Chemical interaction of a graphene surface with molecules in a gas phase produces a change in its electron state. Moreover, the surface electroconductivity change can be induced by electric field or electron transfer to/from a sorbed molecule. That allows us to use graphene as a material for molecular sensors. To enhance the sensor properties of graphene, we have to create reactive centers on its surface such as defects and functional groups [1]. In this case, the electrical graphene properties depend on the degree of its chemical modification (Fig. 1A). Previously, we proposed creating a graphene layer on the surface fluorinated graphite C$_2$F by hydrazine vapour exposure [2]. We can control the degree of reduction by measuring the electrical conductivity of the graphene surface [3]. A part of fluorine molecules remains connected with the graphene layer from the inside and create a positive charge on the graphene. The interaction energy of the reaction centers formed outside the graphene sheet with the molecules of environment is much higher than that for the pristine graphene. This chemical structure of a graphene layer determines its high sensitivity and stability.

The aim of this work is to study the sensing properties of the fluoride graphene reduced surface versus ammonia exposure. The resistance of the reduced surface increases under exposure in NH$_3$ due to electron transfer to the adsorbed molecules. Both the sensor response amplitude and time depend on the recovery degree, which we control by reduction time. To recover the sensor to its initial state, only air purging at room temperature is required. The resistance as a function of NH$_3$ concentration (pressure) follows a general form of the Langmuir isotherm. Comparing the absorption energy extracted from our experimental data with the quantum-chemical analysis, we conclude that the graphene surface has some fluorine atoms attached chemically underneath, which create reactive centers on the top of a graphene surface (free of fluorine).

References


Figures

(A) One cycle of the sensor response showing the difference in amplitude and response time for the samples with the reduction times of 30, 45, 60, and 120 min; (B) Sideview of process of ammonia interaction with the model surface of fluorinated graphene.