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High-efficiency field emission from pressed nickel foam–flat graphene–vertical graphene hybrids

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ABSTRACT

Vertical graphene (VG) was prepared on flat graphene (FG) coated pressed Ni foams by using plasma enhanced chemical vapor deposition. The VG is typical few-layer graphene and is rich in defects. The field emission performance of Ni–FG–VG hybrids with optimal morphology is found to outperform that of VG prepared on pristine Ni foams and Si wafers. It has a low turn-on electric field of 2.03 V/μm and a large maximum emission current density (J_{max}) of 5.91 mA/cm^2 and also exhibits excellent field emission stability at ~50% and ~75% J_{max} over a period of 30 h.

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1. Introduction

Vertical graphene (VG) has been demonstrated as a high-efficiency field emission (FE) cathode material in recent years due to its unique two-dimensional structure and it has shed light on device applications such as lighting lamps and X-ray tubes [1,2]. However, VG grown on Si wafers suffers from inadequate graphene–Si–electrode contacts. Catalytic growth of graphene on metal (Cu or Ni) could bridge this gap [3,4], but the flat-lying graphene edge cannot serve as emission sites in this regime. Preparing VG on metal is thus expected. For example, copper foils and stainless steel have been used to grow VG for FE applications [5–7], but the FE performance of VG prepared on these planar substrates is highly influenced by the field screening [8]. Here we report on the preparation of VG on flat graphene (FG) coated pressed Ni foams by using microwave plasma enhanced chemical vapor deposition (PECVD) and discuss the FE performance of differently shaped Ni–FG–VG hybrids. The FG could not only provide a mass of active emission sites but also improve the VG–metal–electrode contacts and the pressed irregular surface of Ni foams could weaken the influence of field screening, all of them lead to the superb FE properties of the pressed Ni–FG–VG hybrids.

2. Experimental

The pressed Ni foams (~150 μm in thickness) were obtained by pressing the commercially available Ni foams (~1.6 mm in thickness) under 10 MPa for 5 min Fig. 1(a) shows a schematic for the preparation of Ni–FG–VG hybrids. It follows a process of thermal CVD growth of FG in a tubular furnace using methane as the carbon source [9] and microwave PECVD growth of VG using acetylene as the carbon source [10]. Scanning electron microscope (SEM, SU8010, Hitachi, 5 kV), transmission electron microscope (TEM, JEM-2010, 200 kV), and Raman spectroscopy (LabRAM HR800, 532 nm) were used to characterize the structure of samples. The FE tests (sample areas: ~5×5 mm^2) were performed by using a classical diode-type setup at ~1.0×10^{-7} Pa [10]. All the details about the sample preparation and characterization are shown in the *Methods* of the Supplementary material (pages S1–S4, including Figs. S1 and S2).

3. Results and discussion

Fig. 1(b) shows the optical images of the resultant samples in different stages. The dark gray and black colors of the samples demonstrate the growth of FG and VG, respectively. Low resolution SEM image of pristine and pressed Ni foams is shown in Fig. 1(c). The macropores of pristine Ni foam are collapsed after.

the pressing, with irregular surfaces left. Top-view SEM images of pressed Ni–FG–VG hybrids prepared at 300 and 150 W are

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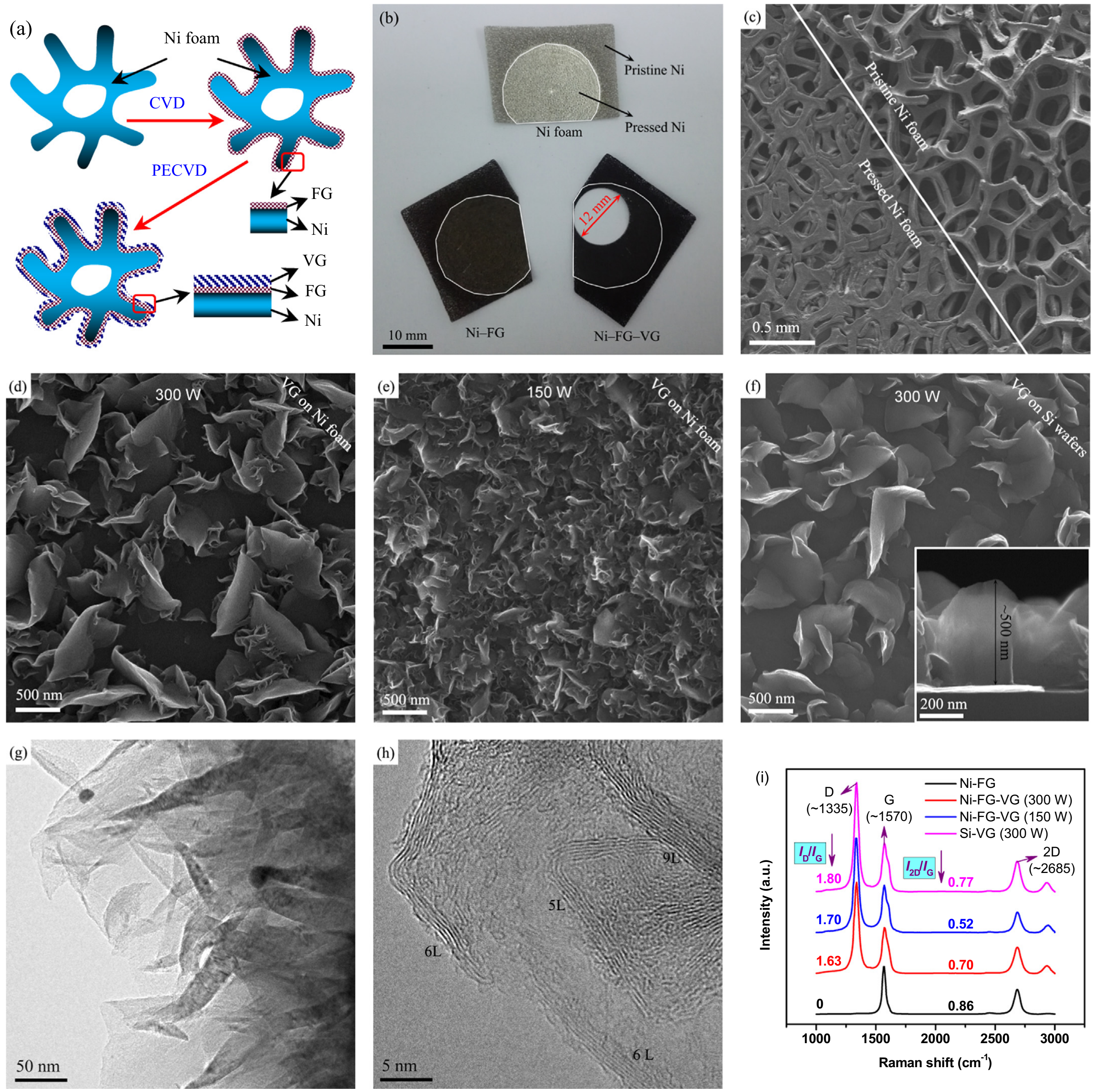

Fig. 1. (a) Schematic for the preparation of Ni–FG–VG hybrids. (b) Optical images of samples in different stages. (c) Low-resolution SEM image of pristine and pressed Ni–FG–VG hybrids. Top-view SEM images of Ni–FG–VG hybrids prepared at (d) 300 W and (e) 150 W. (f) Top-view SEM image of VG prepared on Si wafers at 300 W. Inset: Corresponding side-view SEM image. (g) Low- and (h) high-resolution TEM images of VG. (i) Raman spectra of FG and VG.

shown in Figs. 1(d) and (e), respectively. The VG of the 300 W/Ni sample is ~1 μm in width and is sparsely distributed. Upon the 150 W/Ni sample, the width of a fraction of VG reaches 500 nm, but most of VG is small-sized and densely distributed. This difference of the VG shape is ascribed to the different hydrogen plasma etching when the microwave power is changed, which (including the growth mechanism of VG) has been discussed in detail in our previous studies [2,11]. VG was also prepared on Si wafers for comparison, as shown in Fig. 1(f). The VG is found to have a shape similar to that of the 300 W/Ni one and is ~500 nm in height (inset of Fig. 1(f)). VG prepared on Si wafers was further observed by using a TEM. The low-resolution TEM image (Fig. 1(g)) shows that the VG has corrugations and wrinkles. Fig. 1(h) shows the high-resolution TEM image of the folded edges of VG, which confirms the presence of VG with 5–9 layers. It should be mentioned that most of our VG is typical few-layer graphene with less than 10 layers [12]. Fig. 1(i) shows the Raman spectra of FG and VG prepared in different conditions. Notably, the absence of D peak and the large I_{2D}/I_G ratio (~0.86) of the FG indicate that the FG is high-quality graphene with long-range sp^2 carbon [13]. By contrast, the sharp D peaks and the large I_D/I_G ratios (1.63–1.80) of all the VG samples indicate that the VG is rich in defects [13]. The few-layer nature of VG can also be reflected by its large I_{2D}/I_G ratios (0.52–0.77).

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