Fabricating Large-Area Sheets of Single-Layer Graphene by CVD

Such sheets are components for high-speed digital and RF electronics for defense and commercial communications.

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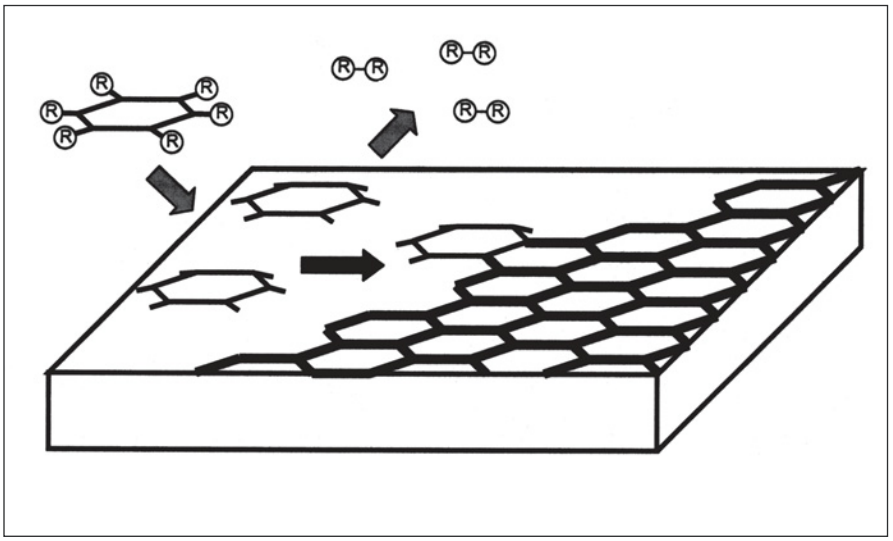
This innovation consists of a set of methodologies for preparing large area (>1 cm^2) domains of single-atomic-layer graphite, also called graphene, in single (two-dimensional) crystal form. To fabricate a single graphene layer using chemical vapor deposition (CVD), the process begins with an atomically flat surface of an appropriate substrate and an appropriate precursor molecule containing carbon atoms attached to substituent atoms or groups. These molecules will be brought into contact with the substrate surface by being flowed over, or sprayed onto, the substrate, under CVD conditions of low pressure and elevated temperature. Upon contact with the surface, the precursor molecules will decompose. The substituent groups detach from the carbon atoms and form gas-phase species, leaving the unfunctionalized carbon atoms attached to the substrate surface. These carbon atoms will diffuse upon this surface and encounter and bond to other carbon atoms. If conditions are chosen carefully, the surface carbon atoms will arrange to form the lowest energy single-layer structure available, which is the graphene lattice that is sought.

A precursor may contain only one carbon atom, such as methane or derivatized methane (e.g., carbon tetrachloride). However, it is more likely that the best precursor molecule will be an aromatic compound that has at least 6 carbon atoms already arranged in the aromatic structure of hexagonal rings, because this will be the geometry of the carbon atoms in the final graphene material. An example of a possible candidate precursor molecule is hexachlorobenzene, an aromatic molecule containing six carbon atoms in a ring, each of which is bound to one additional chlorine atom. In this molecule, the carbon-chlorine bonds are weak enough that they will break under reasonable CVD conditions (a few hundred degrees C); the chlorine atoms will form Cl_2 molecules that will escape into the gas phase, while the six-membered carbon ring will be released upon the substrate surface. If conditions are not too extreme, this six-membered ring will be able to diffuse across the substrate surface while maintaining its structural integrity, eventually encountering other rings of carbon atoms from other precursor molecules. These multiple carbon rings will bond together, their constituent carbon atoms binding together to form the lowest energy structure available — the graphene lattice.



Growth of a Graphene Sheet by CVD using an aromatic molecular precursor gas, shown here as a six-membered ring of carbon atoms, each of which is attached to one additional –R substituent. Carbon-containing precursor molecules impinge upon a substrate and decompose, resulting in gaseous R_2 products and surface-bound rings of carbon atoms. These six-membered rings can maintain their structure as they diffuse toward, and bind to, the growing edge of the hexagonal carbon graphene lattice as shown.

Another method for creating the graphene lattice includes metal-catalyzed CVD, in which the decomposition of the precursor molecules is initiated by the catalytic action of a catalytic metal upon the substrate surface. Another type of metal-catalyzed CVD has the entire substrate composed of catalytic metal, or other material, either as a bulk crystal or as a think layer of catalyst deposited upon another surface. In this case, the precursor molecules decompose directly upon contact with the substrate, releasing their atoms and forming the graphene sheet.

Atomic layer deposition (ALD) can also be used. In this method, a substrate surface at low temperature is covered with exactly one monolayer of precursor molecules (which may be of more than one type). This is heated up so that the precursor molecules decompose and form one monolayer of the target material.

This work was done by Michael Bronikowski and Harish Manohara of Caltech for NASA's Jet Propulsion Laboratory. Further information is contained in a TSP (see page 1).

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