Graphene, a 2D $sp^2$-hybridized carbon sheet with one-atom thickness, has attracted increasing attention in recent years because of its unique structure and special properties. Recently several research publications have described the synthesis of 3D CVD graphene continuous networks\(^1\)\(^,\)\(^2\) grown on Ni foam substrates and their utilization for various novel products and application developments such as high sensitive gas sensor\(^3\), electrode or electrode additive for solar cells\(^4\) and ultra capacitors.\(^5\)

The commercially available Ni foam substrate which is used as a sacrificial template for CVD graphene growth in these publications is typically 1 to 2 mm thick, has a density of $\sim$0.3 g/cm\(^3\) and a surface area in the order of $1 \text{m}^2$/g. This results in the growth of around $0.3 \text{m}^2$ of graphene per cm\(^3\) of Ni foam used. However, higher density 3D graphene material is desired in applications (sensors, batteries, ultracapacitors, etc.) where more graphene material per mass/volume of active material is required to achieve better performance.

In addition, one way to achieve higher yield per batch in a coating process is to use particles instead of flat substrates to exploit the increased surface area available for CVD. This path was already explored to achieve bulk growth of high-quality mono- to few-layer graphene sheets on nickel particles.\(^6\)

We improved upon these prior concepts to produce 10-100 higher batch quantity of 3D CVDGraphene™ material by starting with a micron size filamentary Ni powder (Fig.1) and filling it into multiple, specially designed porous molds that allow the precursor gas to enter the Ni powder easily. This resulted in highly porous sintered Ni pellets (5mm tall, 16mm in diameter, Fig.2), uniformly covered with a graphene skin, as can be observed on the corresponding cross-sectional SEM images (Fig.3). Fig.4 shows the free standing 3D graphene network after Ni is etched away.

This novel way to synthesize graphene on 3D interconnected Ni sintered powder allows to create macroscopic custom 3D structures of CVD\(^3\)Graphene™ which can be further customized by using other 3D shapes and sizes of molds and other types (spherical instead of filamentary) and sizes (nm to \(\mu\)m) of Ni powders. The CVD graphene batch yield can thus be 10-100 times improved (larger quantities and/or denser 3D CVD graphene can be achieved per oven volume) and its cost dramatically reduced.

References
Figures

Fig.1: SEM image of the µm size filamentary Ni powder used as the starting material

Fig.2: Image of the resulting sintered Ni pellets coated with CVD3D Graphene™

Fig.3: SEM images of multilayer CVD3D Graphene™ covering the sintered Ni cellular material substrate made from Ni powder.

Fig.4: SEM image of a 3D multilayer CVD3D Graphene™ carbon network obtained after removal of the sintered Ni structure.