
Xing Xin

Zeyuan Fei, Teng Ma, Long Chen, Mao-Lin Chen, Chuan Xu, Xitang Qian, Dong-Ming Sun, Xiu-Liang Ma, Hui-Ming Cheng, Wencai Ren

1. Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, P. R. China

2. University of Chinese Academy of Sciences, Shenyang 110016, P. R. China.

Contact: xxin13b@imr.ac.cn

Circular Graphene Platelets with Grain Size and Orientation Gradients Grown by Chemical Vapor Deposition

Abstract

Materials that have a continuous structure gradient have unique properties and promising applications that are different from conventional homogenous materials. For instance, the structure gradient in bamboo makes it an interesting material with a hard surface but a soft interior. The gradient chemical composition makes the beak of the squid one of the hardest and stiffest organic materials.^[1] Learning from nature, many materials with unique properties have been developed by designing a structure or composition gradient.^[2,3] Although many such three-dimensional bulk materials have been developed, no two-dimensional (2D) materials have been achieved. Graphene as a typical representative of two-dimensional (2D) materials has attracted great attention.^[4,5] Recent extensive studies show that grain boundary has important influences on the electrical, mechanical, thermal and chemical properties of graphene.^[6-9] Therefore, the graphene platelets with grain structure gradient are expected to have gradient properties, and therefore some potential interesting applications as the cases in 3D materials. Here, we have developed a chemical vapor deposition (CVD) method, using a tungsten (W) foil on which was deposited a very thin layer of copper (Cu) as a substrate, to grow circular graphene platelets with gradients in grain size and orientation distribution in the radial direction.^[10] It was found that the substrate undergoes continuous loss of Cu and the formation of a huge number of small tungsten carbides crystals with different orientations during growth. Because of the different interactions and growth behaviors of graphene on Cu and tungsten carbide, such substrates cause the formation of grain size and orientation gradients through the competition of Cu and tungsten carbide in the dominant role in graphene growth. Our findings not only add a new member to the large family of 2D materials but also provide a general strategy to synthesize other 2D materials with a grain structure gradient by substrate design, which opens up possibilities for investigating the unique properties and applications of such materials.

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Figures

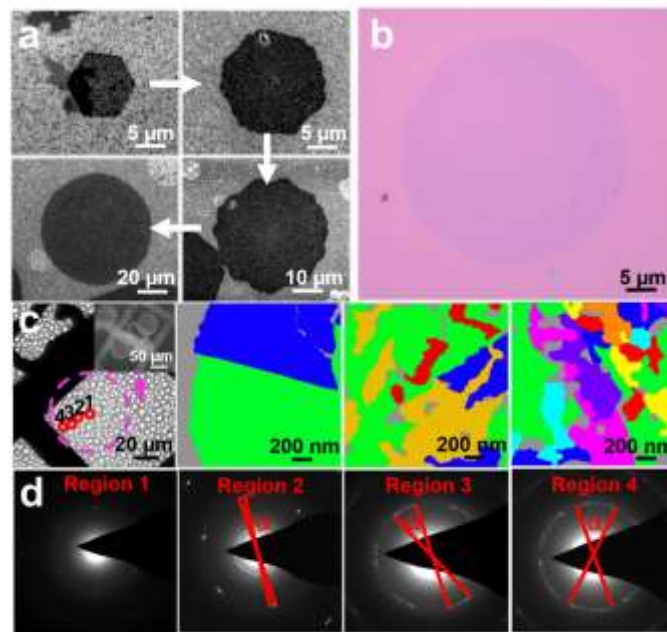


Figure 1: Growth of circular graphene platelets with grain size and orientation gradients. (a) Typical scanning electron microscopy images of graphene islands grown on a W-Cu substrate with a flow rate of 5.0 sccm CH₄ and 300 sccm H₂ for 20, 40, 60, and 90 min, showing the morphology evolution of the islands from hexagonal to circular. (b) Optical image of a circular graphene island transferred onto an SiO₂/Si substrate. (c) A transmission electron microscopy (TEM) image of a circular graphene island and the false-color, superimposed dark-field TEM images of regions 2-4. (d) Selected area electron diffraction patterns taken from the four regions 1-4 in (c). The regions marked 1 to 4 were selected from the center to the edge of a circular graphene island. The above results indicate that the graphene island has a grain size and an orientation-dispersed angle gradient from the center to the periphery.