Fabrication of nanopores in monolayer graphene with controllable edge structures

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Graphene is known for its high charge carrier mobility, which is more than 100 times higher than that of silicon [1]. Various applications were reported that take advantage of this excellent performance. Among them, fabrication of nanopores in graphene have potential applications such as DNA sequencing from one solvent to another as well as single-molecule detection [2], improving the dimensionless figure of merit (ZT) of graphene by reducing the phonon conductance [3]. On the other hand, the electrical conductance behavior of graphene varies strongly depending on its edge structure, such as zigzag, armchair or mixed type, when the width of graphene nanoribbon (GNR) is sufficiently narrow. It suggests that the nanopore with zigzag edge may show different physical behavior from the one with armchair edge. Therefore, it is important to control the edge structures during fabrication of nanopores in graphene. Recently, large area nanopore arrays were realized by focused helium ion beam milling [4], however, despite the fast processing the atomic edge structure cannot be controlled. More controllable but slower is nanopore formation by electron beam sculpting, but the edge structure was not reported [5]. Later, observations using a transmission electron microscope (TEM) showed that the edges of the processed nanopores in graphene comprise a mixed structure of zigzag, armchair and 5|7 ring pairs [6]. Here, we established a way to fabricate nanopores in monolayer graphene with controlled edge structure.

The nanopores were fabricated and observed by a 50 pm resolved transmission electron microscope R005 operated at 80 kV. The graphene was confirmed to be strong enough to maintain its atomic structures under a moderate electron beam irradiation at 80 kV. However, by tightly focusing the beam, etching of carbon atoms is possible. Nevertheless, we found during our observation that point vacancies generated by removal of a single carbon atom disappeared immediately due to migration of another carbon atom from elsewhere. Thus, it is difficult for a point vacancy to grow to a nanopore when irradiated by low density electron beam. Therefore, in our experiment, an electron beam current of ~3000 pA was adopted to reliably form the nanopores. The size and location of the nanopores was controlled by irradiation time and position of the electron beam. We found that the edge configuration of nanopores can be controlled through the temperature during growth. While room temperature grown nanopores are surrounded by zigzag edges, high temperature (600°C) have predominantly armchair edges, as shown in Fig. 1. We believe that the low current density irradiation is crucial for controlling the edge structures of the nanopores, and this is also related to the temperature of the specimen.

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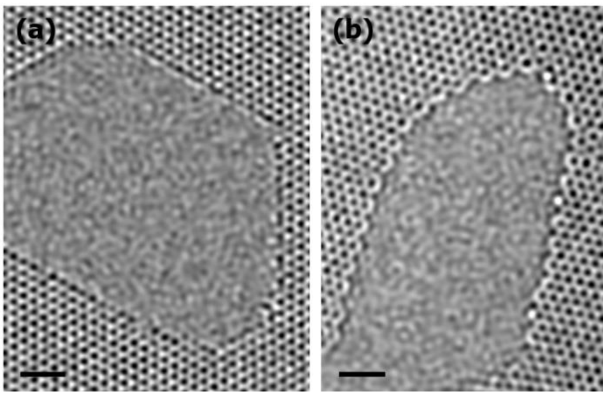

Fig 1. TEM images of nanopores fabricated at room temperature with zigzag edges (a) and at 600°C with armchair edges (b). The scale bars are 1 nm.

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