Journal of Ceramic Processing Research. Vol. 4, No. 3, pp. 115~117 (2003)

J O U R N A L O F
Ceramic Processing Research

TEM-examination and computer simulation of nano-scale multilayers by pulsed cathodic arc deposition

Sung-Yong Chun*
Department of Advanced Materials Science and Engineering, Mokpo National University, 61, Dorim-ri, Chungkye-myon, Muan-Gun, Chonnam 534-729, Korea

Nanometer scale multilayers have been fabricated by pulsed double-cathodic arc deposition with varying thickness of each layer from a few to nanometers and examined by cross-sectional transmission electron microscopy. Computer simulations were performed to obtain a better understanding at least of the nano-scale deposition of multilayers during the cathodic arc deposition process and for comparison with the experimental results. The experimental results are in good agreement with those of simulations suggesting that the limit of the bilayer period of the ultra-thin multilayers is about 3 nm.

Key words: pulsed cathodic arc, multilayers and TRIDYN.

Introduction

In modern materials science, nano-scale multilayers have achieved increased importance. Such multilayers show physical properties and performances in a variety of fields such as in magnetic superlattices, memory devices and biosensing applications [1-3]. Another increasing demand for multilayers is the superlattice structured hard coatings [4]. In the cathodic arc process, a highly ionized metallic plasma induces specific structures associated with the composition of the individual layer. In some cases, the situation is especially complicated as the formation of films by CAD is controlled not only by the ballistic processes, but also by possible chemical or diffusion processes [5]. Experimental investigations of the films formed cannot always give complete information about the mechanisms governing their composition, structure and the mechanism of the film growth. Comparison of the experimental approach with computer simulation of the deposition process offers the possibility of gaining more insight into the physical procedure of the film growth.

In the present study, the formation and growth of nano-scale Fe/C multilayers have been studied by cross-sectional high-resolution transmission electron microscopy (HR-TEM) and computer simulated with the TRIDYN code.

Experimental

The disadvantage of the conventional cathodic arc methods is macro-particle formation in the growing films, which deteriorates the quality of the deposited film. Contamination of the films by macro-particles represents a major obstacle to fabricate nano-scale multilayers. Attempts have been made to solve this problem by removing the macro-particles by filters [6]. During deposition of Fe/C multilayers, the Fe and C ionized cathodic arcs were alternately switched on and off, corresponding with the Fe and C layer deposition period respectively. The dose rates of the simulation in this study was in the range 5×10^{15} to 2×10^{16} ions/cm^2. The ion energy of carbon and iron was 50 eV and 90 eV (mean ion charge state of vacuum-arc-produced iron is 1.8+), respectively. Fe/C multilayers were produced with four different sets of periodic thickness i.e. the thickness of the Fe and C bilayers. The microstructures of the multilayered films formed are examined by cross-sectional HR-TEM.

*Corresponding author:
Tel : +82-61-450-2495
Fax: +82-61-450-2490
E-mail: sychun@mokpo.ac.kr

Results and Discussion

Film formation

Figure 1 shows a schematic diagram of the pulsed vacuum arc apparatus. A cathodic arc plasma source consisting of two 8-mm diameter graphite and iron cathodes and a cylindrical anode was used for the formation of carbon and iron plasmas. The source operated in a pulsed mode with a pulse duration of 25 μs, a repetition rate of 1 Hz, and an arc current peak value of 1000 A. A newly-developed plasma source and macro-particle filter are described in detail elsewhere [7, 8]. The Si(100) substrate was placed within the chamber which was then evacuated to 5×10^{-6} Pa and exposed to the metal plasma at room temperature.

The measured deposition rates of Fe and C were 0.1

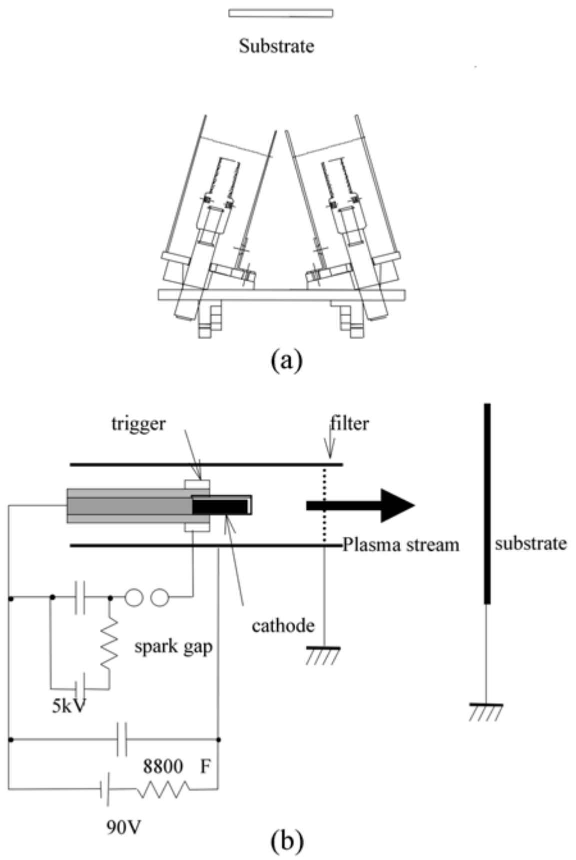

Fig. 1. Schematic illustration of (a) the double-cathode and (b) the pulsed cathodic arc deposition used for Fe/C multilayers.

and 0.05 nm/pulse, respectively. The coating thickness could be controlled from 0.1 nm to a few hundred nanometers by increasing the number of arc pulses.

Cross-sectional HR-TEM

Figure 2 shows the structure of Fe/C multilayers with four different layer thickness in the range between a fraction and a few nanometers. The thickness of the Fe/C layers was varied. For the first, it was 5 bilayers with a bilayer thickness ratio of 0.75 nm, for the second, it was 5 bilayers with a ratio of 1.25 nm, for the third, it was 5 bilayers with a ratio of 2.5 nm, and the fourth, it was 5 bilayers with a ratio of 5.0 nm. The bilayer thickness refers to the thickness (nm) of one Fe/C layer. There are two distinct structures, in which Fe and C are mixed, while in the other the Fe and C layers are distinct. Thus, no multilayer structure is observed by TEM for the bilayers with a period less than 1.25 nm: the film is apparently intermixed.

Based upon these results, the thickness of the thinnest bilayer produced by our pulsed vacuum arc deposition would be ~2.5 nm.

Computer simulation

The dynamic binary collision computer code TRIDYN [9-11] was employed for the simulations of the cathodic arc deposition process. TRIDYN allows us to simulate

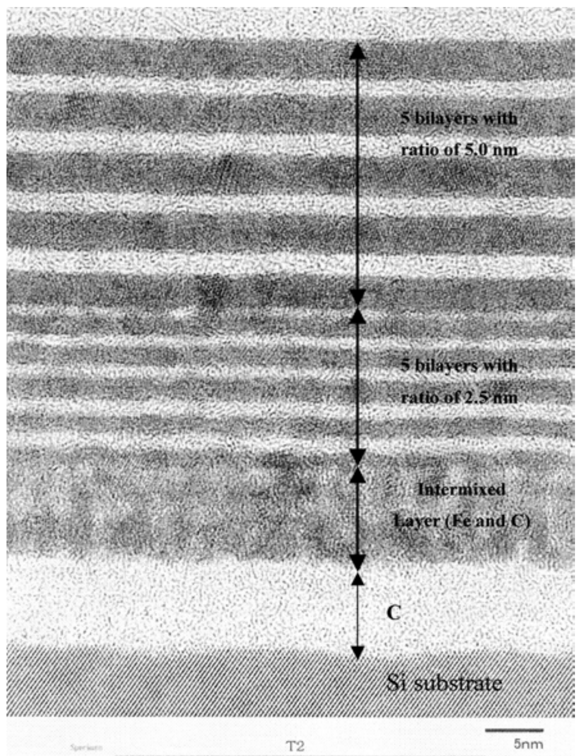

Fig. 2. High-resolution TEM micrograph of Fe/C multilayers with 4 bilayer periods: 0.75 nm, 1.25 nm, 2.5 nm and 5.0 nm in order from the Si substrate, respectively.

dynamic changes of thickness and/or composition of multicomponent targets during high-dose ion implantation or vacuum arc deposition. It is possible to consider homogeneous as well as inhomogeneous targets. In the present case, two cathodic arcs consist of Fe and C components. The fluences also correspond to the experimental conditions. The values for the surface binding energy of iron and carbon were assumed to be the same as in pure Fe and C targets. The values of surface binding energy and the atomic volume were assumed to be the same as in pure Fe and C targets. This is really a simplified assumption. Please note that 4.34 eV is the surface binding energy of Fe, 7.41 eV for C. The volume that carbon and iron atoms occupy in the silicon substrate is chosen as for a pure silicon target.

Figure 3 shows the result of the simulation for the Fe/C multilayered films with a carbon ion energy of 50 eV and an iron ion energy of 90 eV deposition on a silicon target with four different influences. The distributions of C and Fe across one Fe/C bilayer, together with their distributions in the silicon target, are displayed in the figure. It is seen that the Fe/C bilayers are fabricated on the silicon target in Fig. 2(a) and (b) with a bilayer thickness of 4.8 nm and 3.7 nm, respectively. Thus, for fluences higher than 1.0×10^{16} ions/cm^2, periodic Fe/C multilayers were fabricated, which is consistent with the TEM measurements. However, the periodicity of the Fe/C bilayer deteriorates as the thickness of the bilayer decreases to smaller than about

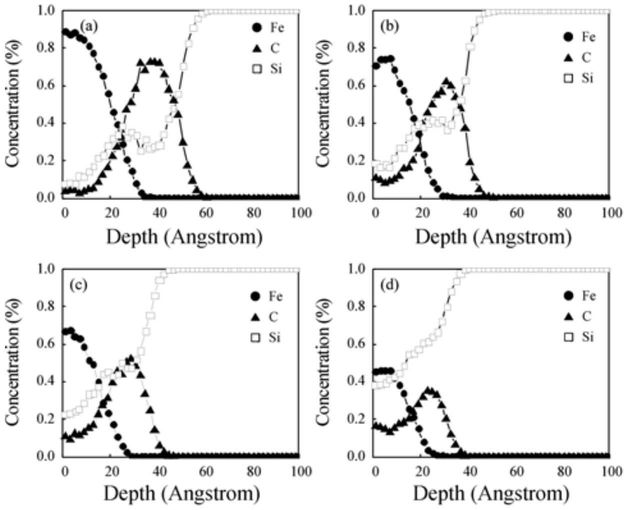

Fig. 3. Limitation of thickness profiles of Fe/C multilayers on Si(100) substrate with 4 fluences by TRIDYN: (a) 1.5×10^{16} ions/cm^2, (b) 1.0×10^{16} ions/cm^2, (c) 8×10^{15} ions/cm^2 and (d) 5×10^{15} ions/cm^2, The corresponding deposition parameters are $E_C = 50$ eV and $E_{Fe} = 90$ eV.

3 nm as shown in Fig. 2(c) and (d). The presence of the carbon distribution on the surface of the silicon target indicates that the carbon and iron layers are so thin that they are completely intermixed. Consequently no periodic structure exists in these multilayers. The influence limit of the simulation of Fe/C multilayers by cathodic arc deposition is approximately 1.0×10^{16} ions/cm^2, which is about 3 nm in thickness.

We therefore believe that the TRIDYN program is a useful tool for studying new unexplored deposition of nano-scale multilayers using cathodic arc deposition.

Conclusions

The computer simulation using a TRIDYN code was performed to compare with the experimental results. For the higher ion fluence (>1.0×10^{16} ions/cm^2) and larger bilayer thickness (>3 nm), the periodicity of multilayers was good. However, the results of the simulations for the lower ion flux and smaller bilayer thickness reveals that each individual layer is intermixed and diffused. The experimental results are in good agreement with those of simulations in that the limit of bilayer period of the ultra-thin multilayers is about 3 nm. The results further indicate that the TRIDYN code can be used for a wide range of applications, from implantation to deposition and may be used for film thickness and composition estimation.

Acknowledgements

This work was sponsored in part by the Basic Research Program in Korea Science and Engineering Foundation under grant No. R08-2003-000-10440-0 and in part by the grant from the Mokpo National University.

References

1. M. Kläui, P.A. Lewis, C.A.F. Vaz, A. Bleloch, R. Speaks, M.C. Blamire, J.A.C. Bland and H. Ahmed, Microelectronic Engineering 61-62 (2002) 593-600.
2. T. Ruckert, W. Keune, W. Sturhahn and E.E. Alp, Journal of Magnetism and Magnetic Materials 240 (2002) 562-564.
3. Selena Chan, Yi Li, Lewis J. Rothberg, Benjamin L. Miller and Philippe M. Fauchet, Materials Science and Engineering: C15 (2001) 277-282.
4. W.D. Sproul, Science 273 (1996) 889-892.
5. W. Moeller, W. Eckstein and J.P. Biersack, Computer Physics Communications 51[3] (1988) 355-368.
6. A. Anders, S. Anders and I.G. Brown, Plasma Sources Sci. Technol. 4 (1995) 1-12.
7. S.Y. Chun and A. Chayahara, Mater. Trans. JIM 41 (2000) 44-46.
8. S.Y. Chun and A. Chayahara, Appl. Surf. Sci. 169/170 (2001) 607-611.
9. W. Moeller and W. Eckstein, Comput. Phys. Commun. 51 (1988) 355-368.
10. J.P. Biersack, S. Berg and C. Nender, Nucl. Instr. and Methods in Phys. Res. Sec. B59/60 (1991) 21-27.
11. M. Posselt and K.-H. Heinig, Nucl. Instr. and Methods in Phys. Res. Sec. B102 (1995) 236-241.