

## Technical Paper

# Numerical modeling of passive layer formation and stabilization in electrochemical polishing process



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## ABSTRACT

The electrochemical polishing generates surface finish in micro-/nano-scale. The process, primarily works on the principle of preferential anodic dissolution owing to the passive layer formation. The current–voltage characteristics curve of the process shows that stable passive layer by anodic dissolution is obtained in plateau region that is a favorable condition for the electrochemical polishing process. This work presents numerical simulations of effect of main process parameters: inlet velocity, diffusion coefficient of electrolyte, inter-electrode gap and roughness of anode surface on uniformity and time to obtain steady passive layer. It is observed that all the above parameters govern uniformity of the passive layer. The inlet velocity and diffusion coefficient of electrolyte mainly govern the time required for stabilization of the passive layer followed by the inter-electrode gap; however, surface roughness at anode has a negligible effect. Further, the process gives better performance when the inlet velocity is 0.007–0.008 m/s, diffusion coefficient is  $10^{-8}$  to  $10^{-9}$  m<sup>2</sup>/s and inter-electrode gap is greater than 6 mm.

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## 1. Introduction

The roughness of a surface plays an important role in reducing friction, surface bearing capacity, resistance against corrosion and overall life-cycle improvement [1]. In the case of precision components, the surface finish requirements are very stringent. In such situations, the processes, which can give the desired surface finish without any adverse effects, are more desirable. The electrochemical polishing process fulfills these requirements, as it is a non-contact process and is independent of mechanical properties of work material. Consequently, it is widely employed in industries for polishing of common materials such as stainless steel and some of the “difficult-to-finish” materials such as biomedical titanium, tantalum, etc. [2,3]. It is also used for the removal of burrs formed during machining processes [4]. See Tailor et al. [4] for an up to date review on the various electrochemical finishing processes.

In the electrochemical polishing process, the workpiece and tool are arranged as an anode and cathode in the electrolyte. The process primarily works on the principle of preferential anodic dissolution owing to the passive layer formation. The passive layer is a non-conductive metal oxide layer, which is formed on the work (anode) surface. This passive layer becomes stable and continuous over the entire work surface in an electrochemical polishing

process, when the current remains constant. On a typical current versus voltage ( $I$ – $V$ ) plot, this region of thin layer formation is called as plateau region [5–8]. It is possible that some of the higher peaks on work surfaces protrude out of this protective passive layer during electrochemical polishing. These peaks of work surface get dissolved because at these peaks, the current density is higher, thereby smoothing the work surface. The passive layer has a high specific gravity and is more viscous than the electrolyte; therefore, it is also called as a viscous layer in electrochemical polishing process. The removal of passive layer also relies on the area of work surface shared by the fluid and abrasives [9–12]. Apart from this, the removal of passive layer depends on other parameters such as flow of electrolyte, concentration and temperature of electrolyte [13,14], inter-electrode gap [2] and relative motion between electrodes [10,15,16]. The anodic dissolution rate is governed by two different mass transport mechanisms: diffusion of dissolved species at the cathode and the acceptor diffusion near the anode [17]. In addition, dissolution also depends on the initial peak to valley distance on a work surface, usually characterized by the surface roughness. It is noteworthy that electrochemical polishing process involves electrical, chemical as well as mechanical action making the process relatively complex and difficult to control and model. Nonetheless, researchers have worked on modeling and simulation of electrochemical process and also employed different experimental techniques. Some of the important works are summarized below.

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### Nomenclatures

$c$	concentration ( $\text{mol}/\text{m}^3$ )
$c_i$	concentration of species $i$ ( $\text{mol}/\text{m}^3$ )
$D$	diffusion coefficient of metal hydroxide ( $\text{m}^2/\text{s}$ )
$D_i$	diffusion coefficient of species $i$ ( $\text{m}^2/\text{s}$ )
$\bar{e}$	electron
$F$	body force ( $\text{N}/\text{m}^3$ )
$M$	metal ion
$M(\text{OH})_n$	metal hydroxide
$M^{+n}$	Metal ion with $+n$ charge
$N_0$	inward flux ( $\text{mol}/(\text{m}^2 \text{ s})$ )
$n$	valency
$\mathbf{n}$	unit normal vector
$\text{OH}^-$	hydroxyl ion
$p$	pressure (Pa)
$p_0$	atmospheric pressure (Pa)
$R_i$	reaction rate for species $i$ ( $\text{W}/\text{m}^3$ )
$T$	transpose
$\mathbf{u}$	velocity vector (m/s)
$u_0$	inlet velocity (m/s)

### Greek symbols

$\eta$	dynamic viscosity (Pa s)
$\rho$	density of electrolyte ( $\text{kg}/\text{m}^3$ )

Jemmely et al. [18] developed two theoretical electrochemical models for passivation in tribocorrosion assuming that passivation occurs on a bare work surface laterally or vertically. In their model, the film growth kinetics and ohmic drop in the electrolyte have been considered. However, electrochemical conditions in the contact zone of the electrodes, the role of third body particles and selective dissolution during the film growth are ignored. Their simulation shows that vertical film growth model should be used after applying the lateral film growth model for a more reliable prediction. Ma et al. [19] suggested a mathematical model to predict the thickness of bulk material removal and reduction of assessed profile height, as a function of inter-electrode gap, finishing time and applied voltage. Kozak et al. [20,21] developed a mathematical model for pulse electrochemical machining (PECM) which predicts the minimum inter-electrode gap so that boiling of electrolyte can be avoided. The model thus developed was employed to simulate PECM as well as to determine the thermal limitations and energy consumption. Kozak [22] developed a mathematical model, which is useful in the simulation of electrochemical process. Marshall and Wolff [23] considered linear secondary current distribution between electrodes and electrolyte in a rectangular electrochemical cell using an analytical solution of the Laplace equation. Hardisty et al. [24,25] performed a numerical simulation of electrochemical machining for a flat pair of electrodes as well as for parabolic tool and stepped work surface. They developed a two-dimensional model that predicts the erosion of work surface by potential and flux distribution in the electrolyte. Tang and Gan [26] demonstrated the electrolyte flow simulation model for electrochemical process and found that electrolyte flow mode directly affects the surface quality in case of complex parts. Nelissen et al. [27] modeled and simulated multi-ion transport phenomenon between parallel plates that shows the effect of convection, diffusion and migration of ions on concentration as well as current distribution. They found that mass transport has a dominant effect on the local concentration distribution as compared to the current density. Dan et al. [28] simulated one-dimensional transient current response considering the rotating electrode for electrochemical process. Their model shows that current density versus overpotential, distribution and

concentration of active ions depends on the rotational speed of the electrode. The electrochemical impedances for various cases were also derived from the developed model.

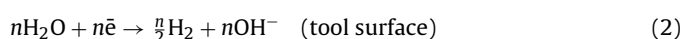
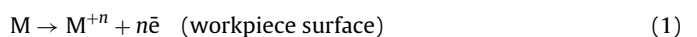
The above studies show that the simulation of electrochemical processes is limited to a fundamental phenomenon of electrolysis, workpiece profile, current or potential distribution, fluid flow analysis, and transportation of ions and their positions. However, formation and stabilization of the passive film/viscous layer, which determines the progress and success of the electrochemical processes has not been adequately studied. The passive layer formation would involve mass and momentum transportation phenomena such as convective and diffusion of various species that are formed during the electrochemical polishing processes. Therefore, this paper presents simulation of formation of passive layer in electrochemical polishing process to understand the effect of mass and momentum transport on the mechanism of the passive layer formation and on the time required for its stabilization as a function of various processing parameters.

## 2. Mathematical model formulation

### 2.1. Problem definition

To simulate the passivation process involving the passive layer formation in electrochemical polishing process, two-dimensional geometry has been prepared as shown in Fig. 1. In this, the workpiece and the tool (electrodes) are considered to be rectangular in shape of size  $60 \text{ mm} \times 1 \text{ mm}$ , enclosed in an electrolyte reservoir of size  $150 \text{ mm} \times 150 \text{ mm}$ . The surface roughness of workpiece (anode) facing the cathode is considered as general variation in overall profile height characteristics. This has been employed by two-dimensional concept average roughness ( $R_a$ ), as the cell geometry is also two-dimensional. The profile skewness (asymmetries) is affected or playing role in surface improvement in electrochemical finishing process when the charge ions follow a cycloidal and suffer from agitation due to additional external source of energy [29–31]. In the present case, there is no any such additional source of energy for changing the straight path of charge ions. Therefore, the profile of surface roughness is made with no skewness and employed as a series of isosceles triangles as shown in Fig. 1(b). The initial Kurtosis (peakedness or sharpness) on peak or in valleys becomes blunt where the passivation occurs first and later behavior depends on the processing conditions [32]. To account for this, corners of peaks and valleys are made sharp by point at which two edges of triangle are met. The roughness of the workpiece surface was varied between 0 and  $200 \mu\text{m}$ . Hereafter, this surface with roughness is addressed by a single straight-line boundary # 8 (Fig. 1) for diagrammatical ease. The geometry of the electrochemical system considered here is same as that of Lee et al. [33] so that benchmarking of the results can be done. It is assumed that the electrolyte enters the reservoir from the bottom and leaves from the top (see Fig. 1).

It is understood from the principle of electrochemical polishing that on the work surface, metal is dissolved releasing electrons ( $M^{+n}$ ) (Eq. (1)). By gaining electrons on the tool surface (cathode), hydrogen ( $\text{H}_2$ ) and hydroxyl ions ( $\text{OH}^-$ ) are produced (Eq. (2)). The hydroxyl ions combine with the dissolved metal ions near the work surface and precipitates metal hydroxides ( $M(\text{OH})_n$ ), which is passive (non-conductive) in nature (Eq. (3)). Therefore, the net reaction of electrochemical polishing process can be presented by Eq. (4)



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