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Detection of limonene using graphene field effect transistor modified by self-assembling peptide

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Abstract

Graphene has shown promise for biosensor applications due to its excellent electronic properties. The key to improving its function as a sensor is the method of surface modification of the probe to complement the target molecule. One method of surface modification is self-assembling peptides. Self-assembling peptides are known to physically adsorb onto the surface of graphene and form a uniform, monolayer-thick structure, allowing the probe to be anchored to the surface without compromising the electronic properties of graphene. In addition, the amino acid sequence can be designed freely according to the target molecule. In this study, we have used low molecular weight, non-polar molecules to demonstrate biosensors. By changing the amino acid sequence of the probe, we observed that the response to the adsorption of the target molecule was altered. This new approach is expected to improve the sensitivity and selectivity of biosensors.

1. Introduction

Graphene, a representative two-dimensional material, has excellent electronic properties due to its high mobility and specific surface area. Biosensors based on graphene field-effect transistor (GFET) are expected to be applied in various fields such as medical diagnosis, environmental monitoring, and security management. A number of studies on biosensors using GFET have been carried out so far. The surface functionalization with probe molecules is crucial, which gives a high affinity to the target molecule. In this process, covalent bonding and π-π interaction have been used to immobilize probe molecules to the surface. However, there are two disadvantages with these detection methods. Firstly, chemical modifications may degrade inherent electronic properties of graphene. Secondly, random adsorption of probe molecules on the surface may result in loss of the probe activity. These lead to a decrease in sensitivity as a sensor.

So far, there have been various demonstration of GFET biosensors reported. The target of the biosensors has been living cells, virus, DNA, and proteins [1]. There is a still challenge here to detect relatively small molecules. These small molecules have low molecular weight and generally do not have a polarization. GFET-based biosensors have a drawback for these small molecules, because the detection mechanism depends on the Coulombic interaction between trapped molecules and graphene. In addition, most of the small molecules are volatile and insoluble. Therefore, in aqueous solutions, it is very difficult to detect volatile molecules dissolved in small amounts. This is also one of the challenges for biosensors.

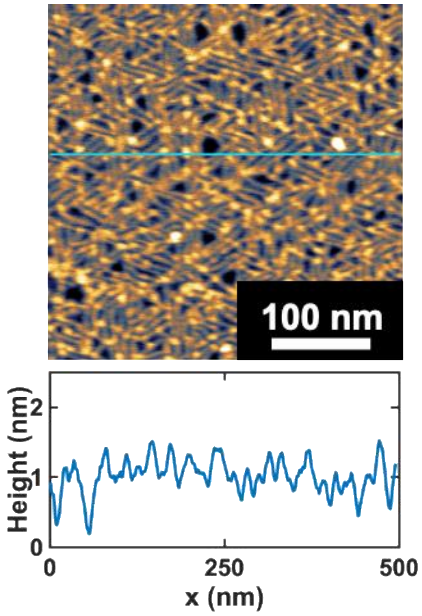

Fig 1. AFM image of self-assembled peptides on graphite surface. A monolayer thickness structure with hexagonal symmetry was confirmed to be formed.

To solve these problems, we suggest an alternative surface modification method using peptide self-assembly [2]. Peptides are known to recognize crystalline structures of inorganic material surfaces and form uniform structures through interactions among molecules. These peptides are physically adsorbed to the surface. Thus, the surface can be modified without losing their intrinsic electronic properties

[3]. These self-assembly of peptides has been demonstrated on gold, boron nitride (BN), transition metal chalcogenides, and graphene in previous studies. In this study, we aimed to develop a new technique to detect an odor molecule, which is relatively small and volatile.

2. Result and discussions

For this sake, we utilized rationally designed peptides as a molecular scaffold and probe peptides on graphene surface [4]. This peptide can maintain it structural uniformity even after rinsing with pure water. The morphology of the self-assembled peptides were characterized by atomic force microscopy (AFM) as shown in Fig 1. Then, we performed odor sensing with limonene, a representative molecule for the smell of lemons. The conductivity of GFETs with respect to the gate voltage was measured with a platinum reference electrode in 10 mM phosphate buffer before and after the formation of the peptide self-assembled structures (Fig 2). The electrochemical interactions in terms of charge neutral point and transistor mobility were investigated, and it was found that ordering of peptides is important for maintaining the intrinsic properties of graphene (Fig 3). It proved that self-assembled peptides are suitable as a molecular scaffold for bio-probes on GFETs. As the result, we successfully detected limonene with high sensitivity. We also found that the sensitivity to limonene was altered by the amino acid sequence of the probe domain.

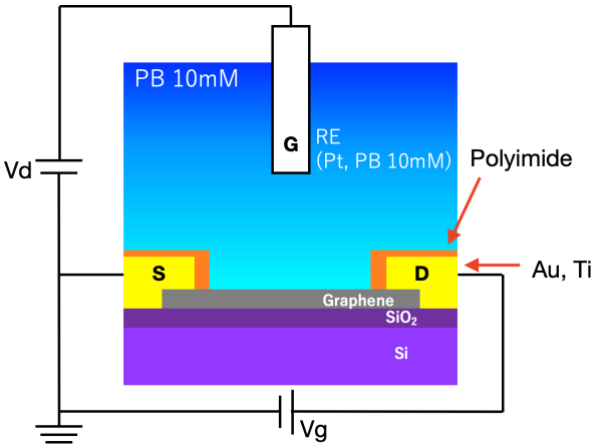

Fig 2. Schematic of the GFET. The source and drain voltages were controlled by the semiconductor parameter analyzer.

3. Conclusions

Self-assembling peptides were used to fix the probe on the surface of graphene and capture the adsorption of target molecules with electrical signals. The peptides formed a uniform structure with a monolayer thickness. The response to the target molecule was altered by changing the amino acid sequence of the probe domain. This new approach is expected to improve the sensitivity and selectivity of biosensors.

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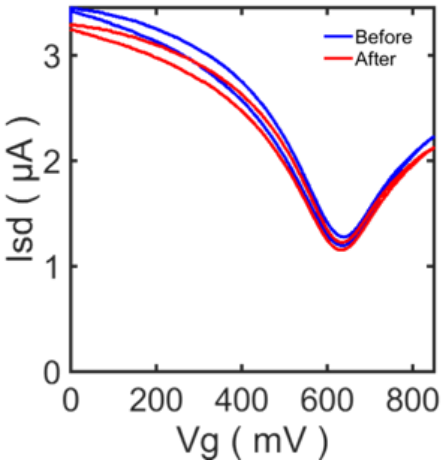

Fig 3. Conductivity of GFET before and after the immobilization of peptides. The gating properties of graphene hardly changed before and after peptide self-assembly.

References

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