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学位論文題目	Synthesis and Characterization of Graphene and Graphitized Nanocarbons （グラフェンおよび黒鉛化ナノカーボンの合成と評価）
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論文内容の要旨

The world today is progressing day by day in every aspect that makes our life smarter and easier by one or more ways. One of the major aspects that directly inhibits our living is the information technology based on the electronics. With advancing thoughts and lifestyle, the search of new materials to continue the pace of this advancement is also being critical. As the era of silicon based electronics technology is in the verge of its peak performance, the exploration of these new materials becomes more essential. Graphene and other graphitized nanocarbons like carbon nanotubes (CNTs) and carbon nanofibers (CNFs) can be significant for this, due to their promising properties and their potential applications. With the introduction of chemical vapor deposition (CVD), scalable synthesis of these novel materials is widely increasing. In this thesis, the controllable synthesis in larger domain size and in growth rate was tackled for this well-known CVD method, as well as the growth position control in solid phase reaction method.

Chapter 1 includes the introduction to the history, the synthesis techniques, the properties and the potential applications of graphene, as well as the brief explanation of the synthesis, properties, applications and the graphitization techniques of amorphous CNFs.

Chapter 2 briefly explains the methodology adopted in the synthesis and characterization of graphene and CNFs with the brief introduction of the materials and devices used in this study.

Chapter 3 introduces a solid source CVD system to tune the graphene growth into anisotropic and isotropic. Controlling the isotropic and anisotropic graphene growth is a critical aspect to understand the growth dynamics for synthesizing large-area single crystals. The effect of carrier gas flow rate and controllability on isotropic and anisotropic graphene growth was established for the atmospheric pressure CVD method. It was found that the idea not only tuned the graphene growth process but also increased the growth rate of isotropic crystals by about 10 times than that of anisotropic crystals without hindering the graphene quality. This achievement can be essential to achieve faster growth of large single crystals to avoid the degradation of electrical properties of graphene due to the presence of graphene grain boundaries.

Chapter 4 discusses the growth of Mo included CNFs (Mo-CNFs) and the graphitization of amorphous carbon in the form of CNFs under the catalysis of Mo during Joule heating in in situ transmission electron microscopy (TEM) processing. For the fabrication of graphene-based nano-scale interconnects, precise control over their position and proper nanoscale soldering are essential. Amorphous CNFs were effectively found to be converted to highly crystalline few-layered graphene during the electromigration of Mo nanoparticles (NPs). It was also found that during the graphene formation process, agglomerated Mo particles can be effectively channeled to the end of graphene by voltage-driven electromigration and acted as a soldering agent, providing the prospect of the further exploration of Mo as a nanoscale soldering material. Thus, this is work explored the double role of Mo: As a catalyst for graphene synthesis and as a soldering material.

Chapter 5 explores the use of low melting point metal Ga in the synthesis of Ga incorporated CNFs and their consecutive graphitization by Joule heating during in situ TEM as well as under normal vacuum annealing. For the first time, the graphitization temperature was explored to be about 600 °C for the material system of the mixture of Ga NPs and amorphous carbon matrix. Increasing the temperature, agglomeration and evaporation of Ga NPs took place together with the graphitization at the periphery of Ga NPs at the surface region. At the same time, in-situ TEM processing led to the accelerated increase in electrical conductivity with the structural change from amorphous to graphitization. This combination of the in-situ and ex-situ TEM observations is considered as a lead step to understand deeper the graphitization process and provide the information in nanoscale.

Chapter 6 summarizes the whole work of this study and explores the future prospects.

論文審査結果の要旨

Graphitized nanocarbon including graphene attracts the significant attention due to the promising properties in a wider range of applications. For the practical application, the controllable synthesis is important. In this thesis, the controllable synthesis in larger domain size and in growth rate was tackled for the well-known chemical vapor deposition (CVD) method, as well as the growth position control in solid phase reaction method.

Chapter 1 includes the fundamentals and the potential applications of graphene and carbon nanofibers (CNFs), as well as the motivation and the purpose of the thesis.

Chapter 2 briefly explains the methodology adopted in the synthesis and characterization of graphene and CNFs.

Chapter 3 introduces the controllable growth of isotropic and anisotropic graphene using a solid source atmospheric pressure CVD system. It was demonstrated that the flow rate of the carrier gas played a crucial role in the controllability of isotropic and anisotropic graphene growth, and that the growth rate of isotropic crystals by ~10 times higher than that of anisotropic crystals was achievable without a degradation of the graphene quality. This achievement can be essential for the faster growth of large single crystals graphene for the device applications.

Chapter 4 discusses the growth of Mo included CNFs (Mo-CNFs) and the transformation of amorphous CNFs to highly crystallized few-layered graphene induced by the electromigration of Mo nanoparticles (NPs) during Joule heating in in situ transmission electron microscopy (TEM) processing. The electromigration of Mo NPs also induced the soldering of the CNF in nanoscale. Thus, this is work explored the double role of Mo: As a catalyst for graphene synthesis and as a soldering material in nanoscale for the future nanodevices.

Chapter 5 explores the use of low melting point metal Ga in the synthesis of Ga incorporated CNFs and their consecutive graphitization by Joule heating during in situ TEM as well as under normal vacuum annealing. The graphitization temperature was explored to be ~600 °C for the material system of the mixture of Ga NPs and amorphous C matrix for the first time. In-situ TEM demonstrated an accelerated increase in electrical conductivity with the structural change from amorphous to graphitization. Thus, the combination of in-situ and ex-situ TEM is considered as a powerful tool to understand the graphitization process deeper.

Chapter 6 summarizes the whole work of this study and explores the future prospects.

Thus, this thesis demonstrated the controllable growth in size, growth rate and position for graphene and graphitized nanocarbon for the future device applications. This is enough worth for PhD thesis.