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THE SIZE DISTRIBUTION OF METAL CLUSTERS PRODUCED IN PLASMA-DISCHARGE HOLLOW-CATHODE SOURCE

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Abstract – *The mean size of Cu clusters produced in a hollow-cathode, dc plasma-discharge source, increases significantly with decreasing flow rate and less pronouncedly with increasing Ar/He abundance ratio and increasing discharge current. The effect of pressure is mixed. Between 0.4 and 0.6 mbar the size is not affected. Above 0.6 mbar, the size increases significantly with increasing pressure. With the help of the above parameters, the mean cluster size can be shifted at will between 20 and 250 Å. Implications concerning the mechanisms associated with clustering in plasma are discussed.* ©*1999 Acta Metallurgica Inc.*

INTRODUCTION

The full utilization of a plasma generating cluster source (such as the magnetron, the pulsed-arc and the hollow-cathode source) is prevented by the insufficient understanding of clustering in plasma. On the other hand, the influence of various source parameters (including temperature, pressure, velocity and type of the gas and dimensions of the condensation cell) on clustering of neutral particles produced in an oven-heated or supersonic-expansion source has been well studied over the years (1). In the present study the size effect of some of the above classical parameters is investigated, utilizing a plasma generating, hollow-cathode source (2), where (in addition to neutral) positive and negative metal particles coexist during the time of cluster formation (3). An other important difference between an oven-heated and a plasma-discharge source is that in the second the initial concentration of metal particles is not an independent variable, since pressure, velocity and type of the inert gas affect not only the clustering but also the sputtering rate. The purpose of the study is to observe whether and/or to what extent these, well known, parameters are also relevant to clustering in plasma. The only previous commensurate data of which we are aware have to do with geometry and temperature effects studied in a magnetron source (4).

EXPERIMENTAL

A detailed description and performance of the hollow-cathode source used has been already presented (2). Briefly, metal atoms are sputtered from the inner walls of a hollow cathode under a dc plasma discharge. The discharge is operated under a forced Ar flow at about 1 mbar pressure, leading to the aggregation of atoms to clusters. The clusters are swept by the gas stream through a nozzle, fly inertly through a skimmer and are deposited on carbon–

coated TEM grids located about 50 cm away from the cathode. The deposited clusters are transferred in the air and examined with a Philips CM20 TEM operating at 200 kV.

RESULTS AND DISCUSSION

In order to observe the effect of flow rate, two different pumps were used. Figure 1 shows that with the smaller pump the mean cluster size and the width of the size distribution (110 ± 22 Å) are about four times larger compared to those obtained with the larger pump (32 ± 4 Å). The parameters affected by pumping speed are interaction times and sputtering rates. The sputtering caused by the larger pump (9.2×10^{16} atoms sec^{-1}) was about three times smaller than that caused by the smaller (29.0×10^{16} atoms sec^{-1}). Therefore, sputtering rates and interaction times are both partly responsible for the mean size seen in Figure 1.

Figure 2 demonstrates the effect of inert gas pressure on cluster size and sputtering rates. For pressure between 0.4 and 0.6 mbar the cluster size remains the same within the experimental error. Considering that at 0.6 mbar the sputtering rate is by far the highest, these data suggest that for equal number of initial Cu atoms the cluster size would significantly decrease between 0.4 and 0.6 mbar, in contradistinction to the expected trend (1). It is usually assumed that a high gas pressure facilitates coagulation due to better cooling. In the present case, however, certain plasma parameters will also be affected. For instance, the changing pressure will change the electron temperature, thus affecting coagulation and cluster size, as it has been demonstrated by Monte Carlo calculations (5).

The present size distributions were found to follow the lognormal distribution (6), a feature also seen in magnetron clustering (7). Results associated with the three specimens of Figure 2 are shown in Figure 3. Lognormal behavior has been identified exclusively in clustering of neutral particles (6). The reappearance of the same distribution in plasma clustering where charged particles are involved, constitutes an experimental evidence in favor of the theoretical contention that lognormal size distribution is a universal feature of clustering, irrespectively of the particular mechanisms involved (8).

The next source parameter investigated was the discharge current. In general, an increase in discharge current caused an increase in cluster size. For instance, the mean cluster

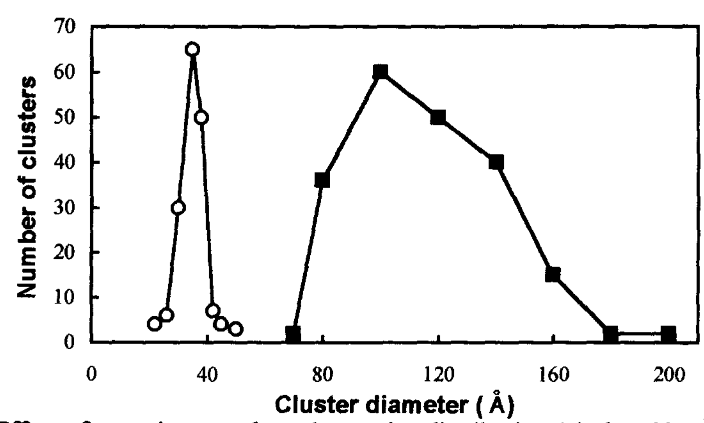

Figure 1. Effect of pumping speed on cluster size distribution (circles: 50 m^3/h; squares: 15 m^3/h)

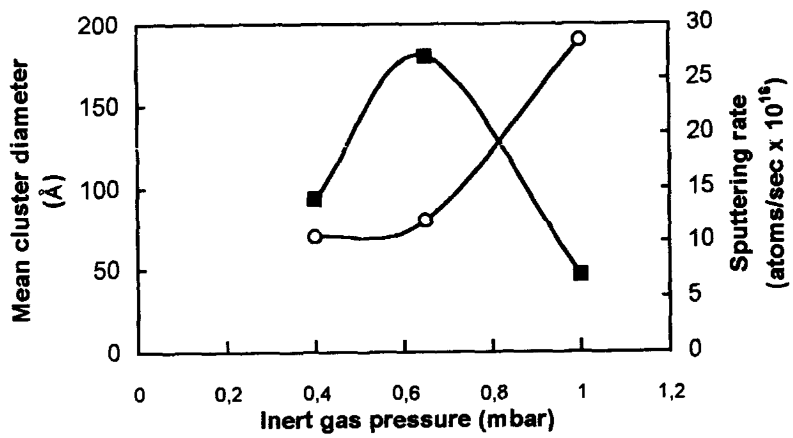

Figure 2. Effect of gas pressure on cluster mean size (circles) and sputtering rate (squares).

diameter increases from 40 Å at 200 mA, to about 60 and 75 Å at 300 and 400 mA, respectively. However, the results are not easily reproducible when big current steps are used. All these seem manifestations of conditioning instabilities known, for instance, to be associated with big voltage changes in particle accelerators.

The last parameter tested was the composition of the inert gas. It was found, for instance, that a mean cluster diameter of 87 Å obtained with a mixture of one part Ar and two parts He, increased to 118 Å when that mixture was replaced by pure Ar. Qualitatively, this is a typical effect of the gas type on size (1).

CONCLUSIONS

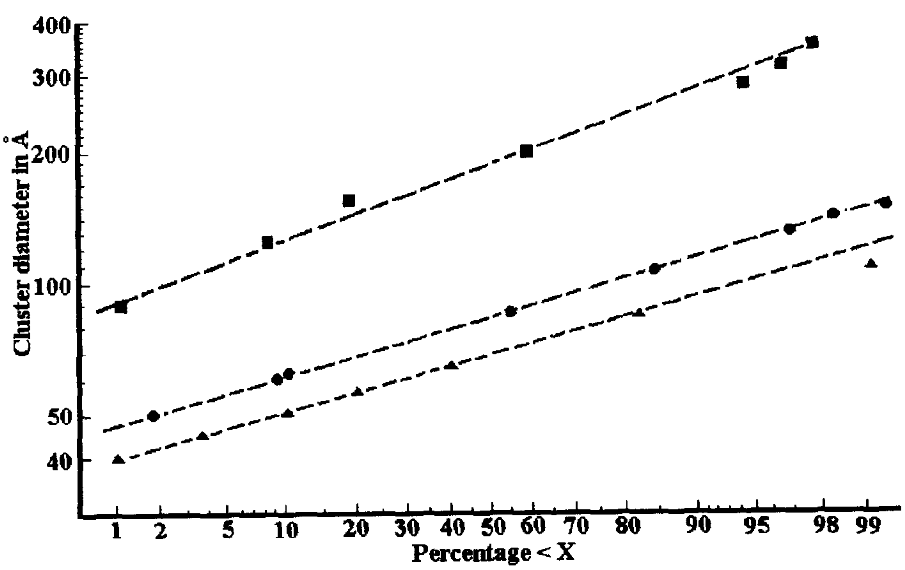

Figure 3. Log normal probability plot of size distribution of the specimens shown in fig. 2. (triangles: 0.4 mbar, circles: 0.65 mbar and squares: 1.0 mbar)

In a plasma-discharge source negatively and positively ionized metal particles coexist during the time of coagulation (3), suggesting that electrostatic interactions should be, at least partially, responsible for clustering in plasma. Nevertheless, the effects of the classical coagulation parameters investigated here (with the exception of pressure) were found, at least qualitatively, in agreement with previous results associated predominantly with clustering of neutral particles (1). Two, more or less mutually exclusive, interpretations seem appropriate. Either the presence of the plasma modifies the interactions between ionized particles to such a large extend that finally they cluster the same way with neutrals; or the examined coagulation parameters are associated with universal size effects, which do not depend on the particular clustering mechanisms involved. We have already intimated that this is the case with the lognormal size distribution, which seems to constitute such a universal property. The effect of flow rate on cluster size must be also universal, because the flow rate determines the interaction time (the later being inversely proportional to the former), which in turn determines the cluster size (the later being proportional to the interaction time). However, the effect of pressure on cluster size presently identified at low pressure, seems to be different in a plasma-discharge source than in an oven-heated source. It should be finally noted that due to the interplay between sputtering and clustering in a plasma-discharge source, a direct quantitative comparison of that source with an oven-heated source demands high quality data, associated with very clearly determined source conditions, which do not seem to be presently available in the literature. In any event, a theory attempting to describe clustering in a plasma source should comply with the size effects presently identified. It should be noted that in a very recent Monte Carlo simulation of clustering in plasma, a lognormal size distribution as well as an effect of pressure on clustering characteristic to the plasma discharge, due to induced plasma electron temperature differences, have been explicitly identified (5).

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