Effect of gas flow on carbon nanotubes growth on SiC

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Carbon nanotubes (CNTs) can be grown by the SiC surface decomposition method [1]. Contrary to graphene growth from SiC, CNT growth requires heating the SiC(000-1) substrate under vacuum with a little amount of oxygen. Previous studies have shown that heating the SiC(000-1) substrate at 1600°C for 30 minutes in a vacuum of about 1×10^{-2} Pa can form high-density, highly oriented CNTs with a length of about 120 nm [1,2]. In these studies, CNT thin film was grown using a large furnace with graphite insulation which emits oxygen during heating. In this furnace, oxygen emission is hard to control. Therefore, in this study, we grow CNTs using a tabletop tubular furnace under controlled introduction of oxygen into a vacuum. In particular, we investigated the effect of gas flow in the vicinity of the substrate surface.

In this study, a substrate cut from a 4H-SiC(000$\bar{1}$) single crystal wafer manufactured by TanKeBlue, with a thickness of about 400 μm, was used. After cleaning, the substrate was placed on a sample stage and introduced into the center of the furnace tube. The temperature was raised from room temperature to 1500°C at a rate of 7.5°C/min and kept at 1500°C for 2 hours. A nitrogen-oxygen mixed gas (N_2:O_2 = 80:20) was introduced at a flow rate of 0.020 sccm when the temperature reached 1100°C, and the pressure during heating was controlled to be approximately 1.0×10^{-2} Pa. The feature of the CNT film was observed by a transmission electron microscope (TEM).

First, we investigated the characteristics of the CNTs formed when the sample stage arrangement was changed, without gas introduction. When any gas was not introduced, air introduced into the furnace by leakage is the main species in vacuum. Figure 1 shows a schematic diagram of the sample stage and TEM images of the CNT film. In sample #1, non-uniform CNTs with a length of approximately 30 nm were formed. In sample #2, CNTs of uniform length of approximately 70 nm were formed. In these cases, the interface between the CNT film and the SiC substrate was flat. On the other hand, when N_2:O_2 gas was introduced, the interface was different from the above results. In sample #3, long CNTs of approximately 100 nm in length were formed, but their interface was rough. These results suggest that the CNT film was thicker when the gas flow velocity near the substrate surface is high, and that some gas molecules other than N_2/O_2 affect the smoothness of the interface.

References:
[1] M. Kusunoki, *et al.*, Chem. Phys. Lett. 366, 458 (2002).
[2] K. Matsuda, *et al.*, J. Appl. Phys. 123, 045104 (2018).

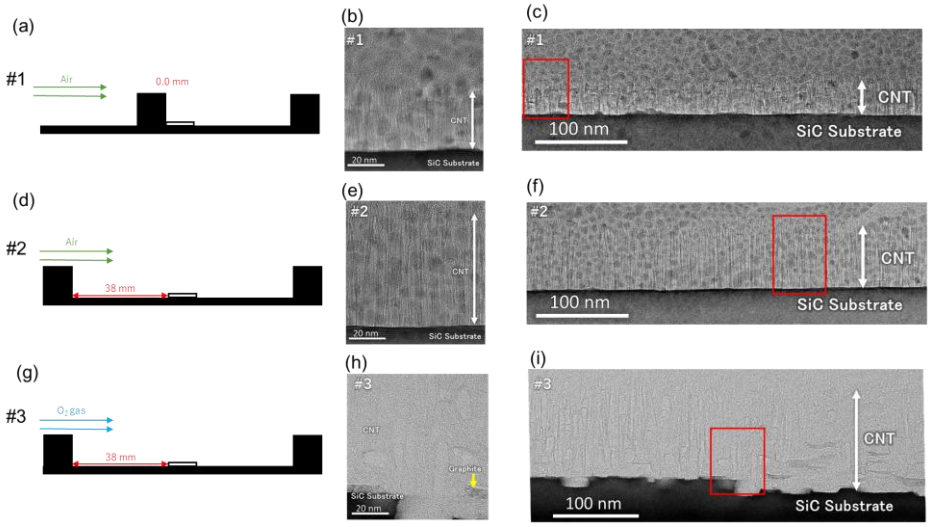

Fig. 1: Schematic diagrams of the sample stage configurations (a) #1, (d) #2, and (g) #3. Low-magnification TEM images of the samples #1, #2, and #3 are shown in (b), (e), and (h), and the corresponding high-resolution TEM images are shown in (c), (f), and (i), respectively.