Article

Impact of the Samples' Surface State on the Glow Discharge Stability in the Metals' Treatment and Welding Processes

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Abstract: The low temperature plasma of glow discharge has found a widespread use as a heating source in welding and surface treatment of metals. The meticulous analysis of glow discharge's instabilities in these processes allowed us to highlight the physicochemical characteristics of the cathode surface (the welded or treated samples) as one of the main reasons of its transition into an electric arc—as a more stable form of gas discharges. The prolonged arc action on the samples surfaces inevitably leads to the disruption of the technological process and, consequently, to undesirable overheating of samples. In this regard, the main aim of this work is to study the influence of the macro- and micro relief of the cathode on the stable glow discharge existence in the processes of metals treatment and diffusion welding. It has been analytically established and experimentally supported that the glow discharge's stability is mainly affected by the sharp protrusions generated on the cathode surface because of samples pre-treatment by machining before welding. It has been established that the rough surface pre-treatment with the Rz about 60–80 µm decreases the pressure range of glow discharge sustainable existence from 1.33–13.3 kPa to 1.33–5.3 kPa compared with the surface machining with the Rz about 10 µm.

Keywords: diffusion welding; plasma; glow discharge; surface treatment; plasma techniques

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1. Introduction

Nowadays, to obtain qualitative permanent joints of heterogeneous materials the methods of welding in a solid state are widely used. The most prevalent of these is a diffusion bonding. The wide nomenclature of compounds creates a complex of specific requirements for diffusion bonding's energy sources. These requirements are mainly related to the acceptability of a wide range of materials and shapes of products, the accuracy of the specific heat capacity control and the ability of the wide regulation of the sample's temperature [1].

The distributed plasma of a glow discharge burning in a rarefied gas atmosphere at a pressure of 0.1–10 kPa is widely used in processes accompanied by the direct action of charged electric particles on the treated or welded materials. Known works consider the possibility of application of gas discharges technique in the field of thin films deposition, metals surfacing, treatment and modification of metals before welding [2–5]. Still, as practice has shown, diffusion welding [6] and thermal and chemical-thermal surface treatment [7] are the most appropriate. This is due to the high technological capabilities of the glow discharge, which in these processes can serve both a processing tool and a source of thermal energy for their implementation simultaneously. Additionally, a glow discharge has high technical, economic, and environmental indicators, for instance, high

heating productivity, energy savings, and last but not least the absence of environmental pollution [8].

However, during surface treatment or welding of metals, the instabilities of samples temperature (discharge cathode in these processes), the pressure of the working gas, the voltage of the power supply, and a number of other factors, can lead to the transition of a glow discharge into another form of gas discharge—an electric arc [9] (Figure 1). In this case, the distributed glow discharge transforms into a contracted form. Its cathode spot narrows to a very small size, increasing the energy concentration dramatically. A prolonged action of a concentrated arc discharge inevitably leads to the local melting and destruction of samples.

Figure 1. The view of the glow discharge on the cathode surface in stable (a) and unstable (b) burning modes during the ion treatment (the bright spots to the right—arc breakdowns).

The probability of arcing increases with the rising of the total and specific power of the discharge. The energy characteristics of the ionic treatment processes are mainly determined by the magnitude of the discharge current and the gas pressure in the working chamber. The development of processes' productivity leads to the necessity of increasing their values. That, in turn, entails rising of average current density (j) in the cathode spot of the discharge and the average specific volumetric power (jE) in the discharge plasma [10]. However, with increasing of discharge energy characteristics in the interelectrode gap, the short-term local arc breakdowns with a duration of one or two half-periods of the rectified current can form (Figure 2).

Figure 2. Oscillogram of the discharge current in stable (a) and unstable (b) mode. (Glow discharge current 4 A. Oscillograms were recorded from a shunt of 3 Ohm connected to the discharge circuit).

In [11,12], the glow discharge's instabilities are closely associated with its contraction (compression) and transition to a cord form of discharge with the increasing of volumetric power (jE) accordingly. The gas in the cord discharge heats up extremely while the burning voltage decreases and eventually the cord switches into an arc electric. Such a mechanism of glow discharge's instability is associated with volumetric processes in positive column of the discharge plasma, and therefore is more characteristic of extended discharges of the laser type [13], where the length of the interelectrode gap is 0.5–1.0 m. In the processes of ion treatment and welding of metals, a stationary DC glow discharge burns between the electrodes with a limited distance of 0.005–0.05 m. In these conditions, processes in the near-electrode regions of the discharge can affect negatively its stability. The largest voltage drop (100–300 V) and the greatest electric field strength accordingly are observed in the cathode region of the discharge where there are the processes of electron ionization and multiplication which determine the glow discharge existence. These processes are affected by conditions both in the volume of the cathode layer and on the surface of the cathode itself. They are quite fully investigated and described in [14–16]. However, they do not consider the impact of the cathode surface characteristics on the stability of the glow discharge while surface treatment and welding of metals, which makes it impossible to determine and establish their optimal values from the point of view of process productivity and discharge stability. In this regard, the aim of this work is to study the effect of the physicochemical characteristics of the samples surface on the stability of a glow discharge and the development, on this basis, of technological recommendations for the selection of values of the mode parameters that ensure the stable discharge existence while diffusion welding and metals treatment.

2. Methods

The main physicochemical characteristics of the metals surface includes its macro- and micro relief, as well as the presence of chemical compounds on it. Even after machining, the surface of the vast majority of structural metals is covered with a thin layer of natural oxide. Oxide films are not ideal dielectrics, but they possess a certain conductivity. The resistivity (ρ) of most oxide films of the processed metals (Fe, Cu, Mo, W, Ti, etc.) is 10^3 – 10^5 Ohm m, and the characteristic value of the dielectric constant is $\varepsilon \approx 2$ – 10 [17]. Therefore, from an electro-technical point of view, an oxide film's capacitance and resistance are connected in parallel. The capacitive properties of the film appear at times $t_0 \leq \varepsilon \varepsilon_0 \rho \approx 3$ (10^{-8} – 10^{-6}) sec [18]. The maximum electric field strength due to the accumulation of charge on the oxide film over this time is defined as

$$E \approx \frac{1}{\varepsilon \varepsilon_0} \int_0^{t_0} j dt \approx j p, \quad (1)$$

where j —the current density at the cathode of the discharge.

In a normal glow discharge, the gas pressure in the working chamber determines the current density in the cathode spot and at the pressures of 1.33–13.3 kPa it can reach 10^2 – 10^3 A/m². Thus, in a normal DC glow discharges at pressures characteristic of technological processes of metal treatment and welding, the electric field may not reach the values of the electric field strength breakdown for thin oxide films. As it was mentioned in [16], the heating of the oxide films to a 1200 K leads to a rapid decrease of its resistivity (four to six orders of magnitude). Therefore, taking into account the simultaneous heating of the films together with the samples and the noticeable decline in the film resistance with increasing of the temperature, the emergence of arcing breakdown is unlikely. Consequently, the presence of an oxide film on the surface of the cathode (product) can make a massive impact on the discharge's stability but only on the cold cathodes. With the heating of cathode, this factor becomes unimportant.

The next parameters characterizing the state of the cathode surface are its macro- and microrelief. In this case, the glow discharge's stability can be affected with the pronounced roughness protrusions obtained after machining, but not with the smoothly changing surface waviness. Figure 3 shows profile curves of the sample's surfaces obtained after treatment with a roughness of 60–80 μm (Figure 3a) and about 10 μm (Figure 3b), respectively. For this purpose, a profilometer TR—200 was used.

Figure 3. Cont.

Figure 3. The profile curves of the samples surfaces obtained after machining by rotational turning with the roughness about 80 μm (a) and 10 μm (b): 1—single placed; 2—ridge-shaped protrusions.

The external view of the device and treated samples themselves are shown in Figure 4.

Figure 4. The view of a profilometer TR-200 (a) which is made up of: 1—display; 2—control panel; 3—sensor (pickup) and treated samples (b).

The undergoing intense ions bombardment, the protrusions heat up much more noticeably than the bulk of the electrode (cathode). This inevitably leads to the superheat point's emergence on the cathode surface to temperatures exceeding the melting or boiling point of the metal. In this case, a glow discharge can transit to the local vapor arcs [19,20]. This can be observed most likely in case when the surface of the protrusions is large enough to provide heating to high temperatures, and the heat transfer to the bulk of the samples is too small.

3. The Analytical Equation of a Glow Discharge Energy Stability while Treatment and Welding of Metals Depending on Cathode Surface State

Cylinders, cones or hemispheres can be taken as a model of individual micro protrusions [21]. The possibility of their heating to a boiling point is provided:

$$q_s = q_v - q_T, \quad (2)$$

where q_s —the energy perceived by the surface of the protrusion from ions bombarding the cathode: $q_s = SjU_c t$ (where S —the area of the lateral surface of the protrusion, U_c —the cathodic potential drop in the discharge, t —time); q_v —the heat content of the protrusion material at the boiling point: $q_v = Vc\gamma T_{boil}$ (where V —the protrusion volume, c —the heat capacity of the metal, and γ —the density); and q_T —the energy diverted from the protrusion into the sample: $q_T = 2\pi\lambda RT[1 - \operatorname{erf}\left(\frac{R}{4at}\right)]^{-1}$, (where λ —the thermal conductivity of the cathode material, R —the radius of the protrusion base, and T —the cathode surface temperature).

In the processes of thermal ion treatment and diffusion welding, when the temperature of the samples is much lower than the boiling point of metal, condition (2) is feasible if $q_T \rightarrow 0$. In order to neglect the heat transfer to the samples, the value of t must be the same order as the glow discharge's transition time into the arc $t = 10^{-4}$ – 10^{-6} sec. So, then

$$q_s = q_v \text{ or } SjU_c t = Vc\gamma T_{boil}, \quad (3)$$

In this case, for the micro relief of structural steels, it is necessary that the S/V ratio is 10^5 – 10^7 cm^{-1} (where S —the area of the lateral surface of the protrusion and V —the protrusion volume). Nevertheless, even at $t = 1$ sec, it is necessary that the value of $S/V \geq 10^2 \text{ cm}^{-1}$. Such ratios of the surface and volume of the micro protrusion are possible only for thin protrusions and burrs. For real surfaces machined by turning or grinding, this ratio is much less than one, which indicates a low probability of fulfilling condition (2). Hence it follows that at the cathode's current densities corresponding to a glow discharge (up to 10^3 A/m^2), the probability of melting and evaporation of the roughness ridges is pretty small. This limits the possibility of thermionic emission from their vertices.

At the same time, sharp protrusions on the cathode surface creates the local distortions of the electric field nearby the cathode surface. Electric field distortions near the protrusions facilitates the attraction of positive ions toward them and as a result, the active points with the increased charges concentration are formed. This, in turn, leads to dramatic increase in the electric field strength in these regions. The length of this region (d_c) is determined by the cathode material as well as kind and pressure (p) of the working gas, and can be found from the ratio:

$$pd_c = c, \quad (4)$$

where c —a constant value for particular gas and its pressure (for a glow discharge burning in a nitrogen, $c = 0.42 \text{ Pa m}$ [22]). At a nitrogen pressure of 1.33–13.3 kPa, the average electric field strength near the discharge cathode is

$$E_{av} = \frac{U_c}{d_c}, \quad (5)$$

where U_c is the cathode potential drop, which is 215 V for a glow discharge in nitrogen.

In these conditions, the E_{av} reaches up to 10^6 – 10^7 V/m. A further increase in the electric field strength inevitably leads to the appearance of a current of field electron emission from the tip of the protrusion due to the ejection of electrons from the surface by a strong electric field. A noticeable field emission current, sufficient for the existence of an arc discharge, appears at the electric field strength of about 10^9 V/m. In this regard, it is advisable to assess the degree of influence of the protrusions surface roughness on a local increase in the electric field strength at the cathode of a glow discharge. Meanwhile, the magnitude of the field strength does not depend on the number of protrusions, but on their size and shape.

In mechanical engineering, the surface machining of samples of the third—sixth class of cleanliness is widely used. In this case, the maximum height of the surface protrusions varies, respectively, from $Rz = 40$ – 80 to 6 – 10 μm . Since the height of the micro protrusion is much less than the interelectrode gap (of 0.01 m or more), for calculation of electric field of such protrusion the electrostatics task has being solved about the conductive ellipsoid in an external field parallel to one of the main axes of the ellipsoid [23]. This is an equivalent to a semi-ellipsoidal protrusion on one of the flat electrodes, or to the gap between two electrodes parallel to each other (Figure 5) which exceeds protrusion height. If the x is the axis of the protrusion in the form of a semi-ellipsoid of rotation perpendicular to the electrode, where $x = 0$, then the field strength on the extension of this axis when x is greater than the height of the protrusion h is [22]:

$$E(x) = E_{av} \left[\left(1 - \frac{\operatorname{arth} \frac{c}{x} - \frac{c}{x}}{\operatorname{arth} \frac{c}{h} - \frac{c}{h}} \right) + \frac{1}{(\operatorname{arth} \frac{c}{h} - \frac{c}{h})(\frac{x^2}{c^2} - 1)\frac{x}{c}} \right], \quad (6)$$

where E_{av} is the average electric field strength created in the cathode region of a glow discharge by a cathodic potential drop; c —half the distance between the ellipsoid focuses located on the x axis (Figure 5), determined, according to [24], as $c = \sqrt{a^2 - b^2}$.

Figure 5. Scheme of a semi-elliptical protrusion on the surface of a flat cathode: a , b are the semi axes of the ellipsoid; h —the height of the protrusion; d_c —the length of the cathodic potential drop region.

The term in square brackets in this expression describes the field strength increasing due to the presence of a protrusion. Denote it by β_E , then the expression (6) can be written as $E(x) = E_{av} \beta_E$. The term in parentheses of expression (6) is always less than one and on the surface of the protrusion at $x = h$ vanishes. Therefore, the main is the second term, the value of which is maximum at minimum x , i.e., at $x = h$ and at the $x = d_c$ simultaneously. Figure 4 shows the dependence of the field enhancement β_E on the ratio h/d_c . Expression (6) describes an increase in the field strength on a flat smooth cathode from a protrusion having a smooth peak with a considerable radius. For a hemispherical protrusion having the same semi axes $a = b$, the field enhancement is $\beta_E \approx 3$ [25]. The results of calculating β_E from expression (6) for surface protrusions in the form of semi-ellipsoidal extended along the x -axis are shown in Figure 6. The graph also shows that, depending on the degree of protrusions elongation, the electric field enhancement at their apex can reach up to 10–30.

Figure 6. The dependence of the field enhancement on the cathode surface on the ratio of the protrusion's height and the length of the cathodic potential drop region: 1—ratio $a/b = 4$; 2—ratio $a/b = 2$.

In practice, after machining protrusions are formed on the surface, having an acute-angled shape, with a radius at the apex within the unit fractions of micrometers (Figure 3). The acute-angled protrusions contribute dramatically to a more intense electric field distortions and an additional increase in its intensity nearby the protrusion. Such an increase in electric field strength can be estimated by the coefficient [21]:

$$\mu = \frac{h}{r}, \quad (7)$$

where h —the height of the protrusion; r —the curvature radius on the peak of the protrusion.

For protrusions with a height of 20–40 μm or more with a radius of vertex curvature from fractions to units of micrometers (Figure 3), the value of μ can reach up to $\mu \geq 10$. Then the field strength near the vertices of such protrusions will be

$$E(x) = E_{av}\beta_E\mu, \quad (8)$$

Under conditions of increased gas pressure, the boundary of the region of cathodic potential drop declines sharply and approaches the vertices of micro protrusions roughness $x = d_c \approx h$. As a result, the local field strength near the vertices of such protrusions can reach $E(x) \geq 10^9 \text{ V/m}$. Such values of the local electric field strength provide for a current density of field emission from the peak of the protrusions of $j_{fe} \geq 10^7 \text{ A/m}^2$, sufficient for an arc breakdowns exciting in the interelectrode gap [26]. The latter is in many orders higher than the current density in the cathode spot of the glow discharge. It is suggested that in these conditions the Joule's heating and evaporation of the vertices of the protrusions entail the arc breakdowns [27]. In this case, condition (2) can already be fulfilled, i.e., the probability of melting and evaporation of protrusions increases dramatically. In turn, the latter contributes to the development of thermionic emission from these areas with the arc formation on the surface of the cathode spot of an arc discharge. As was mentioned above the long-term action of a stable arc discharge on a surface of the samples can lead to their melting and destruction.

Hence, for the prevention of a glow discharge transition into an electric arc, the length of the cathodic potential drop must exceed the maximum height of the roughness protrusions. The cleanliness class of the samples surface treatment determines this.

The adequacy of this assumption has been checked under conditions of ion treatment in a glow discharge in a nitrogen. The cylindrical c with dimensions of $20 \times 60 \text{ mm}$ made of steel A 659 CS Type 1020 (cathode of discharge simultaneously) were used. The glow discharge was powered from a controlled full-wave rectifier with an output voltage of 0–1000 V through a ballast resistor of 80 Ohm. The discharge current was of 4 A. A flat

annular anode located at a distance of 0.008 m from the cathode surface was used as the second discharge electrode (Figure 7).

Figure 7. The schematic view of experiments: 1—treated samples (cathode); 2—anode ring; 3—plasma of glow discharge; d_c —the length of the cathode potential drop.

The steel samples obtained by rough, semi-finishing and finishing had the height of the surface roughness protrusions of 60–80, 30–40, and 10–15 μm accordingly. During treatment, a gradual increase in gas pressure in the working chamber was performed.

As the criteria of glow discharge stability, the limiting pressure at which the short-term arcs at the interelectrode gap emerge developing, apparently from the tops of the highest or most sharp protrusions was chosen. A further rising of a gas pressure is accompanied by an increase in the frequency of microarc discharges formation until a stable electric arc is established in the discharge gap. The cathode (products) temperature while treatment was 700–1000 K. The cathode's temperature was measured with the chromel-alumel thermocouple at three different points in the axial direction and then the results was averaged. The view of treatment by stable glow discharge and in the moments of arcing breakdowns (blurred by flare and multitude of micro-arcs) are shown in Figure 8.

Figure 8. The visualized scheme of treatment in a stable (a) and disturbed (b) by a multiple micro-arcs breakdown glow discharge.

The experimental results are shown in Figure 9 in the form of a curve corresponding to the averaged over the results of a number of measurements, the limiting values of gas pressure depending on the parameters of the surface relief. Here along with the dimensions of the roughness protrusions, the values of the extent of the cathodic potential drop region corresponding to the gas pressures are given.

Figure 9. The experimental dependence of the working gas pressure (P) on the surface roughness parameters (Rz) according to the conditions of glow discharge stability: (d_c —cathodic potential drop region).

The obtained results indicate that the glow discharge's instabilities really begin to appear when the boundary of the cathodic potential drop region (d_c) approaches the vertices of the micro protrusions. A glow discharge remains stable at gas pressures below the obtained experimental curve. Knowing the samples surface characteristics in the form of the $Rz = R_{\text{max}}$ quantity, enables to determine the gas pressure boundary values for the various technological processes of ion treatment or diffusion bonding, as well, in advance.

4. Conclusions

It is shown that the low temperature plasma of DC glow discharge which burns in the active or inert gases at the medium pressures is the perspective source of surface heating in the processes of diffusion bonding and ion treatment of metals. At the same time, a number of factors on the cathode surface can emerge, which leads to the transition of a glow discharge into an electric arc as a more stable form of gas discharges. In this study, the impact of macro and micro relief of the samples on the stability of a glow discharge while diffusion bonding and treatment of metal has been analyzed and analytical equation of a glow discharge energy stability boundary has been obtained. The main conclusions of this study can be summarized, as follows:

1. The emergence of oxide films on the surfaces of specimens during welding or metals treatment does not lead to a significant disruption of a glow discharge stability as long as electrical field strength does not exceed the breakdown values.
2. On the other hand, the roughness of a cathode's surface affects the glow discharge stability at the working gas pressures when the height of the protrusions roughness becomes comparable to the length of the cathode potential drop region d_c .
3. The direct dependence of the d_c length on the gas pressure allows to determine the limit values of the latter based on the given characteristics of the samples surface microrelief which ensures the stable glow discharge existence during ion treatment and welding of metals. In our experiments the increase of the cathode surface roughness from 10–15 μm to 60–80 μm led to a rapid decrease of the region of the limiting

pressure of the stable glow discharge existence from 1.33–13.3 kPa to a 1.33–5.3 kPa, respectively.

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