled in PA-ECM to avoid tip overetching.

On the other hand, the counter electrode of graphite showed little wear under the bipolar pulses. However, after a long usage time, the surface of graphite was altered with slight pitting corrosion.

5.2. Enhancement of the mass transport and anodic reaction by plasma discharges

The previous section demonstrates that PA-ECM can effectively machine microrods with an MRR much higher than conventional ECM. To clarify the process mechanism, the current-voltage characteristics in a pulse train period were analyzed.

Figs. 10 and 11 show the measured current waveform and the amplitude of the current, respectively. During the positive pulse time, the current is constant, and it increases linearly with increasing pulse voltage in both the ECM and PA-ECM methods. The current amplitude, however, shows a considerable difference. As shown in Fig. 11, the anodic current amplitude of PA-ECM is approximately 1.3 times that of ECM under the same positive pulse voltage. In the negative pulse time, the current is approximately zero for the ECM as the power output is zero. In PA-ECM, however, the current shows a sharp increase to approximately 3 A, right after the negative pulse is applied. Then, the current rapidly decreases to approximately 0.15 A in about 50 μs and remains stable, which is due to the formation of the vapor gaseous skin surrounding the electrode, which has the highest electrical resistance in the circuit. On the other hand, it is noticed that compared to the positive current of ECM, the positive current in PA-ECM takes a longer time to reach its maximum value. This change is because the gaseous vapor skin formed in the negative pulse time needed a certain time to dissipate, influencing the current flow.

It has been proven that the plasma discharge occurring in the negative pulse does not result in material removal [23]. However, it has a significant influence on the subsequent anodic dissolution process. As illustrated in Fig. 12(a), in the positive pulse of ECM, a viscous layer is formed on the working electrode surface by the reaction products,
including oxides, metallic cations, and anions. The thick viscous layer causes a serious concentration polarization and results in a low etching rate in the ECM. In PA-ECM, however, as shown in Fig. 12 (b), clustered plasma discharges occur at the electrode surface in the negative pulse time, which forms a positive outward pressure owing to the implosion behavior of the plasma. In the positive pulse time, however, the gas generation rate drops sharply, which can result in negative inward pressure in the surrounding liquid. The cyclic positive and negative pressure can enhance the mass transport efficiency, update the surrounding electrolyte, and thus suppress the concentration polarization. Furthermore, the plasma discharges in the vapor gaseous skin lead to the rapid heating of the electrolyte surrounding the working electrode, promoting subsequent anodic etching by enhancing the kinetics of the electrochemical reaction. As a result, the material removal rate increased by approximately 20 times compared to that of ECM, as shown in Fig. 8 (a).

To experimentally verify the mechanism discussed above, direct observation of the surface phenomena of the working electrode during machining was conducted utilizing a high-speed video camera (Dimax HS1, PCO). Fig. 13 shows the photos taken at different timings in the ECM and PA-ECM respectively. The formation of a viscous layer on the electrode surface can be clearly seen in the ECM. In PA-ECM, however, no viscous layer but a vigorous generation of bubbles and plasma discharges is observed at the electrode surface. In addition, the bubbles oscillate constantly and show periodic expansion and contraction dynamics due to the pulsating plasma discharges generated at the electrode interface. This behavior can be clearly confirmed at the initial stage of PA-ECM when the bubble volume is less, as shown in Fig. 14.

Fig. 10. Comparison of current waveforms between conventional ECM and PA-ECM: (a) in conventional ECM, pulse-off duration is applied during which the current is zero. (b) in PA-ECM, a negative current with a sharp rise to −3 A is observed right after the negative pulse is applied. The current rapidly decreases to −0.15 A in about 50 μs and remains stable due to the formation of plasma surrounding the electrode.

Fig. 11. Difference in the peak current amplitude between ECM and PA-ECM. The anodic current amplitude of PA-ECM is 1.3 times that of ECM under the same positive voltage owing to the effect of plasma.

Fig. 12. Schematic diagram of the working electrode interface: (a) ECM with a viscous diffusion layer and (b) PA-ECM with pulsating plasma to enhance mass transport.
The oscillation of bubbles caused by the pulsed plasma creates a local hydrodynamic flow field near the electrode surface owing to the constantly changing local pressure, leading to enhanced diffusion and convection at the electrode surface.

5.3. Temperature analysis in PA-ECM

To investigate the thermal influence of plasma, the in-process temperature change of the microrod electrode during machining was directly measured using an infrared thermal camera (T660, FLIR Systems). The area of measurement is the part of the microrod immediately above the liquid interface, as shown in Fig. 15. During the measurement, the positive pulse voltage was set to 0 V to prevent the microrod from dissolving. Fig. 15(b) shows the temperature distribution of the microrod at \( t = 60 \) s. It can be found that the temperature at the bottom of the electrode where it is nearest to the plasma site, reaching 71 °C, is the highest. Fig. 16(a) shows the dynamic change in the microrod temperature at the point near the liquid interface. When the cathode plasma is formed, the microrod rapidly heats up within 2 s and then stabilizes at approximately 71 °C. The heat transfer inside the microrod electrode under steady state can be expressed as:

\[
\frac{\partial^2 T}{\partial z^2} = 0
\]

Based on the temperature distribution data measured with the thermal camera, the microrod surface temperature at the plasma area can be predicted using Eq. (4), which is approximately 73.1 °C, as shown in Fig. 16(b). Although it has been reported in the literature that the plasma temperature is approximately 500 °C or higher [31, 32], the heat conducted to the electrode is much less according to our measurement. Furthermore, as shown in Fig. 17, a polished cross section of the microrod electrode after PA-ECM was examined using SEM, and no recast layer or heat-affected zone was found in the subsurface of the rod. Therefore, it is concluded that the plasma in PA-ECM can heat the electrode but exerts little thermal influence on the working surface. On the other hand, it can be predicted that the thermal energy of the plasma can heat the local electrolyte to a higher temperature and thus enhance the kinetics of electrochemical reaction for faster dissolution.
5.4. Improvement in the machining efficiency and accuracy by applying a higher voltage

By increasing the applied voltage, a higher machining efficiency can be achieved owing to the increased current density. However, in previous studies on electrochemical etching, a high voltage above 10 V was rarely applied [34–36]. This is because a high voltage results in a violent etching process and poor surface finish, which are difficult to control. In this section, the difference in the etching accuracy under a high positive voltage of 30 V between ECM and PA-ECM was investigated.

The pulse trains applied are shown in Fig. 18. The resulting microrod surface by ECM in Fig. 19(a) presented a very poor surface appearance with serious burrs and etching pits, especially at the necking area, demonstrating that the high voltage in ECM was not suitable for making precision microrods. In comparison, as shown in Fig. 19(b), the surface of PA-ECM was much smoother with a lower surface roughness of 78.6 nm. Surprisingly, as shown in Fig. 20, the ECM showed an MRR 2.4 times higher than that of PA-ECM under...
the high voltage condition. The microrod diameter resulting from ECM was 51.6 μm, one third of that of PA-ECM. It is considered that the results are caused by the negative differential resistance (NDR) characteristics of the vapor gaseous skin [21]. That is, the current value decreases with increasing voltage in PA-ECM. This change occurs because the increasing voltage promoted gas evolution and heat generation, which made the vapor gaseous skin thicker, thus decreasing the current. This finding has been confirmed by analyzing the positive current during the process. According to the experimental measurement results, the anodic current in PA-ECM was 0.57 A, less than one-third that of ECM, which is 1.93 A. This result was the reason why PA-ECM presented a lower MRR than ECM under high voltage conditions.

The generation of bubbles and the bubble movement in the ECM and PA-ECM methods are depicted in Fig. 21. The process can be divided into three stages: positive pulse, pulse transition and negative pulse (pulse-off in ECM). For both methods, in the positive pulse stage, the tungsten rod is anodically dissolved, accompanied by O₂ bubble evolution. For the next two stages, however, there is a large difference between ECM and PA-ECM. In the ECM, the generated O₂ bubbles diffuse and eventually dissipate during the pulse transition and pulse-off stage. In PA-ECM, however, many more H₂ bubbles are evolved in the pulse transition and negative pulse time due to the high negative voltage. Together with the O₂ bubbles generated in the following positive pulse, a much thicker gaseous skin is generated, which insulates the working electrode and results in a significant decrease in current density.

On the other hand, the phenomena that the gaseous skin limits and confines the anodic dissolution process can be effectively utilized to improve the ECM accuracy, as the insulation effect of the gaseous skin can suppress unwanted stray machining at the working electrode side surface.

5.5. Influence of the negative pulse on the machining of the microrod

To avoid excessive bubble generation at the working electrode surface, the duty ratio of the positive pulse is varied to control the bubble generation process.
The experimental results are shown in Fig. 22. At a duty ratio of 70%, the resulting microrod by PA-ECM was much thicker than that by ECM. However, when the duty cycle is set at 90%, the diameter of the microrod processed by PA-ECM is thinner than that processed by ECM, and the necking phenomenon is weakened. Specifically, the rod straightness error is drastically reduced from 66.8 μm to 14.6 μm, on the order of 78%. These results demonstrate that PA-ECM can improve both the machining efficiency and machining precision simultaneously. Furthermore, the rod surface integrity of PA-ECM was much better than that of ECM. From Fig. 23, it can be found that with increasing duty ratio, the ECM efficiency decreased, while the efficiency of PA-ECM increased instead. The decrease is because the viscous concentration boundary layer formed in the ECM became much thicker with increasing duty ratio, which intensifies the concentration polarization and results in a lower anodic dissolution efficiency. In PA-ECM, however, the plasma discharges can enhance the mass transport effect, resulting in higher processing efficiency. In addition, a shorter negative pulse time leads to fewer bubbles, thus increasing the current density.

5.6. Optimization of the bipolar pulse train

Based on the above results, the cathodic gas evolution process should be well controlled to enable the occurrence of plasma discharge while avoiding excessive gas evolution. To optimize the process, a pulse-off time is considered necessary in the bipolar pulse train. Therefore, a specially designed bipolar pulse train, as shown in Fig. 24, was proposed. Based on the position of the negative pulse in the pulse-off period, the bipolar pulse train was divided into three types: (a) preposition, (b) middle-position, and (c) postposition. The experiments corresponding to each type were carried out to verify the effects. The other experimental conditions were kept the same, as shown in Table 2. The experimental results are shown in Figs. 25 and 26. The middle-position pulse train type presented the highest material removal rate, followed by the postposition and preposition types. When the negative pulse was postpositioned in the pulse-off time, there was insufficient time for bubble dissipation before the next positive pulse, resulting in a thick gaseous skin, reducing the current density and consequently the machining efficiency. In the preposition type, however, it was difficult to ignite plasma discharges during the negative pulse time because the bubbles generated at the previous positive pulse interfered with the hydrogen evolution and electrolyte vaporization in the negative pulse and consequently affected the formation of the gas skin in a short time. The middle-position type is the optimal bipolar pulse train type because the discharge can be ignited easily and the generated bubbles in either the positive pulse or negative pulse can be effectively dissipated, leading to a high etching efficiency.

Furthermore, the influence of the duty ratio of the positive pulse was investigated. As shown in Fig. 26, with the same pulse period of 1 ms and the same negative pulse-on time of 20 μs, the MRR was higher when the positive pulse time was shorter than 700 μs in comparison to 900 μs. This finding proves the above discussion that a longer pulse-off time can facilitate the dissipation of bubbles and improve machining efficiency. Notably, an optimal machining rate was achieved when the positive pulse time was 700 μs, and the negative pulse was middle-positioned. As a result, a microrod with an average diameter of 18 μm and a high aspect ratio of 55:1 was successfully machined within 5 s, and a maximum diameter reduction rate of 36.4 μm/s was achieved. To our knowledge, this is the highest efficiency of microrod fabrication that can be achieved so far. In addition, the smallest diameter of the microrod was only 6 μm, and the resulting surface roughness of Ra was 46 nm.

To conclude the above results, the optimal bipolar pulse train suitable for PA-ECM of microrod tools should possess the following characteristics: (1) a larger duty ratio of positive pulses to improve material removal, (2) the capability of the generation of plasma discharges to enhance anodic dissolution, (3) a relatively shorter negative pulse time to avoid excessive bubbles, and (4) a certain pulse-off time to improve mass transport. Within the suitable process windows, PA-ECM is beneficial for enhanced machining precision and efficiency.

5.7. PA-ECM for microdrilling and milling

PA-ECM drilling and milling can be realized by feeding the plasma-coated microtool toward the workpiece electrode based on the proposed principle above. Fig. 27 shows the proposed method and experimental conditions. Compared to the pulse waveform of ECM, a short but high amplitude voltage pulse during the pulse-off time (Fig. 27(b)) is applied in PA-ECM to induce a plasma around the microtool electrode. To avoid microtool wear due to reverse current during the machining [42], a diode is connected in both circuits. As the machining gap condition is very different from that of microrod fabrication, the machining characteristics will change. Fig. 28 shows a comparison of blind holes on the tungsten workpiece resulting from ECM and PA-ECM drilling. It can be seen that when the machining voltage $U_1$ is 5 V, PA-ECM and ECM achieve a very close machining precision with the same hole entrance width. The resulted hole depth of PA-ECM, however, is larger than that of ECM, which indicates the effect of plasma for enhancing the machining efficiency. When the machining voltage $U_1$ is increased to 15 V, the PA-ECM results in a smaller machining gap (represented by hole entrance width) than that of ECM, indicating that the induced bubble and plasma are effective for improving the machining precision by the side surface insulation effect under high voltage. On the other hand, the less material removal amount of PA-ECM implies a lower machining efficiency. This result is probably caused by the narrow machining gap and plasma which hinders the electrolyte comes into contact with the machining area, thus resulting in a reduced anodic current. Further optimization of the process is necessary to improve PA-ECM accuracy and efficiency.

Fig. 29 shows several examples of microstructures machined by PA-ECM cutting and milling by controlling the micro tool feed on the workpiece surface. The material used here is stainless steel (SUS304). As in Fig. 29(a), a microrod can be cut by PA-ECM with high dimensional uniformity using a micro tool electrode. In Fig. 29(b) and (c), micro-patterns are successfully machined with a low surface roughness of Ra 23.5 nm, demonstrating the capacity of PA-ECM for micromachining.

<table>
<thead>
<tr>
<th>D=70%</th>
<th>D=90%</th>
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<tbody>
<tr>
<td>(a) ECM</td>
<td>(b) PA-ECM</td>
</tr>
<tr>
<td>(c) ECM</td>
<td>(d) PA-ECM</td>
</tr>
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![Fig. 22. SEM image of microrods resulting from ECM and PA-ECM under different duty ratios of positive pulses. ($U_p$: 30 V, $t$: 5 s).](image)
6. Conclusions

In this study, a novel hybrid plasma-assisted electrochemical machining method combining ECM and an electrolytic plasma is demonstrated for microtool and microstructure fabrication. The process includes enhanced cathodic gas liberation, plasma formation, and plasma-assisted anodic dissolution. To evaluate the proposed method, comprehensive experiments were conducted. Some of the key conclusions of the study are as follows:

1. In machining of microrod, the introduction of a cathodic plasma increased the anodic current by 1.3 times under the same anodic...
potential, while the volumetric removal rate showed an increase of approximately 20 times. The major plasma influence is attributed to the increase in local temperature and the formation of a local hydrodynamic flow induced by the pulsating plasma, which is not only able to enhance the kinetics of the electrochemical reaction by increasing temperature but also improves mass transport in the ECM process, thus leading to enhanced machining efficiency.

2 In PA-ECM, the vapor gaseous skin formed around the working electrode shows an insulating effect, which can significantly improve the ECM precision, especially under high voltages. In PA-ECM of microrod, the machining precision (expressed by microrod straightness error reduction) improved by 78% at high anodic potentials of 30 V.

3 The working electrode surface temperature at the plasma site was very low (73.1 °C in microrod machining), and no discharge crater or other thermal damage resulting from the plasma was observed, indicating that the material removal is primarily determined by anodic dissolution and plasma-chemical reactions.

4 The PA-ECM performance is significantly influenced by the formation and diffusion behavior of the induced gaseous bubbles and
plasma, which can be controlled by designing the pulse voltage waveform. By inducing a cathodic plasma during the anodic pulse interval, followed by a pulse-off time, anodic dissolution can be remarkably enhanced, and PA-ECM performance can be maximized. As a result, a maximum diameter reduction rate of 36.4 μm/s was achieved for machining a microrod tool with a high aspect ratio of 55:1.

5 PA-ECM drilling and milling of microstructures are successfully performed, and the results show that PA-ECM drilling also provides improved machining precision as compared to ECM owing to plasma insulating effect. However, unlike machining of microscopical narrow machining gap in PA-ECM drilling/milling gives rise to difficulty of electrolyte flow, thus resulting in a reduced machining rate under high voltages.

Overall, this study has shown that PA-ECM has the potential to enhance the conventional ECM performance. With suitable process windows, improvement in both machining precision and efficiency can be realized.

Credit author statement

Shunda Zhan: Conceptualization, Data curation, Investigation, Validation, Visualization, Roles/Writing - original draft. Yonghua Zhao: Formal analysis, Funding acquisition, Methodology, Project administration, Supervision, Resources, Writing - reviewing & editing.

Declaration of competing interest

We declare that we have no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (NSFC) (grant number 51905255), the Shenzhen Knowledge Innovation Plan (grant number JCYJ20180504165815601), the Shenzhen High-level Innovation and Entrepreneurship Fund (grant number KQTD20170810110250357), and the Shenzhen Science and Technology Innovation Commission (grant number JCYJ20190809143217193).

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