The Role of Radical and Cluster in Thermal Plasma Flash Evaporation Process

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The plasma flash evaporation method developed by our group is one of the most promising candidates for the large-area practical processing of multi-component fine films, in which mixed fine powders of constituents are continuously injected into a thermal plasma to be co-evaporated completely and co-deposited onto substrates. This novel processing has some interesting characteristics such as high-rate deposition under various reactive soft-vacuum (about 200 Torr) atomospheres. We have applied it to the deposition of high Tc superconducting oxide and have obtained excellent ($Tc = 92K$, $Jc = 3\times10^5$ A/cm^2) $YBa_2Cu_3O_{7-x}$(YBCO) films on $SrTiO_3$ (100) with relatively large-area of 7×7 cm^2 . The prominent features may be caused by high net flux of atomic oxygen and cluster deposition mechanism. In order to clarify such features quantitatively, the atomic oxygen flux (Γ_o) and cluster size (d) was measured by the following procedure. For measuring Γ_o, QCM (quartz crystal microbalance) covered with Ag thin films were placed at plasma tail flame and exposed for 10ms. Ag is mainly oxidized by atomic oxygen, and Γ_o can be deduced from the small weight change (around 1 μg/cm^2) detected by QCM. The dependences of Γ_o in 200 Torr RF thermal Ar–O_2 plasma on substrate position and RF input power were measured, and the maximum value of Γ_o was estimated to be more than 10^{19} atoms/sec cm^2 corresponding to the 7 % frozen atomic oxygen. On the other hand, in order to estimate d, the Si wafer with sub-micron trenches was put on the substrate holder. The cluster size d can be described as a function of diffusion coefficient D and sticking probability η. These D, η, d were determined by observing film thichness feature in the trenches. The cluster size of YBCO was estimated to be about 0.6 to 10 nm. Allowing few nm cluster for deposition species under high atomic oxygen flux should lead us to following advantages. (1) Cluster deposition from high vapor concentration results in high rate deposition. (2) Deposition of multi-component films with the same composition of the fed powders without difficulty. (3) High rate deposition because of the increase of the sticking probability of clusters even if relatively high substrate temperature is used. (4) Under higher substrate temperature condition, the critical size for 2D surface nucleation increases resulting in good epitaxy. (5)Deposition under high radical flux leading to promote effective chemical reaction. Actually, we have demonstrated extremely high rate deposition (around 2.2 μm/min) of fine epitaxial YBCO films with $Tc = 90K$ on $SrTiO_3$(100) from cluster of 0.8nm diameter.