Effects of pulsing frequencies on macro-particle contamination during pulsed vacuum arc deposition

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Abstract

Offering high deposition rates and being suitable for thin film fabrication, vacuum arc deposition (VAD) has been investigated extensively. However, macro-particles (MPs) emitted from the vacuum arc degrade the properties of the deposited samples and various types of magnetic filters have been proposed to mitigate macro-particle contamination. Unfortunately, the resulting deposition efficiency is inevitably compromised. In this work, we used a direct current (DC) based pulsed vacuum arc to deposit copper on a glass substrate. DC ensures continuous arc burning to achieve better stability, and the pulsed power provides high instantaneous and discontinuous energy that affects the generation of MPs. Four sets of experiments were done, and the relationship between the pulsing frequencies and MPs deposited on the substrate was studied. Our results show that the MPs tend to be smaller when the pulsing frequency increases, and the MP ratios vary with the pulsing frequencies in a different way than duty cycles. Several factors related to this phenomenon are discussed.

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1. Introduction

The technique of vacuum arc deposition (VAD) has been studied widely in recent years. It offers high deposition rate and high ionization rate of the cathode, and is well suited for thin film fabrication [1]. VAD has been applied commercially to deposit hard protective coatings on cutting and forming tools, EMI shielding materials, medical catheters, as well as decorative coatings such as gold colored TiN [1]. However, this technique is plagued by macro-particles (MPs) emitted from the source and subsequently deposited onto the samples. The generation of MPs is an intrinsic characteristic of VAD because the metal vapor generated during the deposition process is converted to liquid-state droplets which become the source of MPs [2]. Much effort has been made to avoid MP contamination and the proposed remedies include the use of a negative bias on the substrate [2,3], a shielded plate placed between the substrate and cathode [4], a magnetic filter [5], and so on. A negative bias will push away MPs that are negatively charged but the problem cannot be completely eliminated. A shielded plate can reduce MP contamination but it will also affect the temperature of the substrate and reduce the deposition rate. A magnetic filter is a very popular method, and many types of filters such as linear filters, torus filters, knee-like filters, dome filters have been designed. While MPs can be significantly reduced, the common disadvantage of this approach is the lower deposition rate [6].

A pulsed arc offers many advantages including high ionization rate [7], simpler electrode design [1], and increased velocity of the cathodic spot [8]. Pulsed arc deposition is still an area of intensive research and the influence of the pulsing parameters on the quality of the coatings has been investigated and the results have been compared to those obtained by direct current (DC) arc deposition [9]. In this work, we use the pulsed arc based on DC to deposit copper on glass substrates. DC ensures continuous, stable burning of the arc and the pulsed power provides high instantaneous energy [8] that affects the generation of MPs. The relationship between the pulsing frequency and contamination ratio of the...
MPs deposited onto the samples is studied. An image processing software package [10] is used to monitor the diameter of MPs and MP contamination ratios.

2. Experimental details

Our experiments were performed in a multi-purpose ion implanter composed of a 900 mm tall and 900 mm diameter chamber. The radius of the cathode (copper) was 80 mm, and the distance between the cathode and substrate (glass) was about 480 mm, which was fixed for each set of experiments. Since it has been pointed out that the ion current varies with the angle from the normal of the cathode plane [7,11,12], our glass substrate was placed at the maximum ion density direction. The flow rate of the background gas (argon) was constant at a pressure of $1.7 \times 10^{-1}$ Pa. A rectangular pulsed power supply was used as an arc source, and the frequency was chosen as 2.4 Hz, 27 Hz, 37 Hz, 63 Hz, and 210 Hz. The duty cycle was 15.5%, 40%, 61% and 82.7%. The typical waveform is depicted in Fig. 1. An arc was generated by the contact ignition method using a trigger electrode. The door of the vacuum chamber was kept closed for every two sets of experiments, and so contamination to the cathode could be kept to a minimum. We chose copper and glass as the cathode and substrate, respectively, because a Cu film on a glass substrate has fine surface morphology and is suitable for the observation of MPs. Before deposition, the glass substrates were cleaned ultrasonically with acetone 3 times and dried, since the surface conditions of the glass substrates can influence the results of deposition.

Scanning electron microscopy (SEM) was performed on a JSM-6460 SEM made by JEOL. The magnification was 1000 and ten regions were chosen randomly for statistical purposes.

![Fig. 1. Representative waveform of the arc (duty cycle = 15.5%; arc frequency = 209.21 Hz).](image1)

![Fig. 2. Images processed by the software: (a) original and (b) after particle recognition.](image2)

![Fig. 3. (a) Counts of MPs: 564; area fraction of MPs: 2.9% and (b) counts of MPs: 419; area fraction of MPs: 5.7%. (b) shows more serious MP contamination which has a larger area fraction value.](image3)
Fig. 4. Area fractions of MPs for different duty cycles (mean values of 10 data points; dc mean duty cycle).

Fig. 5. Critical particle diameter for different duty cycles. (Particles smaller than this diameter constitute 60% of the total MPs deposited on the films.)
The photos of the MPs were processed by an open source image processing package [10], which has the ability of adjusting the threshold of the intensity of the pictures, recognizing the particle edges, and measuring the area of each particle automatically. Two representative images before and after image processing are depicted in Fig. 2. The MPs can be easily distinguished from the copper substrate, and so the round or non-round circles shown in Fig. 2(b) are adequate in identifying the MPs.

In previous articles about MPs, many researchers chose the particle number density as the criterion to evaluate the contamination of MPs [13,14]. However, when considering the situations involving many small particles (depicted in Fig. 3a) with the other case with few big particles (Fig. 3b), there may be confusion because the first one might induce a higher particle density. Therefore, we have chosen the area fraction covered by the MPs ($f_{MPs}$) as the main criterion, with the amounts of particles (equal to particle density, since the total area is constant) assisting in our interpretation. The parameter $f_{MPs}$ is defined as:

$$f_{MPs} = \frac{A_{MPs}}{A_{total}}$$

where $A_{MPs}$ is the area of all MPs in a photo and $A_{total}$ is the total area.

3. Results

All the images acquired from the ten regions are processed, and the average $f_{MPs}$ of each frequency is calculated. The results are shown in Fig. 4. The line in each panel is linearly fitted using 5 points with the error as weight. It can be concluded that $f_{MPs}$ tends to decrease when the frequency increases. This trend is violated when the duty cycle is 40%. It should be pointed out that from the fitted line, the data acquired from a duty cycle 82.7% do not obey this trend perfectly, but from the scattered data points, the trend can be reasonably assumed.

The diameter of every particle is tracked and sorted in an ascending order. There exists a critical diameter. Particles smaller than that constitute 60% of the total MPs. This critical diameter is plotted in Fig. 5. It is clear that the size of the MPs tends to decrease when the frequency is increased. The linear fitted line shows a generally increasing trend for a duty cycle of 40%, although some data scattering is observed.

Fig. 6 shows the relationship between the frequency and amount of MPs determined from the ten measured regions. This plot is similar to that of the particle number density and the trend resembles that displayed in Fig. 4.

4. Discussion

Considering the formation of liquid droplets preceding the formation of MPs, some good information can be obtained. When an arc begins to burn on the surface of the cathode, very small cathodic spots migrate randomly on the surface [15]. At every place the spot passes, the cathode metal is evaporated and the vapor may gather at the vicinity of the cathode. If there is no other force to disperse the vapor, liquid droplets will form and fly towards the substrate along with the plasma flux to form MPs. Hence, the density of the metal vapor plays a key role on the formation of MPs [16].

For a pulsed arc source, the arc burns only when the voltage is high. During the rest of the time, the arc may be extinguished...
due to the absence of a high power input. For one arc burning and extinguishing cycle, when the frequency is increased, the time of arc burning diminishes. This will lead to more small “evaporating pool”, but not less big ones. Therefore, the density of the vapor that may gather to form liquid droplets will tend to be small, thereby producing more small diameter MPs, as indicated by the trend revealed in Fig. 5. Since $f_{ MPs}$ is strongly related to the diameter distribution and amount of MPs, the trend of Fig. 4 can be explained using the results exhibited in Figs. 5 and 6. If only one considers the change in the critical diameter, MP contamination should be milder when the frequency is increased under all duty cycles. However, as shown in Fig. 6, the amounts of MPs increase for duty cycles of 40% and 82.7%. Therefore, $f_{ MPs}$ increases for these two duty cycles (Fig. 4).

The absolute differences between the minimum and maximum values of $f_{ MPs}$ at different duty cycles can be found in Fig. 4. When the duty cycle is equal to or greater than 40%, there is only a small difference (about 0.15) among the values of $f_{ MPs}$ at different frequencies. Furthermore, the difference in the surface morphology with different frequencies is also very small. As we know, with increasing duty cycles, the input power increases. Since the power of the arc source influences the shape and velocity of the cathodic spots [15], it will also influence the metal vapor. If the power is increased, a larger amount of the metal will be evaporated, and it is easier to produce a larger number and bigger liquid droplets. When this phenomenon becomes dominant, the aforementioned factor about decreasing size of droplets will play a more limited role. Hence, it is observed that for higher duty cycles, there is a smaller difference in the surface morphology of the MPs at different frequencies.

5. Conclusion

Experiments on pulsed vacuum arc deposition were conducted and the surface images of the films were processed by a software package. The results show influences on the MP diameter and contamination ratio by the pulsing frequencies. With increasing frequencies, the size of the MPs tends to decrease in our experiments. Nevertheless, the MP contamination ratio decreases when the duty cycle is 15.5% and 61%, increases when it is 40% and 82.7%.

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