論 文 の 要 旨

題　目　In-flight Coating of Carbon Nanotubes by Plasma-Enhanced Chemical Vapor Deposition Process
（プラズマ CVD プロセスによるカーボンナノチューブの浮遊コーティング）

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Plasma-enhanced chemical vapor deposition (PECVD) processes are used for the preparation of micro and nano structures. The very fast processing time, flexibility for wide substrates and new morphologies are the attractions of PECVD process. Metal oxides and thin films have been successfully prepared by PECVD process. Coatings on carbon nanotubes (CNTs) by PECVD is focused due to the uniform and complete coatings on the surface. Coatings on CNTs are important for specific applications, involving light structures and enhanced activity. In this dissertation, PECVD is used for the in-flight coating of metal oxides and polymers on CNTs.

First, the effect of process conditions on coatings of titania (TiO_2) coated CNTs by PECVD was studied. Aerosols of CNTs and vapor of titanium tetraisopropoxide were simultaneously fed into a plasma zone. In-flight conditions of CNTs were maintained by means of carrier gas. The resulting products were collected and analyzed. The effects of process parameters on the morphology of the coated layers were studied with the variation of process conditions. The concentration of coating precursor and pressure of the system was varied at three different conditions. At higher values, the thickness and surface roughness of the layers increased. The increase of residence time in the plasma zone changed the morphology of layers from sparse and granular to thick and continuous ones. Role of plasma input power on morphology was also evaluated. Higher plasma input power with unchanged flow conditions, changed the morphology of layers from fine to aggregates. These results show that the morphology of coating layers can be controlled by the process parameters.

Next, the crystallization of TiO_2 coating layers of structures prepared by PECVD was investigated. Coating precursors and CNTs at in-flight conditions were subjected to plasma exposure. TiO_2 layers of typical thickness of ~140 nm on the surface of CNT was obtained. Obtained layers were amorphous in nature. Elemental analysis by EDS showed that the layers consisted of the TiO_2. The layers were converted to anatase TiO_2 by annealing at 450 ºC for 12 h in air. Annealing at 600 ºC in air showed the loss of CNTs with damages in the outer layer containing TiO_2. Annealing the structures in N_2 atmosphere at 900 ºC showed that the products retained CNTs with coating of rutile phase. Morphological changes were observed when the annealing was carried out first in N_2 and then in air at 700 ºC. This operation caused the crystallization and densification of the coating layer. CNT played the role of template and hollow TiO_2 nanotubes were obtained. The detailed evaluations of the hollow nanotubes showed that these had uniform structures of TiO_2 with anatase and rutile phase.

The in-flight coating of polymers on CNTs by PECVD was also experimented**.** Suitability of PECVD process for the preparation of the polymers from monomers and subsequent coating on CNTs was evaluated. Monomers of methyl methacrylate in the presence of triethyl amine was vaporized and fed simultaneously with

CNTs for the plasma exposure. Chemical analysis of the resulting products indicated the functional groups of polymethyl methacrylate (PMMA) in the coated structures. Complete and uniform coatings on undestructed CNTs were obtained. Coating layers with thickness of 150-350 nm were obtained. Higher feed rate of precursors resulted in rougher coatings, while smooth coatings were obtained at low feeding rates. The obtained products consisting of PMMA on CNTs suggest the applicability of PECVD process for polymeric coatings.

Finally, the summary of all findings was highlighted and scope for further investigations of coatings by PECVD was suggested.