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## 主 論 文 の 要 旨

論文題目 Study on synthesis processes and characterization of tin and carbon nanocomposites  
(錫と炭素のナノ複合材料の合成プロセスと特性評価に関する研究)

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### 論 文 内 容 の 要 旨

In this dissertation, from the viewpoint that the plasma processes are advantageous to surface modification and nanomaterial synthesis, the surface modification of carbon films and tin oxide-graphene (SnO<sub>2</sub>-graphene) nanocomposite formation was clarified. It was realized that SnO<sub>2</sub>-graphene composite can be synthesized at the relatively low temperature and atmospheric pressure in a single step by in-liquid plasma. Upon studying gas sensing response of SnO<sub>2</sub>-polyaniline (Sn-PANI) composite, it was suggested that the composite has a better gas sensing response at room temperature than its component materials due to an overall increase in surface area for gas adsorption and formation of a p-n junction between p-type PANI and n-type SnO<sub>2</sub>. The chapter wise summary has been summarized below.

#### Chapter 1: Introduction

In chapter 1, the structures, properties and point of view for application development of metal oxides (SnO<sub>2</sub>), conducting polymers (PANI), carbon materials

(graphene) and composite based on SnO<sub>2</sub>, polyaniline, and graphene are described. The need for composites and problems associated with traditional composite formation approaches are summarized. It has been pointed out that the traditional graphene synthesis methods are not suitable for graphene-based composite synthesis, the in-liquid plasma assisted graphene synthesis can be superior to the composite for the graphene-based composite synthesis.

### **Chapter 2: Experimental setup and evaluation methods**

In chapter 2, the synthesis methods for SnO<sub>2</sub>, PANI, carbon films and graphene were described. The atmospheric pressure plasma surface treatment process and apparatus were also described. Various material evaluation methods were discussed along with their operational principle. The gas sensing measurement set up and sample preparation for gas sensing measurement has been shown and discussed.

### **Chapter 3: Ex-situ binding of Sn-PANI composites – Synthesis and gas sensing properties**

In chapter 3, the sensing properties of Sn-PANI composite synthesized by ex-situ approach has been shown. The composite was synthesized by mixing SnO<sub>2</sub> and PANI powders and grinding together. The surface morphological, crystal structure and chemical composition studies showed the mutual presence of SnO<sub>2</sub> and PANI in a composite sample. The composite showed improvement of ammonia gas and humidity sensing response at room temperature. The synergetic effect due to the addition of SnO<sub>2</sub> improved gas sensing characteristics in the case of the composite.

### **Chapter 4: Surface and bulk modifications of magnetron-sputtered carbon films by post treatments of atmospheric pressure plasma**

In chapter 4, surface and bulk modification of magnetron sputtered carbon films were achieved by the atmospheric pressure plasma (APP) treatment and controlling

process parameters during deposition. A relationship between surface/bulk structures and optical/electrical properties of carbon was studied. It was found that variation in power density during deposition can control bulk and surface properties of the carbon films. As an effect of post APP treatment, the surface roughness of carbon films increased with treatment time along with the selective removal of  $sp^2$  clusters. The removal of  $sp^2$  clusters was observed by NEXAFS studies. The APP treatment caused the increase in the sheet resistance as well as the optical band gap of the carbon films. Consequently, a technique to control structural, optical and electrical properties for bulk and surface of the magnetron-sputtered carbon films, by controlling power density variation during deposition and the subsequent APP treatments has been developed.

#### **Chapter 5: SnO<sub>2</sub> nanoparticle bound graphene composites - In-liquid plasma assisted synthesis using SnO<sub>2</sub> dispersed ethanol**

In chapter 5, a hybrid approach for SnO<sub>2</sub>-graphene composite synthesis has been discussed. A hybrid approach for SnO<sub>2</sub>-graphene composite formation based on the in-liquid plasma was developed by dispersing SnO<sub>2</sub> nanoparticles in ethanol, which was a single precursor for the graphene. A uniform distribution of SnO<sub>2</sub> nanoparticles on graphene sheets was achieved. As found in the Raman analysis and XRD, the SnO<sub>2</sub> dispersion caused the formation of disorder and less crystalline graphene.

The studied hybrid approach was simple, low cost, atmospheric pressure, and room temperature operation and can be easily scalable to fabricate gas sensing devices based on SnO<sub>2</sub>-graphene composite.

#### **Chapter 6: One-step, in-situ binding of tin oxide nanoparticles to graphene nanosheets by in-liquid plasma**

In chapter 6, an in-situ approach for SnO<sub>2</sub>-graphene composite synthesis has been discussed. The in-situ binding of SnO<sub>2</sub> nanoparticles to graphene sheets by in-liquid plasma method was achieved to fabricate SnO<sub>2</sub>-graphene composite at low temperature

and atmospheric pressure in a single step processing using dissolved SnCl<sub>2</sub> in ethanol as the only precursor. As observed from TEM, SnO<sub>2</sub> nanoparticles of size around 2-3 nm were uniformly distributed and attached to both sides of graphene sheets. XRD and Raman analysis showed the formation of well-crystalline materials. The chemical composition studies from FTIR showed SnO<sub>2</sub> and graphene functional groups indicating the formation of SnO<sub>2</sub>-graphene composite. The studied in-situ binding synthesis route was facile, low cost and can be easily used to fabricate devices based on SnO<sub>2</sub>-graphene composite.

**Summary:**

It was found that plasma processes are advantageous for the surface modification of carbon films and synthesis of graphene-based composite due to non-equilibrium physical-chemical processes. The graphene-based composite can be formed by in-situ, ex-situ and hybrid approaches using in-liquid plasma efficiently at relatively low temperature, atmospheric pressure with single step processing, which collectively reduced the complexity of the composite formation process.