Pulsed non-self-sustained glow discharge with a large-area hollow cathode for nitriding of iron-based alloys

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Abstract. The paper reports on a study of a pulsed non-self-sustained glow discharge with a hollow cathode area of 2.3 m² and pulse current of up to 370 A. The ignition and operation of the discharge at low pressures (0.4–1 Pa) was sustained through electron injection from the plasma of an arc discharge based on a cold hollow cathode. The current-voltage characteristics of the pulsed glow discharge were measured. Specimens of 12 Cr18Ni10Ti, Cr12, and 40Cr steels were nitrided in the glow discharge plasma with double thermal shields at a temperature of 520 ºC and pressure of 0.9 Pa for 4 h. The nitrided layer thickness for 40Cr, Cr12, and 12Cr18Ni10Ti steels was 240, 90, and 50 μm, respectively.

1. Introduction

Research in low-pressure discharges for surface modification of materials is assuming more and more significance. New avenues for surface treatment are opened up by pulsed discharges [1] which allow operation at increased powers (one-two orders of magnitude higher than those for steady discharges) and control of discharge pulse durations [2]. Among the methods of ion nitriding as a tool of chemical heat treatment is the use of a pulsed glow discharge which allows control of its operating voltage and treatment of different metal materials [3]. In this type of discharge, the average ion current density and temperature of treated parts are controlled by varying the discharge pulse duration at a constant pulse repetition frequency [4]. Despite much progress made in ion nitriding technologies, there still remains a series of problems such as surface roughness of treated parts due to etching, their nonuniform heating, and initiation of microarcs on their surface. These problems are urgent for both pulsed and steady glow discharges. It has been shown that a steady low-pressure non-self-sustained glow discharge based on a hollow cathode have a high potential for technological applications, including ion nitriding of Fe- and Ti-based alloys [5–10]. A special feature of this discharge is the possibility of its operation at decreased pressures 0.4…1 Pa which enables efficient surface cleaning during nitriding. Moreover, the discharge is initiated at low voltages 100…350 V, making it possible to greatly decrease surface etching and to attain minimum surface roughness. In pulsed mode, this type of discharge provides a possibility to independently control the main parameters of the process: the energy of ions bombarding the surface of treated parts, their temperature, and pressure in the chamber.

By now, no theoretical and experimental research has been performed on the formation of a low-pressure (0.4–1 Pa) non-self-sustained glow discharge with a large-area hollow cathode (>2 m²) in pulsed mode at a pulse current more than 100 A and pulse power higher than 30 kW. Also no experimental studies have been reported on nitriding of Fe-based alloys in this type of discharge under the conditions specified above.
The objective of the present study was to investigate the characteristics of a pulsed non-self-sustained hollow-cathode glow discharge and the possibilities of its use for nitriding of 12Cr18Ni10Ti, Cr12, and 40Cr steels.

2. Experimental setup

The experimental setup used in the study was built on the basis of NNV-6.6-I1 commercial ion plasma unit (Fig. 1). The vacuum chamber was pumped with a TMN-500 turbomolecular pump to a pressure of $1 \times 10^{-2}$ Pa. The operating pressure was controlled in the range 0.4…1 Pa by varying the gas (nitrogen) flow rate.

The glow discharge was ignited between a hollow cathode (inner surface of the chamber) and a plane anode. The cathode area was $2.3 \times 10^4$ cm$^2$, the anode area was 1350 cm$^2$, and the ratio of these areas was thus 17/1. The main discharge was powered by a stabilized voltage source with an output voltage of 30…320 V, output pulse current of 0…500 A, and maximum average output power of 25 kW; the output voltage instability was ±5%. The power supply was equipped with a protection system which precluded initiation of microarcs at the cathode surface. The discharge current was measured using a Honeywell CSNJ481 Hall sensor with a current output. The voltage was measured using a probe with a divider of 1:100. The signals were transmitted to a TDS2014C oscilloscope and averaged over 16 points. To provide a flat peak during the entire voltage pulse, a capacitor bank with a capacitance of 7.2 mF was used.

For easier ignition and stable operation of the glow discharge at low pressures, an electron source based on an arc discharge with a cold hollow cathode was used [6]. The working gas to the discharge chamber was supplied through this electron source arranged at the chamber gate. Electrons were emitted to the hollow cathode (chamber) through an electrode shaped as a truncated cone and covered on all sides with a fine grid of dimensions 0.4×0.4 mm and geometric transparency 45%. This electrode served as the anode of the auxiliary discharge and was simultaneously at the potential of the main discharge cathode. The plasma generator was powered by a welding rectifier which ensured a constant arc current of up 200 A at an arc voltage of 30…40 V; the current of the magnetic coil in all experiments was 0.18 A. The current and voltage of the auxiliary discharge were measured using pointer instruments with recording of their effective values.

Figure 1. Schematic of the experimental setup (top view).

The current-voltage characteristics of the pulsed non-self-sustained glow discharge were taken from oscillograms of the current and voltage at 320 µs of the discharge pulse during the operation with
a frequency of 1 kHz and duty factor of 34 %. When these characteristics were recorded, the discharge voltage varied in the range 40…320 V. The pressure dependence of the main discharge current was determined at an auxiliary discharge current of 40…120 A. Before the measurements, the chamber walls were subjected to ion cleaning for which a pulsed non-self-sustained glow discharge was initiated in the working chamber through nitrogen supply and its walls were bombarded by ions for 30 min at a voltage of 300 V and average current of 25 A.

To decrease thermal loss at the water-cooled walls of the vacuum chamber, double shields made of polished stainless steel were used in nitriding. In this experiment, the thermal shield walls facing the discharge gap served as the hollow cathode of the glow discharge.

For nitriding, the 12Cr18Ni10Ti, Cr12, and 40Cr steel specimens made to dimensions 15×15×5 mm were fixed in a holder placed at the potential of the hollow cathode. The working gas mixture contained 90 % of nitrogen and 10 % of helium. The addition of helium to nitrogen was made to increase the nitriding rate [9]. The specimens were nitried for 4 h at a gas pressure of 0.9 Pa, discharge operating voltage of 280 V, and average ion current density of 1.7 mA/cm$^2$. The specimen temperature was 520°C and was controlled at a constant ion energy by varying the pulse duty factor which was 40 % at a pulse repetition frequency of 1 kHz (pulse duration 400 μs, pause 600 μs). The ion current density to the specimens reached 4.4 mA/cm$^2$ in a pulse. Heating and cleaning of the specimens was through bombardment by ions accelerated in the cathode layer of the non-self-sustained glow discharge. Before arrangement into the vacuum chamber for nitriding, the specimens were grinded and polished, washed with benzene and acetone in an ultrasonic bath to remove organic impurities, and wiped with ethanol-wetted calico.

The nitriding efficiency in the pulsed non-self-sustained glow discharge plasma was estimated by measuring the Vickers microhardness on lateral sections in depth from the specimen surface with a step of 10 μm at an indenter load of 0.2 N. The measurements were performed on a PMT-3 device. The in-depth microhardness distribution was measured for each specimen until its initial microhardness was reached, whereupon the result was averaged over three measurements at each point.

3. Results and discussion

Waveforms of the main discharge current at two values of the auxiliary discharge current are presented in Fig. 2. The current rise and fall times, regardless of the pulse duration, are rather long due to a gradual increase in plasma density when voltage is applied to the discharge gap and to deionization of the discharge gap when it is removed. The discharge current in the experiments reached 370 A in a pulse. The discharge pulse power was about 90 kW.

Figure 2. Waveforms of the current and voltage of the non-self-sustained glow discharge at two auxiliary discharge currents: $I_1=60$ A and $I_2=100$ A; discharge operating voltage $U_d=240$ V, pulse repetition frequency $f=1$ kHz, duty factor $\gamma=34$ %, pulse duration $t_p=340$ μs, operating pressure $p=1$ Pa.
Figure 3a shows a current-voltage characteristic of the discharge at an operating pressure of 0.65 Pa.

The discharge current is the cathode current and equals to the product of the amount of injected electron current from the auxiliary discharge and $\gamma$-electron current formed as a result of the secondary ion-electron emission from hollow cathode walls to the average number of ionizations produced by these electrons after acceleration in the discharge cathode layer. In turn, $\gamma$-electron current is equal to the product of the ion current to the secondary-ion-electron emission factor $\gamma$. Increasing the discharge voltage increases the average number of ionization events realized by each of the electrons accelerated in the cathode layer. Because the voltage in the near-cathode layer at the initial portion of the current-voltage characteristic is low, the secondary-ion-electron emission factor $\gamma$ is small and $\gamma$-electrons have low influence on the characteristic. Beginning with a discharge voltage of about 200 V, the rate of rise of the current increases which can be explained by a considerable increase in the number of $\gamma$-electrons [11].

The dependence of the main discharge current on the auxiliary discharge current (Fig. 3b) suggests that the efficiency of the hollow cathode increases with increasing discharge operating voltage. The characteristics are linear because the cathode current is proportional to the amount of auxiliary discharge electron current and current of $\gamma$-electrons. When $U_d = \text{const}$ the secondary ion-electron emission factor remains constant and the growth of the cathode current is carried out by increasing of the electron current from the auxiliary discharge and the ion current to the cathode. Increasing the discharge operating voltage increases the rate of rise of the discharge current due to an increase in the fraction of electrons formed in secondary ion-electron emission. Moreover as noted above increasing the discharge voltage increases the average number of ionization events realized by each of the electrons accelerated in the cathode layer.

The dependence of the main discharge current on the operating pressure at a constant discharge voltage (Fig. 3c) demonstrates that the discharge current has a slight decrease in the investigated range of operating pressures.

Figure 3a. Current-voltage characteristic of the non-self-sustained glow discharge at an auxiliary discharge current of $I_a=40$ A.

Figure 3b. The main discharge current vs the auxiliary discharge current at a constant discharge voltage.
Figure 3c. The main discharge current vs operating pressure at a constant discharge voltage.

This can be explained by the fact that for a given area ratio of anode and cathode the upper limit of the pressure range within which there will be a constant value of the discharge voltage is 0.33 Pa. Above this value, according to [12] there will be an increase of the discharge voltage at a constant current discharge. The dependences in Figure 3c shows that at the constant discharge voltage in the pressure range (0.3 - 1) Pa discharge current decreases slightly.

Figure 4 shows in-depth microhardness distributions for 12Cr18Ni10Ti, Cr12, and 40Cr steels nitrided in the glow discharge plasma. The result of nitriding in the nitrogen-helium mixture for 4 h is a substantial increase in surface layer microhardness.

Figure 4. In-depth microhardness distributions for 12Cr18Ni10Ti, Cr12, and 40Cr steels nitrided in the glow discharge plasma for 4 h at a temperature of 520 °C and nitrogen-helium pressure of 0.9 Pa. Dotted lines – initial microhardness.

In-depth microhardness distributions show that the nitrided layer depth for 40Cr steel is 240 μm; for Cr12 steel, it is 90 μm; and for 12Cr18Ni10Ti, it is 50 μm. This result evidences that the surface of the steels is nitrided with a sufficiently high efficiency. In comparing the nitriding processes in pulsed and steady modes, it should be noted that the nitriding rate for 40Cr steel in a steady non-self-sustained hollow-cathode glow discharge was about the same [9]. Despite this, an important advantage
of the pulsed non-self-sustained glow discharge is the possibility to control the temperature of treated parts by varying the pulse duty factor at a specified discharge voltage amplitude over a rather wide range of operating pressures.

4. Conclusions

Thus, we realized a pulsed non-self-sustained glow discharge with a large hollow cathode area (>2 m²), pulse current of up to 370 A, and pulse power of up to 90 kW. The measured current-voltage characteristics of the discharge demonstrate stability of the discharge current in the operating pressure range 0.4–1 Pa, showing promise for chemical heat treatment of materials and parts with a large surface area.

Nitriding of 12Cr18Ni10Ti, Cr12, and 40Cr steels in the non-self-sustained glow discharge plasma at a temperature of 520 ºC for 4 h was realized. The nitrided layer depth for 40Cr, Cr12, and 12Cr18Ni10Ti steels was 240, 90, and 50 μm, respectively.

The research results suggest that pulsed non-self-sustained glow discharges with a large-area hollow cathode have a high potential for increasing the efficiency of nitriding of steels and alloys.

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