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論　文　要　旨
Thesis Abstract

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※報告番号	第　　　号	氏　名 (Name)	KETSOMBUN EKKAPHOP

主論文題名　(Title)

Optimization of molybdenum(V) chloride intercalation process to graphene for conductor applications

内容の要旨　(Abstract)

Conductor materials such as copper (Cu) have been used for electron devices. Recently, Internet of Things (IoT), artificial intelligence (AI), and 5G are leading the development of electronic device technologies including conductors. For electronic device applications, conductors are used as the materials for inductors, interconnects, and transparent conductive films. Doped graphene has attracted people's interest as the new conductor material due to its potential of low-resistance, high reliability, and high inductance density. Few layer graphene (FLG) is expected to be used in applications that include LSI interconnects and transparent electrodes of solar cells. Doped multilayer graphene (MLG) is the one candidate for inductors to achieve both continuous sizes scaling while fulfilling the inductance and performance requirements due to the increasing of kinetic inductance after doping. This study reported the optimization of process guidelines for the $MoCl_5$ intercalation process which is known as a stable doping method. $MoCl_5$ intercalation for FLG, exfoliated highly oriented graphite (e-HOPG) to mimic the high crystallinity of MLG, and chemical vapor deposition-MLG (CVD-MLG) as a practical deposition method. This study also proposes a practical fabrication process for doped CVD-MLG patterns as a potential method for inductor applications.

The optimization of the FLG was expected to be used as a scaling down of the interconnects and transparent electrodes. Moreover, the results of the FLG was used for the process guidelines of thicker graphene layers. This work proposes a $MoCl_5$ intercalation process for doping FLG at a low temperature of 150 °C using a high concentration of $MoCl_5$ chemicals. Bilayer graphene (BLG) was successfully doped at 150 °C without serious damages within a short time process of 30-60 min. The uniformity of doping was improved by increasing the reaction time. However, the damage on the narrow width was found. To avoid the damage, the work proposed to reduce the chemical concentration of the $MoCl_5$ intercalation process.

For the reduced chemical concentration, this work found that optimum intercalation temperature and time depend on the layer number of FLG. The bilayer graphene (BLG) was intercalated at 175°C without serious damage, besides that higher temperature of 200°C was required to intercalate tri-layer graphene (TLG) with a fixed reaction time for 60 min. Although we can reduce the intercalation temperature for TLG, a longer reaction time is required and higher damage is found. After considering both viewpoints of the effective doping and low damage, the high reaction temperature with short reaction time conditions may be suitable for the TLG intercalation process.

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The stacked upper layer is considered to protect the underlying layer from chemical damage during the intercalation process and it leads to the higher activation energy for intercalation at the same time. Therefore, higher reaction temperature or longer reaction time are required for intercalating FLG with more layer numbers.

Thick MLG, of the order of micrometers, is required for MLG applications in high-frequency devices, such as inductors. e-HOPG films were used to mimic a high crystallinity MLG films. Damage-less doping process with $MoCl_5$ intercalation for e-HOPG has been developed by optimizing the chemical concentration and temperature. This work found that the thick e-HOPG films are more susceptible to the intercalation damage than the FLG. The damage was found to be reduced by lowering the chemical concentration. This work found that there is a trade-off relationship between the doping efficiency and damage. An efficient doping with 77% reduction of sheet resistance without serious damage was obtained with the further optimization of temperature and time. The strain and hole density induced during the intercalation process were analyzed using G and 2D peak correlation plots. The changes in strain and carrier density agreed well with the observed damage and sheet resistance. Moreover, the intercalation process obtains high stability after storage in the N_2 box for 40 weeks. It is suggested that the optimization of intercalation conditions is essential for low-resistance and high-inductance applications of doped e-HOPG.

CVD-MLG is expected to be a more practical method for device manufacture than e-HOPG. The optimization of CVD temperature provides a guideline for the $MoCl_5$ intercalation process with patterned CVD-MLG. High-crystallinity CVD-MLG with a G/D ratio of 88 was obtained with a high CVD temperature of 900 °C. A higher crystallinity of the G/D ratio more than 20 was required for $MoCl_5$ intercalation. The morphology did not change after the intercalation process. Cross-sectional SEM images showed the stacked layers of CVD-MLG and Ni. Part of the Ni layer was removed after intercalation at a temperature of 300 °C. It is suggested that the doped of CVD-MLG can be used as a more practical method for the doped-MLG inductor fabrication.

For the practical fabrication process of the doped MLG pattern such as inductor, this work proposed the selective CVD-MLG on Ni catalyst and stable $MoCl_5$ intercalation process with the G/D ratio above 20. Finally, stage 2 intercalation and above was achieved by increasing the intercalation temperature and using high-crystallinity CVD-MLG. The sheet resistance was reduced after intercalation.

In this study, we investigated the optimization of $MoCl_5$ intercalation, which is known as stable intercalation, to MLGs of various film thickness. There is a trade-off relationship between the doping density and the damage induced by the $MoCl_5$ intercalation process, therefore the optimization will be inevitable depending on the MLG thickness. The optimized condition will lead to obtain a low-resistance and stable MLG for various conductor applications in electron devices to support the advancement of the IoT, AI, and 5G technologies and beyond.

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