



Contents lists available at ScienceDirect

Surface & Coatings Technology

journal homepage: www.elsevier.com/locate/surfcoat

Electric field effect on surface layer removal during electrolytic plasma polishing

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ARTICLE INFO

Article history:

Received 24 May 2016

Revised 18 July 2016

Accepted in revised form 22 August 2016

Available online xxxx

Keywords:

Electrolytic plasma polishing

Stainless steel

Electric field simulation

Vapour gaseous envelope

3D scanning

Treatment uniformity

ABSTRACT

In this paper, electric field distribution in the electrolyser during electrolytic plasma polishing (EPPo) is analysed. The analysis takes into account field distribution in the electrolyte and the voltage drop in the vapour gaseous envelope (VGE), providing strong bridging to the surface properties using the results of 3D scanning. A numerical approach is used for simulation of the field in the electrolyte which is treated as a linear conductive medium, taking into account a non-linear voltage drop in the thin vapour gaseous envelope formed around the anode. The resultant current density distribution from the electrolyte can be used for evaluation of material removal profile via Faraday's law and current efficiency. The results of 3D scanning show a good agreement with the theoretical results. The average thickness of the surface layer removed after 15 min of EPPo treatment reaches 20–40 μm , with the surface roughness R_a decreasing from 0.3–0.5 to 0.06–0.08 μm , providing a mirror-like surface finish. The removed layer profile change around the sample cross-sectional perimeter exhibits high peaks of the volumetric material loss at the edges, which is consistent with the theoretical profile. The study reveals several important features of the EPPo process mechanism. Firstly, the mechanism is predominantly electrochemical with an estimate of the current efficiency at about 30%. The VGE provides surface oxide removal by hydrodynamic flows and shifts the anodic reaction balance from water electrolysis to the metal dissolution. Secondly, despite presence of plasma in the VGE, the discharge does not cause damage to the surface, due to its diffused type and low intensity. Thirdly, the VGE provides a uniform treatment, especially at higher voltages, because the negative differential resistance of the VGE balances out the current density distribution over a complex shape of the sample, leading to a uniform removal of the surface layer. However, this works only for the surface features of size larger than the VGE thickness ($>3\text{--}5\text{ mm}$); otherwise, the feature becomes exposed to the electrolyte without VGE shielding and is rapidly dissolved because of the inrush of the current density. Finally, the proposed approach contributes to understanding of the mechanisms underlying electrolytic plasma processing and provides a reliable tool for modelling these non-linear processes.

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1. Introduction

Electrolytic plasma processes (EPP) cover a wide range of applications in surface engineering [1]. These include oxidising treatments, primarily plasma electrolytic oxidation (PEO), and non-oxidising treatments, e.g. case hardening and cleaning [2,3]. A distinct feature of the non-oxidising EPPs is a thin vapour gaseous envelope (VGE) formed around the working electrode when a high voltage, leading to intensive ohmic heating of the electrolyte, is applied [4]. This results in either a rapid heating of the working electrode, which is used for nitriding and/or carburising (PEN/PEC), or intense surface cleaning used for polishing and/or coating removal (EPPo/EPCS) [5–7]. In this study, we focus on the anodic EPPo of a stainless steel.

The vapour gaseous envelope has the highest electrical resistance in the circuit; therefore, the majority of the voltage drop occurs across it. The VGE is a quasi-stationary object, since it exists only during the electrolytic plasma process. Moreover, it is also a non-linear object; this follows from the EPP current-voltage characteristics (CVC) exhibiting a negative differential resistance (NDR) region corresponding to the operational regime [1]. This occurs because growing voltage increases the specific amount of heat liberated in the vicinity of the working electrode, making the VGE thicker, so decreasing the current. Another source of non-linear NDR behaviour is a glow discharge which appears in the VGE due to the high electric field. Depending on the EPP type, the VGE thickness varies in the range from 0.01 to 5 mm, resulting in the values of electric field of up to $10^6\text{ V}\cdot\text{cm}^{-1}$, which is close to the breakdown values in vapour-gaseous media [1,8,9]. Although the importance of VGE is generally recognised, limited theoretical studies exist dating back to the 1980s [10,11].

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Recent trends in the EPP research show increasing interest to the assessment of the treatment uniformity [5]. This includes electric field analysis for the electrolyte as a conductive medium. Theoretical studies of electric fields in both the electrolyte and the VGE for a cylindrical coaxial system was carried out in [8] and [12] for PEN/PEC and EPPo processes, respectively. This is a 1D field problem, and more complex shapes have not so far been considered. The voltage drop in the electrolyte is often neglected, with that the voltage across the VGE assumed to be equal to the total applied voltage, and the current density at the working electrode being averaged and considered constant [4,13]. While the former is fair for the majority of EPPs with a VGE, the latter holds only when using the simplest electrode shapes and layouts.

Electric field modelling is routinely used for the analysis of electrochemical processes, simulation and structural optimisation of electrolysers, especially in Al reduction, fuel cells and batteries, anodic coatings and other applications [14–16]. The nonlinearity of EPPs could be a reason why the effect of electric field has not so far been deeply explored for this group of processes.

Therefore, the aim of this study is to investigate electric field distributions in the electrolyte and the vapour gaseous envelope, and correlate associated results with surface characteristics obtained after electrolytic plasma polishing of stainless steel components.

2. Theoretical

2.1. General approach

This research attempts to join the results of theoretical and experimental studies of EPPo treatment of a stainless steel. Firstly, a 2D analysis of longitudinally invariant electric field distribution is carried out. Consequently, a current density distribution along the sample perimeter is obtained and translated into the weight loss and surface profile change. Secondly, an experimental study of the EPPo with the analysed layout is performed at different voltages corresponding to the treatment range boundaries and providing similar average current densities. Further, changes in the actual surface profile of the sample are assessed using a 3D scanner and compared to the theoretical estimates, to reveal the VGE role in the material removal.

2.2. Electric field modelling

2.2.1. Assumptions

The following assumptions can be adopted:

- 1) the problem is solved for the case of a bubble boiling in the VGE during anodic EPPo under potentiostatic conditions;
- 2) the electrolyte is agitated; therefore, its temperature T is independent on spatial coordinates;
- 3) the electrolyte is considered as a linear homogeneous conductive medium with constant specific conductivity γ ;
- 4) the plasma discharge is distributed uniformly over the VGE, with no filamentation or micro-discharging occurring;
- 5) the system nonlinearity is determined by the resistance of the VGE;
- 6) the VGE thickness changes in the range of 0.1 to 2.0 mm, which is significantly less than the interelectrode distance (100...200 mm);
- 7) the problem is solved for a longitudinally invariant field which corresponds to a common case of processing long workpieces.

The above assumptions allow the electrolyte temperature to be used as a constant, depending on the CVC of the process; therefore, instead of a multiphysics problem of the heat- and mass transfer in the electromagnetic field, a stationary problem of a current density distribution could be solved at this stage.

2.2.2. Boundary value problem

The 2D field distribution in the conductive medium (electrolyte) is obtained by numerically solving Laplace equation $\nabla^2\varphi=0$ in Cartesian

coordinates (x, y, z) with respect to the electric potential φ [17]. Based on assumption (7), the 2D problem (Fig. 1) is solved for the longitudinally invariant field $\varphi = \varphi(x, y)$. A rectangular plate anode is placed in the centre of the system at position $(x_1; y_1)$ and the cathode with size of $x_3 \times y_3$ forms the perimeter of the system. Dirichlet boundary conditions, $\varphi = 0$ and $\varphi = U$, are adopted at the cathode and anode respectively. Therefore the non-uniformity of current density can be assessed along the perimeter of the workpiece cross-section parallel to the xOy plane.

The Laplace equation adapted to this case

$$\frac{\partial^2\varphi}{\partial x^2} + \frac{\partial^2\varphi}{\partial y^2} = 0$$

can be solved using finite difference or finite element method for a mesh covering the area of interest so that for each node with coordinates (i, j) of the mesh the potential $\varphi_{i,j}$ is calculated. This solution can be achieved using any electric field modelling software, e.g. COMSOL, ElCut and others.

2.2.3. System non-linearity

The system non-linearity is formalised in an integral form as a non-linear CVC obtained experimentally for the average anode current density:

$$\delta = f(U, T)$$

This determines the current I :

$$I = \delta \cdot s,$$

where s is the surface area of the anode.

2.2.4. Modelling approach

The modelling approach takes into account the electric field in the electrolyte and the VGE, and consists of the following three steps.

The first step is dedicated to the solution of the electric field problem with respect to the distribution of potential φ in the electrolyte. This solution corresponds to a given value of voltage U without taking into account the voltage drop over the VGE. The result is a matrix $m \times n$ of potentials $\varphi_{i,j}$. The resulting potential distribution helps obtaining the electric field \vec{E} using a gradient operator:

$$\vec{E} = -\nabla\varphi$$

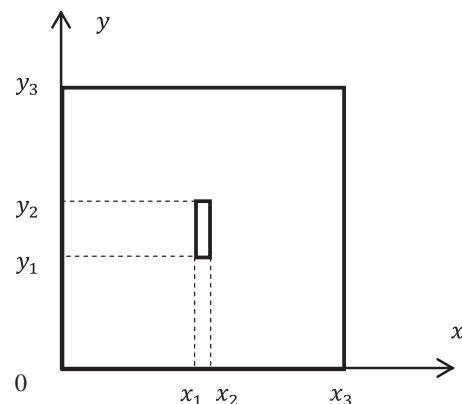


Fig. 1. Electrode layout for the electric field analysis.

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