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Annual Technical Progress Report - II

"In-Situ" Electron Microscope Investigation of the Nucleation
and Growth of Sputtered Thin Films

MASTER

by

Thos. E. Hutchinson and Dudley Sherman

University of Minnesota
School of Mineral and Metallurgical Engineering

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the instruments at 125 kilovolts have indicated that these design goals have been reached in the present model.

IV. ULTRA-HIGH VACUUM STAGE

The ultra-high vacuum stage has been received from Varian Assoc. The Electro Ion pump purchased by this contract has been received and tested on a comparable ultra-high vacuum system for approximately 50 hours and has shown excellent pumping characteristics. Vacuums of the order of 2×10^{-8} torr have been realized in these tests. The ultra-high vacuum pump is presently being fitted to the high vacuum, high temperature specimen stage in order to allow investigations of the nucleation and growth of sputtered atoms on a variety of substrates at temperatures up to 600°C .

V. NUCLEATION AND GROWTH OF COPPER ON AMORPHOUS CARBON SUBSTRATES

Data have been obtained on the nucleation and growth of copper deposited by inert gas ion sputtering on amorphous carbon substrates at room temperature and at 350°C . The amorphous carbon substrates were prepared in a modular high vacuum unit operating at 10^{-6} torr by depositing carbon on clean glass slides to approximately 200 angstroms. The carbon thin films were then removed from the substrates in distilled water and placed on electron microscope grids. Depositions were carried out at 5 K.V. and an ion beam current at $250 \mu\text{A}$.

The results of sputter deposition at 350° during observation in the electron microscope are shown in Figure 1 - 11, and the number density of nuclei is shown in Table I. Several observations of significance can be made

from these data. It seems likely that the initial nuclei shown in Figure 1 result from activated state nucleation possibly due to foreign material contaminants existing on the substrate initially. These initial nuclei are certainly larger than would be expected on the basis of non activated state nucleation. The secondary nuclei observed in Figure 3 and following, are more indicative of non activated state nucleation. The growth of these nuclei follows a relatively normal pattern, seen previously in vacuum vapor deposition. The number density, however, is somewhat higher than that observed in vacuum-vapor deposition. These nuclei show less crystallographic faceting in the early stages than similar nuclei obtained by vacuum vapor deposition. This indicates a higher energy surface bond between the nuclei and the substrate. They do not develop significant crystallographic faceting until a monolayer coverage of approximately 2 is reached.

The two electron diffraction patterns, Figure 8 and 11, were taken at approximately one ~~monolayer~~ coverage and five monolayer coverage respectively. The presence of a strong (220) reflection in the one monolayer diffraction pattern indicates a preferred orientation of the original nuclei (i.e. activated state nuclei), having a [111] axis perpendicular to the substrate. This is much less pronounced in the five monolayer coverage indicating a relatively random orientation of the secondary nuclei. Several examples can be seen in the figures of nuclei which are near coalescence. No attempt, nowever, has yet been made to determine the mechanism of coalescence.

VI. TECHNIQUE FOR RAPID DETERMINATION OF NUCLEUS DENSITY AND ORIENTATION

A technique developed by D. B. Dove ⁽¹⁾ has been adapted for use in thin film nucleus density and orientation studies. The technique employs a greatly magnified electron diffraction pattern formed only with elastically scattered electrons. The theory of this technique is based on the correlation diffraction effects between individual nuclei at spacings up to approximately 200 angstroms. The lens settings of the electron microscope are usually adjusted to conveniently reveal diffraction effects of 5 angstroms and less. In the diffraction patterns of Figure 8 and 11, the (220) ring diameter plane is roughly 5 centimeters. Since the diffraction pattern is a picture of reciprocal space, larger spacings than this will be closer to the undeviated central spot. It is necessary therefore to magnify the image by approximately 10 in order to obtain significant diffraction separation for particles on the order of 200 angstroms mean separation. Features in the area close to the center spot are generally masked by the low angle inelastically scattered electrons having lower energy than the elastically diffracted electrons. It is therefore necessary to reduce this contribution. This is done, in the system designed in this laboratory, by a series of mesh grids at potentials

(1) D. B. Dove, private communication.

slightly less than the primary beam energy. Electrons having energies less than these grids are thereby repelled and not allowed to reach the imaging system. Electrons having energy higher than this are reaccelerated and are recorded. Experimental details and initial testing of this instrument will be presented in a separate report in preparation. The correlation function between nuclei (similar to a radial distribution function) will be recorded directly with respect to nucleus separation. Theoretical studies of D. B. Dove have allowed determination of particle size in agglomerate systems of thinned bulk material. This will be used to determine the nucleus size as well as the distribution.

VII. GRADUATE STUDENT PROGRESS

Mr. D. M. Sherman has been the graduate student primarily responsible for the bulk of the experimental work in this project. His progress in this area has been quite good, as the results would indicate from the earlier sections of this Progress Report. He has completed all necessary requirements for the Ph. D. with the exception of completion of the thesis work and satisfaction of the final examination. He is most likely to complete this in September or October of this year.

Mr. J. S. Maa and Mr. C. C. Lin have been added to the graduate student research staff in order to more rapidly complete the work outlined in this and the previous proposal. Both of these men have shown significant diligence and initiative and seem likely to develop into excellent graduate students.

VIII. CONCLUSION

The work done in the past 12 months has demonstrated the technique of insitu sputtering by inert gas ion bombardment of targets inside the electron microscope is feasible. Data have been obtained on the nucleation and growth characteristics of sputtered copper formed at room temperature and 350°C on amorphous carbon. The data indicate activated state nucleation as well as non activated state nucleation. Initial nuclei show strong $[111]$ preferred orientation perpendicular to the substrate, whereas nuclei composing a five monolayer film demonstrate random orientation. Further data will be taken on the system copper on amorphous carbon, the results of which will be correlated with theories of nucleation and mechanisms of coalescence.

IX. ACKNOWLEDGEMENTS

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TABLE 1

<u>Picture No.</u>	<u>Monolayer Coverage</u>	<u>No. of Nuclei/cm²</u>
1	0.25	4×10^8
2	0.50	4×10^8
3	0.65	1×10^{10}
4	1.00	7×10^9
5	1.25	7×10^9
6	1.25	2×10^{10}
7	1.50	2×10^{10}
9	2.20	2×10^{10}
10	5.25	2×10^{10}

Figures 1 - 11 are electron micrographs and diffraction patterns of copper sputtered on amorphous carbon at 350°C. The magnifications are noted below.

Figure	1.	36,000
	2.	36,000
	3.	36,000
	4.	93,000
	5.	93,000
	6.	93,000
	7.	220,000
	9.	93,000
	10.	220,000

Fig. 1

Fig.2

Fig. 3

Fig.4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

Fig. 9

Fig.10

Fig. 11