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Steel nitriding by atmospheric-pressure plasma jet using N_2/H_2 mixture gas

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ABSTRACT

We have succeeded in hardening surface of tool steel with a new plasma-assisted nitriding treatment, in which the pulsed-arc plasma jet is generated with N_2/H_2 mixture gas and the jet plume is sprayed onto steel surface under atmospheric pressure. We discovered that the thickness of the formed hardened layer is varied by H_2 mixture ratio of the operating gas. Moreover, there exists an optimal H_2 ratio for nitriding, the value of which is 1% under our experimental condition. From experimental results of optical emission spectroscopy for the jet plume and X-ray diffraction from treated steel surface, we consider that the production of NH radicals and surface oxidation are possible to relate to the mechanism governing the dependence of nitriding quality on H_2 ratio.

1. Introduction

Plasma-assisted nitriding (PAN) treatment is one of the surface-hardening technologies utilized for a number of mechanical products such as automobile components and dies [1]. In recent years, PAN treatment for austenitic steels is actively researched to improve wear and corrosion resistance of stainless steels [2–26]. Such state-of-the-art PAN treatments include surface patterning of steels [27]. On the other hand, the technology of PAN treatment for ferrite steels, e.g., tool steels, has been well researched and matured to become widespread especially in automobile industry [28–48]. Nitriding for tool steels produces the diffusion layer composed of Fe–N solid solution and nitride precipitates, and the compound layer composed of iron nitride. These layers increase the wear, fatigue, and corrosion resistances. In industry, PAN treatment is performed using a DC or pulsed DC glow discharge generated in a low-pressure nitrogen/hydrogen gas mixture or NH_3. This technique is referred to as ion nitriding [19,47]. On the other hand, a number of novel PAN technologies have been developed in academic researches on the basis of low-pressure plasma technology [28–48]. As the treatment requires a vacuum system, however, the conventional PAN treatment can be operated basically as a batch process and the capital cost becomes very high. To eliminate these shortcomings, we are developing a new PAN treatment using atmospheric-pressure plasma technologies. To the best of our knowledge, there are two literatures on surface hardening by atmospheric-pressure plasma nitriding except us. Yan et al. have succeeded in hardening steel surface using a dielectric barrier discharge [49]. This technique exhibits that the hardness profile strongly depends on the discharge gap length. On the other hand, Hassel et al. used a transmitted arc discharge [50]. They found that the treated surface was hardened only slightly, but the deeper area showed a rapid fall of hardness down to the value lower than that of the base material.

Of a wide variety of atmospheric-pressure plasmas, we have adopted the pulsed-arc (PA) plasma jet for three reasons. First, the plume temperature of the PA discharge is much lower than the DC arc discharge because of pulse excitation, thereby enabling surface treatment without the material melted, roughened, and improperly annealed. Second, the PA discharge is suitable to produce high density reactive species [51], owing to the fact that the input power can be increased in contrast to barrier discharges. Third, the visible plume length of the PA jet extends from ca. 20 mm to 200 mm in N_2 atmosphere when the residual O_2 decreases to less than 0.3% [52], which should be advantageous in treating complex-shaped workpieces with holes or narrow gaps. These three advantages indicate that the PA plasma jet has a potential to offer a new practical method of nitriding. In our previous investigation, only spraying the nitrogen plasma jet did not harden the sample surface. Instead, an oxide (Fe_2O_3) layer of 10 μm in thickness was formed on the sample surface. This result indicates that the formed oxide layer is likely to interrupt nitriding. Therefore, we have added the H_2 gas to the ambient nitrogen atmosphere to reduce the residual O_2 for preventing undesirable oxidization. As a result, we have succeeded in hardening the steel surface [53].

Here, the characteristics of plasma-jet nitriding are reported in detail, particularly the effect of H_2 addition on the quality of nitriding. We have recently revealed that the H_2 addition to the operating N_2 gas provides advantages for industrial use, compared with the H_2 addition to the ambient N_2 atmosphere which was reported in [53]. Thus, here we concentrate on the use of N_2/H_2 mixture gas as the operating gas for discharge. The detailed characteristics reported here for the

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first time will be most valuable for future practical use of plasma-jet nitriding.

2. Experimental procedure

The jet nozzle is composed of a coaxial cylindrical electrode system as shown in Fig. 1. The grounded external electrode measures 35 mm in inner diameter and has an orifice of 4 mm in diameter at the tip. The curvature radius of the tip of the internal electrode is 4 mm. The discharge gap is approximately 18 mm. N_2 gas and H_2 gas are introduced into the nozzle through separate mass flow controllers (HORIBA STEC, SEC-E60, SEC-E40, respectively). The purities of the used N_2 and H_2 gas are 99.99% and 99.97%, respectively. The low-frequency voltage pulse (4–5 kV in height and 21 kHz in repetition as shown in Fig. 2) is applied to the inner electrode using a high voltage power supply (plasmatreat, FG3001). The maximum of the discharge current is ca. 1 A. The afterglow of the generated PA plasma is spewed out from the orifice, forming the jet plume.

Fig. 3 shows the schematic diagram of our experimental setup. Plasma-jet nitriding is performed in an air-tight cylindrical container (153 mm in inner diameter and 223 mm in height) made of stainless steel. The container is fitted with a quartz viewport for spectroscopic observation of the plasma jet plume. The jet nozzle is inserted into the container from the upper end. Prior to the generation of plasma jet, residual oxygen (O_2) inside the container was purged by N_2 gas introduced through the nozzle at 20 slm. We estimated from the plume extension mentioned in the previous section that the purge duration of 10 min is adequate to eliminate residual O_2. The gas flows out through four exhaust ports (3/8 in. in diameter) mounted at the lower end. The container is not fitted with any pumping system so that the pressure inside the container is kept at 1 atm.

Nitriding response is examined by spraying the jet plume onto the surface of disk-shaped samples (20 mm in diameter and 4 mm in thickness) made of die steel JIS SKD61 (corresponding to AISI H13) which is frequently used to benchmark nitriding [28,35,38,42–44,46]. The composition is the following: 0.38% C, 0.42% Mn, 0.92% Si, 5.12% Cr, 1.19% Mo, and 0.8% V. The hardness of samples has been adjusted to 600 Hv by heat treatment. The sample surface is mirror finished with alumina powder (0.3 μm) and degreased in an ultrasonic acetone bath. The sample is put on a ceramic heater to control the treatment temperature. The distance between the nozzle tip and the sample surface is 15 mm, which was determined for the reason that the plume temperature at this distance is lower than 500 °C. As a result, the treatment temperature of the sample can be controlled by the ceramic heater independently of heating by the plume. In this experiment the treatment temperature is set to be 540 °C. The treatment duration is 2 h. The visible jet plume covers the entire surface of the sample during the treatment as shown in Fig. 4.

The hardness profile is measured using a micro Vickers hardness tester (Akashi, HM-124). To obtain the metallographic structure, the sample cross-section is chemically etched with 3% nital and observed using a metallograph (Nikon, MM-800/LMU). The X-ray diffraction pattern is observed using X-ray diffractometer (XRD) (Philips, X'pert), where Cu Kα radiation is used. The nitrogen distribution is measured using an electron probe micro analyzer (SHIMADZU, EPMA-1720).

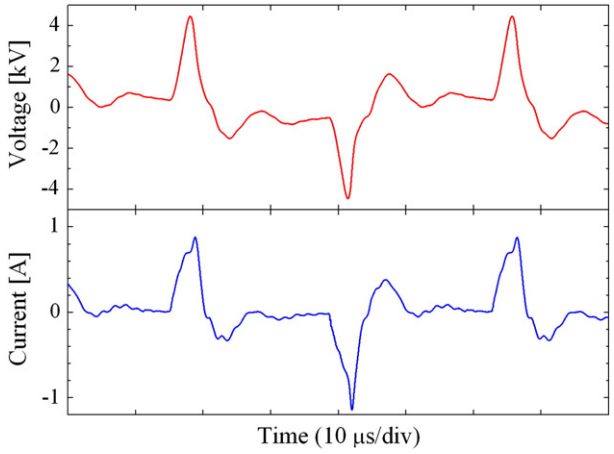

Fig. 2. Typical waveforms of the applied voltage and discharge current.

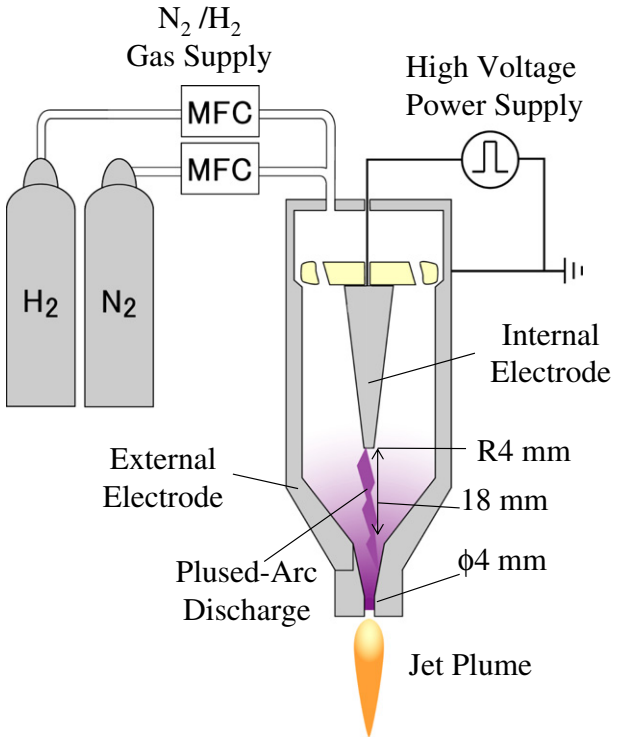

Fig. 1. Schematic of the pulsed-arc plasma jet system.

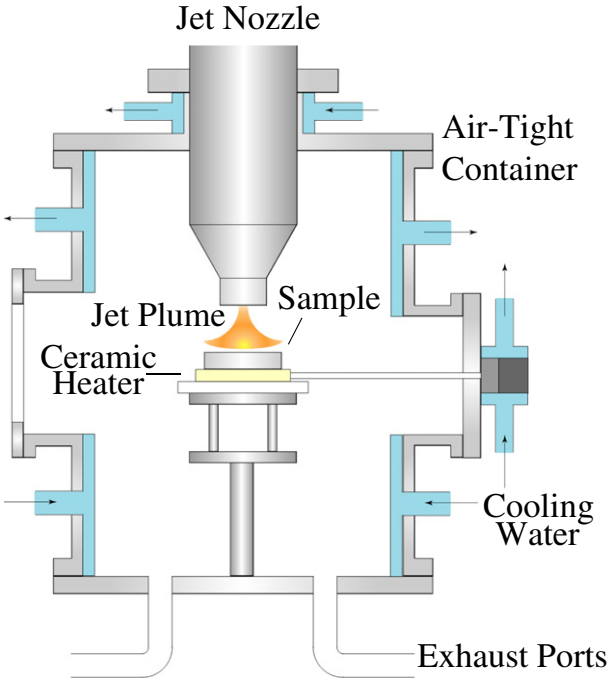

Fig. 3. Schematic of the experimental setup.