Title:

Transferred-Arc Cleaning of Tungsten

Author(s):

K. J. Hollis, R. G. Castro, B. D. Bartram

Submitted to:

<http://lib-www.lanl.gov/la-pubs/00796481.pdf>

Transferred-Arc Cleaning of Tungsten

K. J. Hollis, R. G. Castro, B. D. Bartram
Los Alamos National Laboratory, Los Alamos, NM 87545 USA

Key Words: Transferred arc, cleaning, roughening, cathode spot, tungsten

Abstract

Transferred-arc cleaning is being investigated as a solvent-free cleaning method for various metallic substrates. With the recent increase in attention given the hazards involved in the storage, use and disposal of organic solvents, cleaning methods which promise comparable cleaning effectiveness with reduced hazards are being sought. Transferred-arc cleaning of tungsten substrates has been studied to identify the effect of processing conditions on cleaning and roughening characteristics. A Box-Behnken response surface designed experiment varying the chamber pressure, substrate standoff distance and plasma torch arc current while observing the transferred-arc voltage, current, surface cleanliness and surface roughness was performed. The results of the analysis show the effect of the various independent variables on the measured responses. Particular difficulties in roughening tungsten are due to its exceptionally high arc voltage for metal arc attachment. The results presented here provide an enhanced understanding of the arcing properties of various cathode materials. Such information is useful in obtaining the desired cleaning and/or roughening of the substrate.

Introduction

The initiation of an arc at a cathode spot is thought to take place by a combination of thermionic and field-enhanced emission of electrons [1]. The specific location of the cathode spot depends, among other factors, on cathode surface variabilities such as oxides, contaminants, inclusions, grain boundaries or metallic edges or protrusions. The presence of these surface variabilities attracts the cathode spot. When the arc attaches, the surface variabilities are removed by melting, ablation and evaporation. The cathode spot then moves to another surface variability and the process continues until the surface is relatively clean and uniform.

Transferred-arc (TA) cleaning and roughening as discussed in the patent by Muhlberger [2] is often used as a surface preparation method prior to low pressure plasma spray (LPPS) deposition of coatings. TA surface preparation has been shown to improve coating bond strength as a result of cleaning and roughening of the substrate material [3-6]. The increased surface roughness of the substrate after TA preparation causes increased mechanical bonding between the substrate and coating. The removal of oxide and other contaminant layers promotes metallurgical bonding of the coating to the substrate under specific conditions.

Another possible application of TA cleaning is the removal of thin films of contamination. Such contamination could include oxides, other compounds formed with the substrate material, organics, and metals. In some cases the roughening of the sample is desirable (such as in preparation for coating deposition) but, in other cases, cleaning without roughening is desirable (such as in cleaning finished parts). In order to achieve cleaning without roughening, a good understanding of the arc behavior is needed and proper control of the process is required. Such a study was previously performed for copper and was reported in Ref. 7. In this present work, the TA cleaning of tungsten substrates was investigated to determine the effect of cleaning parameters on the measurable response variables of the cleaning process.

Experiment

The experimental apparatus for TA cleaning experiments is described elsewhere [7] and will be briefly described here. The TA experiments were carried out in an inert environment LPPS chamber using a Praxair SG-100 torch and a Miller Electric, Inc. DC constant current TA power supply. The torch gas used

was argon and the flow rate was 40 standard liters per minute for all experiments. The TA cleaning current for each sample was 70 A. The cleaning process was monitored using a digital video camera while TA voltage and current were recorded on a digital oscilloscope. The samples were characterized by mass and surface roughness measurements before and after cleaning. The experimental setup is shown in Fig. 1.

Figure 1. Plasma torch and TA power supply configuration used for cleaning experiments.

The TA experiments were carried out on tungsten disks 25 mm in diameter by 1 mm thick. The disks were mounted on a copper pedestal, which was connected to the TA power supply and shielded by a ceramic sheath. A hole in the center of the pedestal was attached to a vacuum pump to provide suction to keep the sample in place. The plasma torch was swept over the sample in a single back-and-forth pattern.

TA cleaning runs were carried out at 3 different settings for each of the three independent variables of torch current (250, 325, 400 A), substrate standoff distance (76, 102, 127 mm) and chamber pressure (4, 10.67, 17.33 kPa). The type of statistical design chosen was a response surface method using the Box-Behnken data point sampling design. The Box-Behnken design avoids the extreme vertices used in the central composite design. For the experiments described here, the points at the extreme vertices often did not even produce an arc between the torch and the substrate. Using the Box-Behnken design allowed more valid data points to be collected for the same range in independent variables. A total of 15 different experiments were conducted including three center points. The experiments were carried out in random order.

In order to assess the effectiveness of surface cleaning, one tungsten sample was exposed to a deuterium/carbon plasma to deposit a deuterium containing carbon film on the sample surface. The deposition was performed by Dr. Russell Doerner at the PISCES-A facility at the University of California-San Diego. Surface carbon and deuterium levels were measured for several samples before and after cleaning at the Los Alamos Ion Beam Materials Laboratory using the elastic recoil detection technique [8]. The other samples used in this study were heated in air to cause the formation of an oxide film on the surface of the tungsten. Extent of removal of the dark oxide layer was determined by optical inspection of the bright metallic samples.

Results and Discussion

Tungsten Arc Voltage Characteristics

Table 1 shows the arcing voltage for several clean metal electrode surfaces. This data indicates that the arcing voltage for tungsten is higher than that for other metals. This means that if there is any other metal that is electrically attached to the power supply in the vicinity of the tungsten sample, it will have a lower arcing voltage and will thus attract the arc before the tungsten does. This undesired arcing to other materials is a common problem while TA cleaning tungsten. The arc is more likely to attach to the fixturing or the powder lead than the tungsten sample. Careful shielding of the conduction path components is necessary to confine the cathode spot to the tungsten sample.

Table 1. Representative arcing voltage to clean metal electrodes [9].

Material	Arcing volts
Stainless Steel	16.2
Al	16.7
C(graphite)	20
Cu	21.5
Mo	24
W	26

An analysis of the voltage data taken while cleaning the sample revealed its dependence on the chamber pressure, torch current, substrate standoff distance and interaction terms. The cleaning arc voltage was statistically fit to determine its dependence on the independent variables. The pressure and substrate standoff distance were found to be statistically significant (95% confidence interval) while the torch current and the interactions were found to be statistically insignificant. The model is given by Eq. 1

$$V_{\text{cleaning}} = 44.0 + 0.000524 (P-10.67) + 0.185 (S-102) \quad (1)$$

where V_{cleaning} is in volts, P is the pressure(kPa) and S is the standoff (mm).

After the surface contamination is removed, the arc can attach to the tungsten metal. However, the voltage required to attach to the base metal is significantly higher than the corresponding voltage for arcing to the contaminant layer. For the conditions in the experimental matrix, the power supply was not able to generate sufficient voltage to maintain a tungsten metal arc. Therefore, the metal arc voltage for most of experimental conditions could not be measured. However, tungsten metal arcing was observed on two of the samples. The cleaning arc voltages were 49.2 and 42.2 V while the metal arc voltages were near 60 and 55 V respectively for these samples. This indicates that the metal arc voltage increase over the cleaning arc voltage is in the range of 12 V. The inability of the TA power supply to sustain a metal arc to the tungsten samples prevented an estimate of the metal arcing voltage for the other samples.

The arc voltage values for tungsten described above can be compared to similar values found for copper and stainless steel in previous investigations [7]. Fig. 2 shows the cleaning and metal arcing voltage for tungsten, copper and stainless steel. The various values correspond to different chamber pressures, torch

Figure 2. Cleaning and metal arcing voltages for tungsten, copper and stainless steel as a function of cleaning conditions.

currents and substrate standoff distances. The metal arcing voltages descend in order for W, Cu and SS with differences in voltage which are in good agreement with Table 1. The higher overall voltages in Fig. 1 than Table 1 are due to different arcing parameters used in the present experiment than those used in Ref. 9. As expected, the cleaning arc voltages are lower than the metal arc voltages in all cases. However, the

difference between metal arc voltage and cleaning arc voltage is a strong function of material. The stainless steel actually showed the highest cleaning voltage (though close to that of copper) while the tungsten showed the lowest.

The significance of these results is that if samples of these three materials are close together and all connected to the power supply, the cleaning will take place on the tungsten first then the copper and stainless steel. The metal arcing will take place on the stainless steel first and will occur less frequently or not at all on the copper and tungsten. For instances where cleaning is desired but metal arcing (and the surface roughening which results) is not desired, materials with large differences in the cleaning and metal arc voltages are preferred. The large difference in voltage allows a more distinct transition from cleaning to metal arcing allowing the process to be halted after cleaning but before significant roughening has taken place.

Erosion rate

The sample weights before and after cleaning were used to determine the amount of sample mass lost during arcing. The values for arc erosion rate are commonly reported in units of $\mu\text{g}/\text{Coulomb}$. Therefore, a measurement of the current integrated over the time of cleaning is necessary for reporting the erosion rate. As described above, cleaning arc time far exceeded the metal arc time for all samples. Several samples had no metal arcing and no sample had more than 5% of the arcing time showing metal arcs as indicated by the video footage. Some spurious arcing to the sample holder was observed for several samples thus leaving uncertain the exact arcing time to the tungsten sample. In addition, arcing to the sample holder may have caused material deposition on the sample that would increase the uncertainty of the mass loss measurements. With these limitations in mind, the erosion rates were measured to be from 12 to 36 $\mu\text{g}/\text{C}$. The average value was 23 $\mu\text{g}/\text{C}$. The estimated uncertainty is $\pm 6 \mu\text{g}/\text{C}$.

Reported values from the literature for the metal arc erosion rate of tungsten include 55 $\mu\text{g}/\text{C}$ [10] and 62 $\mu\text{g}/\text{C}$ [11]. The cleaning arc erosion rate is expected to be much smaller with typical values less than 10 $\mu\text{g}/\text{C}$ [12]. It is expected that the tungsten erosion during TA cleaning would be between these values of 10 and 62 $\mu\text{g}/\text{C}$. Furthermore, since there was considerably more cleaning arc time than metal arc time, the net erosion should be closer to 10 $\mu\text{g}/\text{C}$ than 62 $\mu\text{g}/\text{C}$. This is indeed the case in the average tungsten erosion rate of 23 $\mu\text{g}/\text{C}$ measured here.

Surface roughness

The surface roughness of three samples was measured before and after TA cleaning. Since the erosion takes place locally at the cathode arc spot, the surface is roughened by differential material removal. Two of the samples were polished to an initial average roughness (R_a) value of 0.08 μm while the third had an initial R_a value of 1.6 μm resulting from water jet cutting. After cleaning, the R_a values for the samples were 0.85 and 1.4 μm for those initially polished and 2.2 μm for the third sample. Fig. 3 shows the after-cleaning surface of one of the initially polished samples. The final roughness values do not represent either a maximum or minimum value achievable but are merely representative of the TA conditions chosen. In general, however, higher TA current produces higher surface temperatures and larger craters [13] resulting in a rougher surface.

Figure 3. Surface of an initially polished tungsten sample after TA cleaning.

Surface cleaning

The tungsten sample which had the carbon/deuterium film deposited on its surface was analyzed before and after cleaning to determine the impurity content. Fig. 4 shows the before and after results for carbon and Fig. 5 shows the results for deuterium. As Fig. 4 indicates, the carbon peak has been completely removed by TA cleaning. Fig. 5 indicates a similar reduction in the deuterium such that the peak is not detected after cleaning.

The mechanism for removal of surface contaminants by TA cleaning is surface ablation. The current density of cathode spots is approximately 10^8 A/cm² [14]. For a typical cathode voltage drop of 15 V this gives a power density of 1.5×10^9 W/cm². Such high power densities cause high surface temperatures and can cause severe ablation of the surface. Additionally, the high power densities produce large near-surface thermal gradients even for materials with high thermal conductivity. This large thermal gradient acts as a driving force to displace deuterium or other gases from under the sample surface to the sample surface where it can be transferred to the surrounding atmosphere.

Figure 4. Ion beam elastic recoil detection results for carbon contamination before and after TA cleaning.

Figure 5. Ion beam elastic recoil detection results for deuterium contamination before and after TA cleaning.

Summary and Conclusions

The following conclusions resulted from this study on the transferred-arc cleaning of tungsten:

- The cleaning arc voltage was found to be a statistically significant function of chamber pressure and substrate standoff distance only.
- The cleaning arc voltage for tungsten is lower than that for copper or stainless steel.
- The metal arc voltage for tungsten is higher than that for copper or stainless steel.
- The high metal arc voltage for tungsten makes spurious arcing to other surfaces in the chamber more likely.
- The large difference between the cleaning arc voltage and the metal arc voltage make cleaning without roughening easier for tungsten than for copper or stainless steel.
- The average erosion rate for TA cleaning tungsten was measured to be 23 $\mu\text{g}/\text{Coulomb}$.
- Average surface roughness values after TA cleaning ranged from 0.85 to 2.2 μm .
- The use of TA cleaning to remove a carbon/deuterium surface film was shown to be highly effective.

Future investigations planned for TA cleaning include the precision cleaning of components while introducing minimal roughening. The development of precise control methods, which are needed to achieve this goal, is planned for future research.

Acknowledgements

This work was supported jointly by the Technology Partnership Program at Los Alamos National Laboratory and the DOE Office of Fusion Energy.

References

1. G.A. Farrall, "Arc Ignition", *Handbook of Vacuum Arc Science and Technology*, R. Boxman, P. Martin, D. Sanders, Eds., Noyes Publications, 1995, p 28-72.
2. E. Muhlberger, R. Kremith, "System and Method for Plasma Coating", US Patent 4,328,257, May 4, 1982.
3. R. Henne, M. Bradke, G. Schiller, W. Schnurnberger, W. Webber, "Vacuum Plasma Spraying of Oxide Electrocatalytic Materials", Proc. of the 12th Int. Thermal Spray Conf., paper 112, 1989.

4. A. Itoh, K. Takeda, M. Itoh, M. Koga, "Pretreatment of Substrates by Using Reversed Transferred Arc in Low Pressure Plasma Spray", *Thermal Spray Research and Applications*, T.F. Bernecki, Ed., ASM International, 1991, p 245-51.
5. K. Honda, I. Chida, T. Fuse, T. Murakami, K. Kobayashi, "Effect on Reverse Transferred Arc Testing on Bonding Strength in Low Pressure Plasma Spraying" *Thermal Spray: Practical Solutions for Engineering Problems*, C.C. Berndt, Ed., ASM International, 1995, p 405-11.
6. D. Apelian, M. Paliwal, R. Smith, W. Schilling, "Melting and Solidification in Plasma Spray Deposition - Phenomenological Review", *Int. Metals Reviews*, Vol. 28, No. 5 (1983), p 276.
7. K. Hollis, R. Castro, B. Bartram, "Investigation of Transferred-Arc Cleaning for Thin Film Removal" *Proceedings of the United Thermal Spray Conference*, March 17-19, 1999, Dusseldorf, p. 202-207.
8. *Handbook of Modern Ion Beam Analysis*, Eds. Joseph R. Tesmer and Michael Nastasi, Materials Research Society, Pittsburgh, PA (1995).
9. M.P. Reece, *Proc. Instn. Electr. Eng.*, 110:793-802 (1963).
10. I.G. Brown, H. Shiraishi, *IEEE Trans. Plasma Sci.*, 18:170-171 (1990).
11. C.W. Kimblin, *J. Appl. Phys.*, 44:3074-3081 (1973).
12. V.F. Puchkarev, "Summary of Cathode Spot Phenomenology", *Handbook of Vacuum Arc Science and Technology*, R. Boxman, P. Martin, D. Sanders, Eds., Noyes Publications, 1995, p. 257.
13. B. Juttner, V. Puchkarev, E. Hantzsche, I. Beilis, "Cathode Spots", *Handbook of Vacuum Arc Science and Technology*, R. Boxman, P. Martin, D. Sanders, Eds., Noyes Publications, 1995, p 124.
14. A.E. Guile, B. Juttner, *IEEE Trans. Plasma Sci.*, v. PS-8, no. 3, p. 259-69.