

# Transient Simulation of the Removal Process in Plasma Electrolytic Polishing of Stainless Steel

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## Introduction

Plasma electrolytic polishing (PeP) is an electrochemical method for surface treatment. In detail PeP is a special case of anodic dissolution [1] that unlike electrochemical polishing requires higher voltage and uses environment friendly aqueous solutions of salts.

In recent years, a lot of studies have been made. Nevertheless, at present, a few research works have been focused on the understanding of the process and even less on simulation. Due to the fact that PeP is a complex combination of chemical and physical processes, it is challenging to simulate this process.

To investigate the basics of PeP a transient 2D simulation model was developed. In this model, a special interest is focused on the plasma-gas layer and the electric potential. The thickness of the plasma-gas layer and its conductivity are based on experimental data [2 – 4]. Material removal is realised as a function of the current density at the workpiece surface.

The paper shows that the main voltage drop in PeP occurs in the plasma-gas layer and that primarily the profile of the surface determines the distribution of current density. Both effects have a main significance in the polishing process. Furthermore, the polishing effect on the surface profile will be analysed.

## Theory

In the literature there is a lot of information on solutions used for polishing of different metal alloys like steels, aluminium, titanium and others [2; 5 – 11] and on process parameters like temperature, electrolyte concentration and voltage. For example, 3% - 6% ammonium sulphate solutions are widely used for polishing stainless steel workpieces with common voltage and temperature range 250 V – 350 V and 70 °C - 90 °C respectively [2, 9, 11]. But as mentioned above, only few research works have been focused on the simulation [12, 13].

A principle scheme of the PeP process is shown in Figure 1. The workpiece is anode and connected to a DC energy source. Due to high voltage the formation of the plasma-gas layer on the anode occurs. The polishing process requires the presence of this plasma-gas layer. The plasma-gas layer is stable in a range from 200 V to 400 V [2, 5, 7].

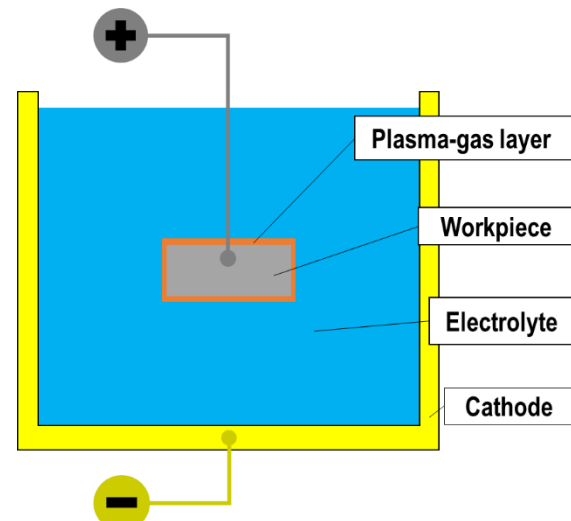


Figure 1. Principle scheme of PeP

However, PeP has some limitations. Firstly, mainly metal parts can be polished. There is few information, if it is possible to polish semiconductors. Secondly, process energy source determines the maximum part size. For example, around 5 kW electrical power is required to polish 40 cm<sup>2</sup> workpiece surface. Thirdly, each metal requires electrolyte adaption. For example, titanium can't be polished in ammonium sulphate which is used for stainless steels. Lastly, treating internal cavities is challenging.

Figure 2 shows schematically a typical current-voltage characteristic. The first section AB is a conventional electrolysis process that can be described by classical electrochemistry.

The section BC is a transient or switching mode, when a plasma-gas layer periodically occurs on the anode. This leads to unstable current with many drops.

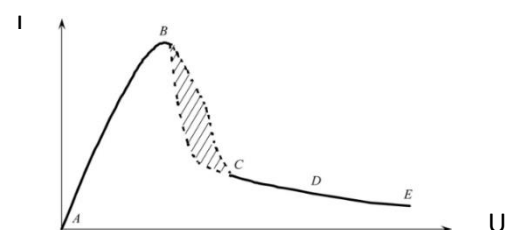


Figure 2. Schematic current-voltage characteristic [2]

The section CD is an electrolytic plasma mode [2] when plasma-gas layer is stable and polishing is possible.

At the section DE the plasma-gas layer becomes unstable. Voltages above 400 V cause disruption of

the plasma-gas layer and stop the polishing process.

At the sections BC, CD and DE increase in the voltage leads to the decrease in current, because of increase in thickness of plasma-gas layer [2, 3, 12]. At the same time, based on data from literature sources, it can be derived that this layer has significant resistance.

PeP is a technology that is used as a finishing surface treatment of metal workpieces. After processing, the surface of a workpiece is smoother and has higher gloss level. Because of small achievable roughness ( $R_a < 0.02 \mu\text{m}$ ) and small removal rates [1 – 3], this process can be applied for finishing of precision parts.

Although this technology is known since the 1970s, the processes taking place in the plasma-gas layer are not fully described.

### Model development

The developed model is used to simulate electrical phenomena and removal process during PeP after the appearance of a stable plasma-gas layer.

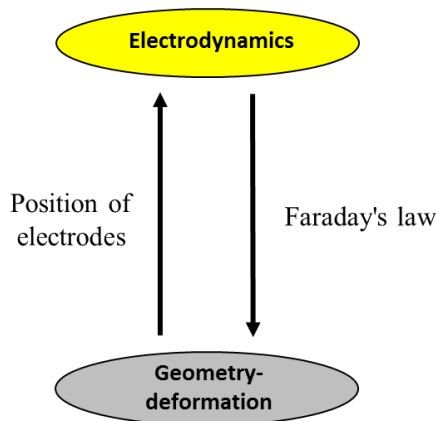


Figure 3. Coupling scheme of the multiphysical model

The model is based on the assumption that PeP can be considered as an electrochemical polishing. The coupling scheme is provided on Figure 3.

The model set up and calculation were made in COMSOL Multiphysics ®. Electric Currents and Deformed Geometry interfaces were chosen for this model. The initial anode surface profile was generated in COMSOL Multiphysics® using Spatial Frequencies method [15] with the equation:

$$y = A \sum_{m=-N}^N (m^2)^{\frac{-b}{2}} g1(m) \cos(2\pi ms + u1(m)) \quad (1)$$

This was made to simulate the polishing effect of PeP and to analyse the current density distribution on a surface. Parameters that were used for this are provided in table 1.

Table 1: Parameters for Spatial Frequencies method

Parameter	Description	Value
N	Spatial frequency resolution	30
b	Spectral exponent	0.2
A	Scale parameter in y coordinate	0.0005
s	x coordinate	
g1	Gaussian random function	
u1	Uniform random function	

Modell geometry and boundary conditions are based on principle scheme shown in Figure 1 and provided in Figure 4. The bath with the electrolyte is defined as cathode. The bath has dimensions of 20 cm x 20 cm. The anode is completely immersed in the bath to a depth of 5 cm. The anode is surrounded by a plasma-gas layer.

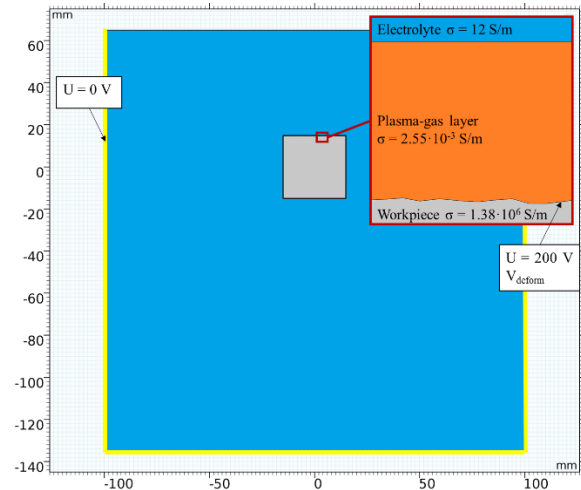


Figure 4. Modell geometry and boundary & domain conditions

Table 2: Simulation parameters

Parameter	Value
Voltage	200 V
Anode conductivity	$1.38 \cdot 10^7 \text{ mS/cm}$
Electrolyte conductivity	120 mS/cm
Plasma-gas layer conductivity	$2.55 \cdot 10^{-2} \text{ mS/cm}$
Plasma-gas layer thickness	0.15 mm
Removal coefficient K	$1.54 \cdot 10^{-11} \text{ m}^3/(\text{A} \cdot \text{s})$
Anode relative permittivity	1
Electrolyte relative permittivity	55
Plasma-gas layer relative permittivity	1

The model has 3 domains: electrolyte, plasma-gas and anode. Simulation parameters are provided in table 2.

Side and bottom boundaries of the model are defined as grounded. A voltage of 200 V is applied to the workpiece boundaries.

Ammonium sulphate was chosen as an electrolyte for this simulation. Electrical conductivity was set 120 mS/cm, what corresponds to concentration of 50 g/l solution at 75 °C [14]. This value is common for polishing stainless steels. Steel 304 was chosen as material for the anode.

The thickness of plasma-gas layer based on the literature was chosen 150 µm [2 – 4]. Electrical conductivity of the plasma-gas layer was calculated based on experimental values and data provided in literature. In extended literature common value of electrical field are provided: 10<sup>4</sup> V/cm - 10<sup>5</sup> V/cm [2 – 4, 14]. Such a high value allows to assume, that almost all voltage drops in the plasma-gas layer. Than based on the thickness of 150 µm and voltage of 200 V electrical field can be calculated:

$$E = \frac{V}{dh} = \frac{200 V}{0.015 cm} = 13333 V/cm \quad (2)$$

This corresponds with the above mentioned range. Current density is defined:  $j_n = \sigma \cdot E$ . Average  $j_n$  based on experimental data from Rajput et al. [14] for 200 V was 0,3399 A/cm<sup>2</sup>. Knowing  $E$  and  $j_n$ , conductivity can be calculated:

$$\sigma = \frac{j_n}{E} = \frac{0.3399 A/cm^2}{13333 V/cm} = 2.55 \cdot 10^{-2} mS/cm \quad (3)$$

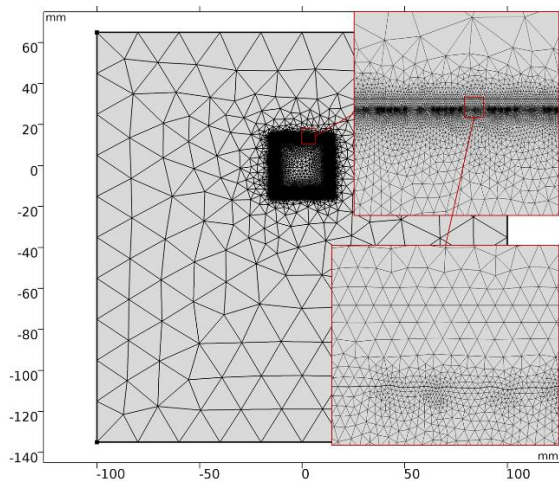


Figure 5. Mesh

In Figure 5 a visualization of the model mesh is provided. Complete mesh consists of 344424 domain elements and 13818 boundary elements.

Mesh parameters are provided in table 3. The finest mesh is realised near the anode surface, where the removal take place.

Table 3: parameters for mesh

Parameter	Electrolyte and plasma-gas layer	Workpiece
Maximum element size	20 mm	20 mm
Minimum element size	0.005 mm	0.005 mm
Maximum element growth rate	1.5	1.2
Curvature factor	0.2	0.2
Resolution of narrow regions	1	1

The simulation has two studies: stationary study, where initial values for electrical variables are calculated and time depended study, where electric currents physics and mesh deformation are solved. Mesh deformation is calculated according to equation below:

$$V_{deform} = K \cdot (-j_n) \quad (4)$$

where:

$K$  - removal coefficient calculated based on experimental data from Rajput et al. [14];

$j_n$  - normal current density.

$K$  is calculated from experimental data: average material removal rate (MRR) and average current density for 200 V:

$$K = \frac{MRR}{j_n} = \frac{5.24 \cdot 10^{-8} m/s}{3398.69 A/m^2} = 1.54 \cdot 10^{-11} m^3/(A \cdot s) \quad (5)$$

Removal simulation was made for 120 s.

## Simulation Results

In Figure 6 can be seen the result of electrical potential. As expected, the main voltage drop occurs in in the plasma-gas layer. That can be seen in Figure 7 and Figure 8. Figure 7 and Figure 8 show the electrical potential near the workpiece surface at different time steps.

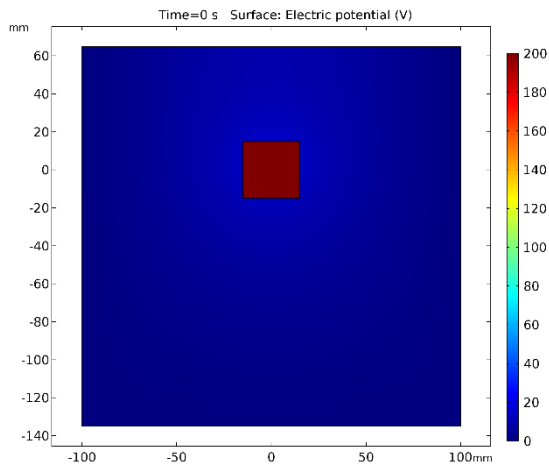


Figure 6. Electric potential

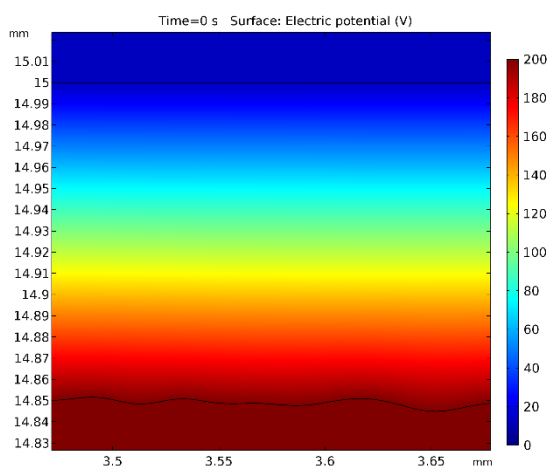


Figure 7. Workpiece surface and electric potential at t=0 s

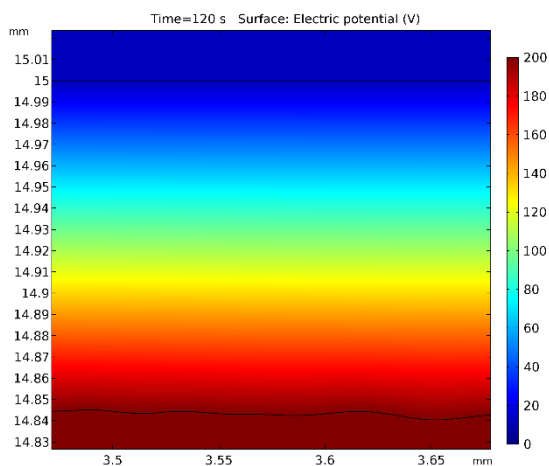


Figure 8. Workpiece surface and electric potential at t=120 s

Because almost total voltage drops inside it, plasma-gas layer can be considered as a special electrochemical cell, where the interface between plasma-gas layer and electrolyte acts as a cathode.

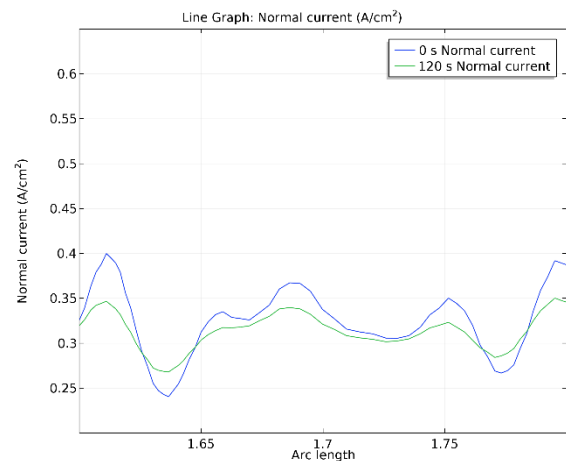


Figure 9. Normal current density at 0 s and 120 s

In Figure 9 can be seen, that the normal current density in the cavities is lower than at the peaks. The normal current density is mainly influenced by the shape of the surface. Taking into account the electrochemical character of the process, this leads to a faster removal of the material on the peaks.

In Figure 9 it also can be seen that at the current density at the deeper cavities raises with the processing time. Average current density in model is  $0.313 \text{ A/cm}^2$  comparing to  $0.340 \text{ A/cm}^2$  in experiment from Rajput [14].

Figure 10 shows the surface profile before and after 120 s polishing. It can be seen, that despite the fact that the overall shape of the surface is saved, the peaks were visibly removed. That can be explained by the higher current density on the peaks.

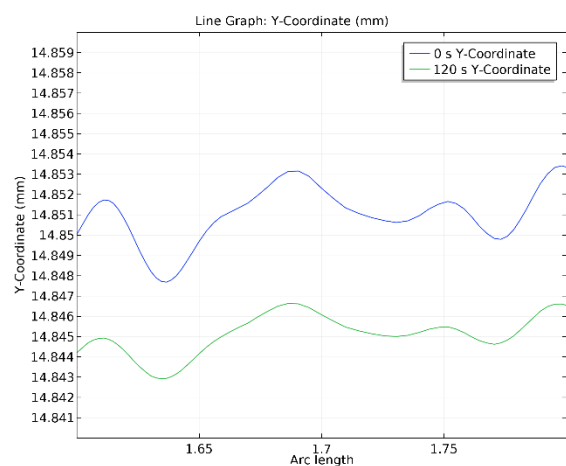


Figure 10. Surface profile at 0 s and 120 s

Figure 11 shows the average heights as function of time. MRR in this model is  $3 \mu\text{m}/\text{min}$  or  $5 \cdot 10^{-8} \text{ m/s}$ . In the experiment it was  $5.24 \cdot 10^{-8} \text{ m/s}$ .

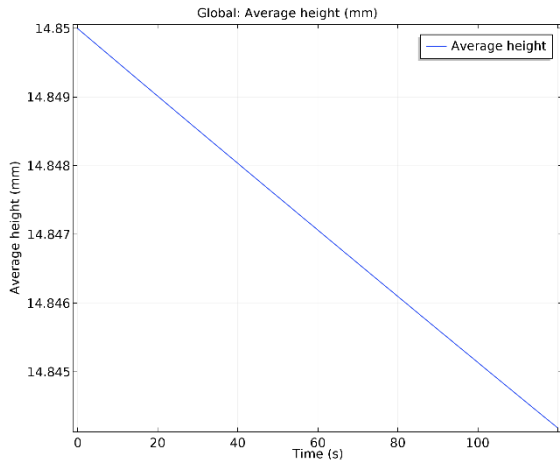


Figure 11. Average height during polishing

To analyse the polishing effect, the roughness parameter Ra was calculated. The equation for Ra calculation was developed based on the next formula:

$$Ra = \frac{1}{l} \int_0^l |h(x)| dx \quad (6)$$

where:

$l$  - evaluation length

$h(x)$  – deviations from the mean line

$$h(x) = |y - \bar{y}| \quad (7)$$

To calculate this in COMSOL, next component couplings were used: intop1 - integration over a boundary 19; p10 and p12 - maximum functions in points 10 and 12 respectively; aveop1 – average over a boundary 19. Points and boundary can be seen in Figure 12.

To calculate  $l$  next equation was used:

$$l = p12(x) - p10(x) \quad (8)$$

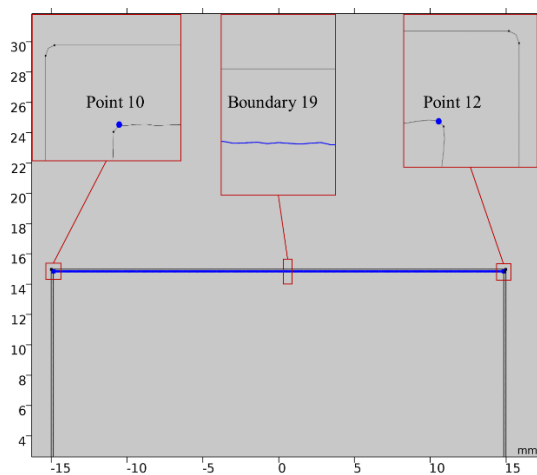


Figure 12. Couplings boundary and points.

Then mean line is calculated with aveop1 function.

Applying everything to the equation (6):

$$Ra = \frac{1}{(p12(x) - p10(x))} \int_{p10}^{p12} |y - \bar{y}| dx \quad (9)$$

Results of this calculation can be seen in Figure 13. It can be seen the roughness decreases according to exponential decay. This corresponds to experiments and data from the literature [4].

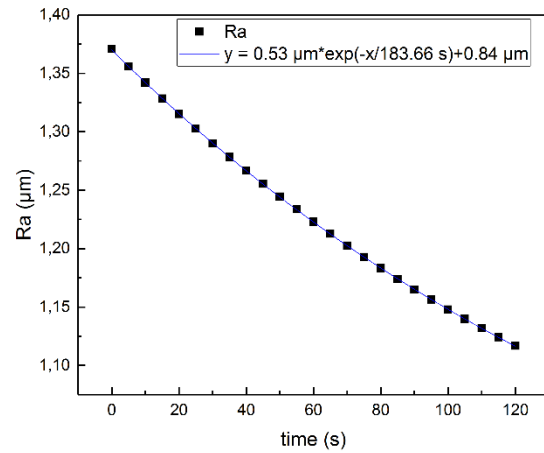


Figure 13. Selected results for Ra as function of time with fit curve

Mukaeva[4] has shown in her work that roughness Ra can be approximated by the following parametric dependence on time  $t$ :

$$Ra = A \cdot \exp((-t) / \tau) + C \quad (10)$$

where:

$A$  – max decrease in roughness;

$\tau$  – time constant;

$C$  – min achievable roughness;

$t$  – processing time;

These parameters were calculated for the model and provided in the table 4.

Table 4: Ra approximation parameters

Parameter	Value
C	$(0.84133 \pm 0.00128) \mu\text{m}$
A	$(0.52878 \pm 0.00123) \mu\text{m}$
$\tau$	$(183.66349 \pm 0.60566) \text{ s}$
Reduced Chi-Sqr	5.04874E-8
R-Square(COD)	0.99999
Adj. R-Square	0.99999

According to this data, the minimal achievable roughness Ra in this model has a value equals  $0.84 \mu\text{m}$ .

## Conclusions and Outlook

It was shown by help of simulation, that the main voltage drop in PeP occurs in the plasma-gas layer and that the surface form determines the distribution of current density. This plays an important role in the polishing process. Current density on the peaks is higher than in cavities. Because of electrochemical character of the process, that leads to faster removal of the peaks. This results in roughness reduction, which was shown and calculated in this paper. The results of Ra calculation from the model approve the exponential roughness decay which was shown by

Mukaeva [4].

Based on this model it can be concluded, that PeP of stainless steel can be simulated as an electrochemical machining process taking part inside the plasma-gas layer.

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