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Graphene - Magic of Carbon

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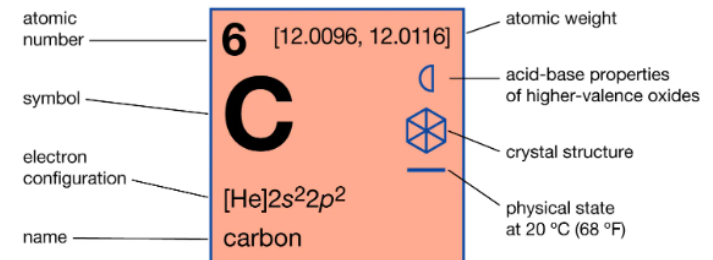


In the second part of the eighteenth-century carbon was first identified as an element. The name "carbon" comes from the Latin word carbon, meaning "charcoal".

Carbon (C) is the sixth element on the periodic table and the sixth most abundant element in the universe.

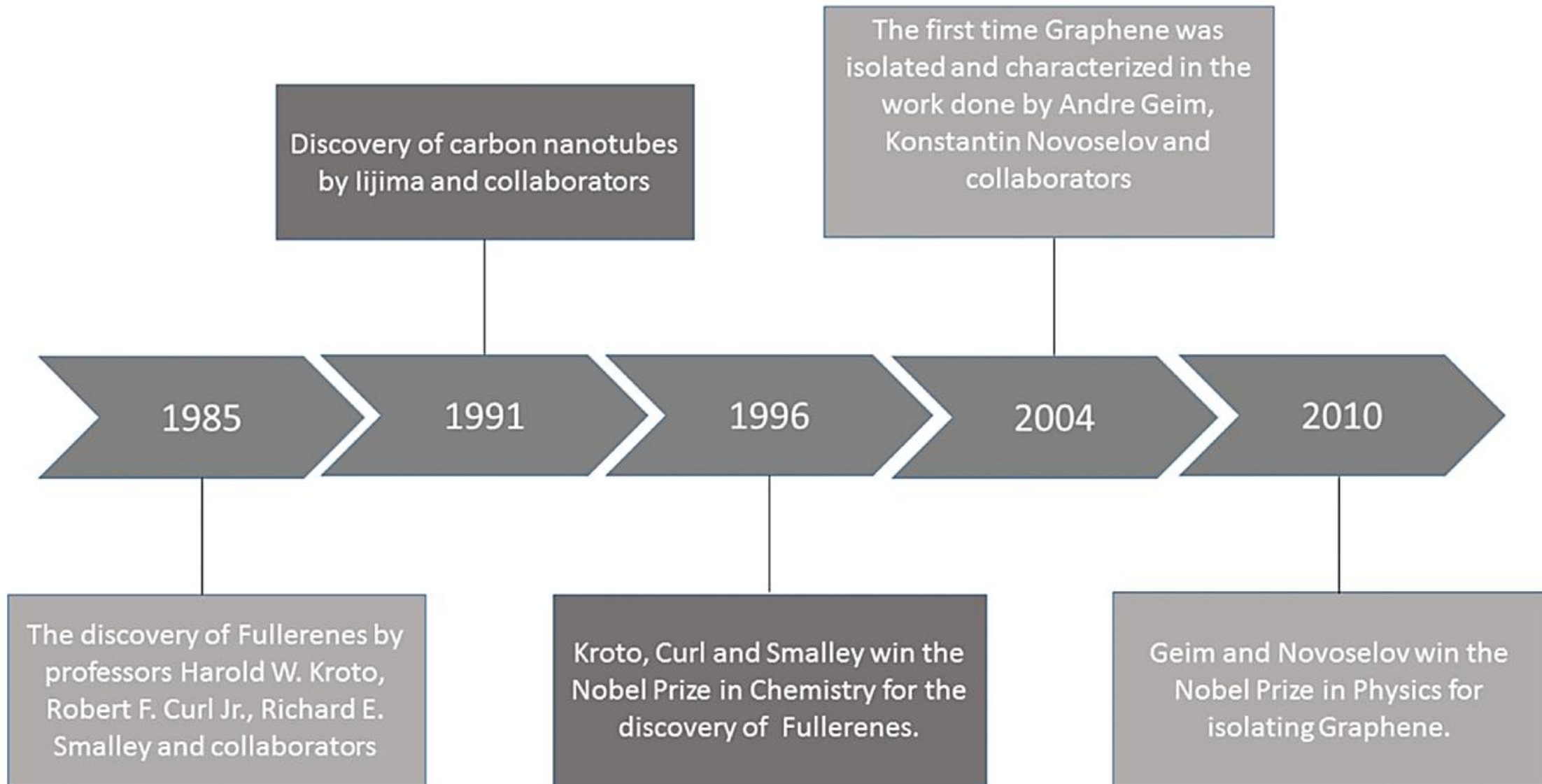
Carbon is unique because of its four valence electrons, which make it very versatile and allow it to bond with many other elements.

Carbon



Other nonmetals	Solid
Hexagonal	Weakly acidic

<https://www.britannica.com/science/carbon-chemical-element>



Allotropy is the property of certain chemical elements to exist in two or more distinct forms (phases), known as elemental allotropes.

Allotropes have different structure: the atoms of the same element are bonded together in different ways.

The allotropes of carbon include:

diamond

graphite

graphene

fullerenes

carbon nanotubes

Q-carbon

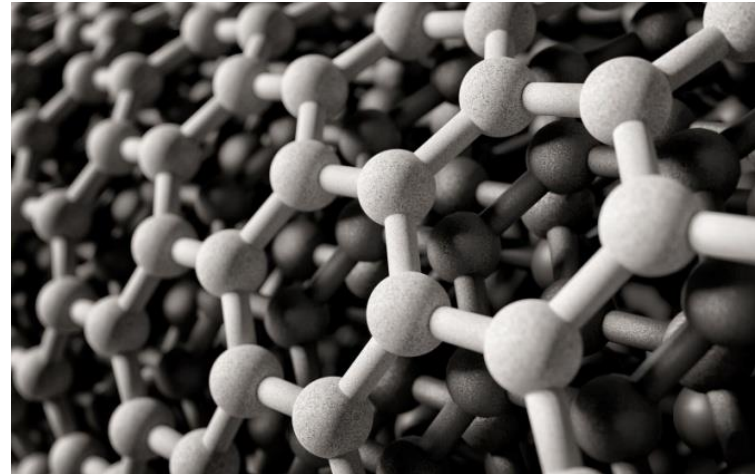
Carbyne

amorphous carbon

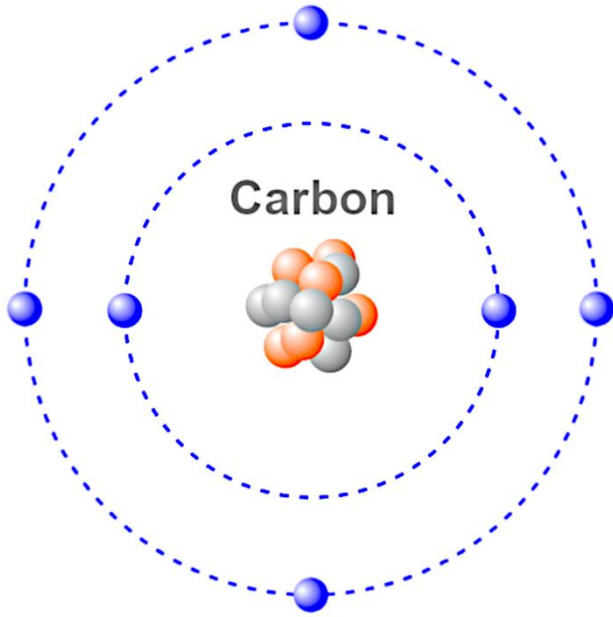
schwarzites

cyclocarbon

glassy carbon



What is the reason for structural variations and cardinal changes in properties?



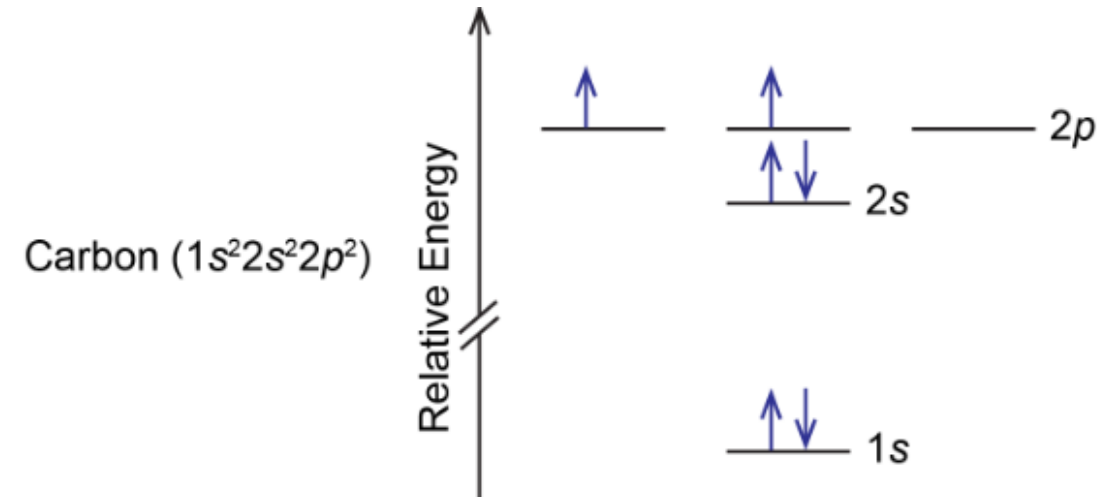
Carbon atoms are made up of a nucleus of six protons and six neutrons (^{12}C isotope) and surrounded by six electrons.

The electron configuration of carbon is $1s^2 2s^2 2p^2$.

The 1s orbital is the lowest energy level. Since 1s can only hold two electrons the next 2 electrons for C goes in the 2s orbital, and it can also hold a maximum of two electrons.

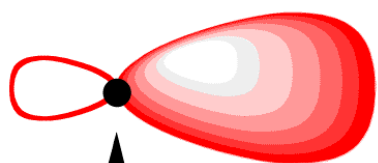
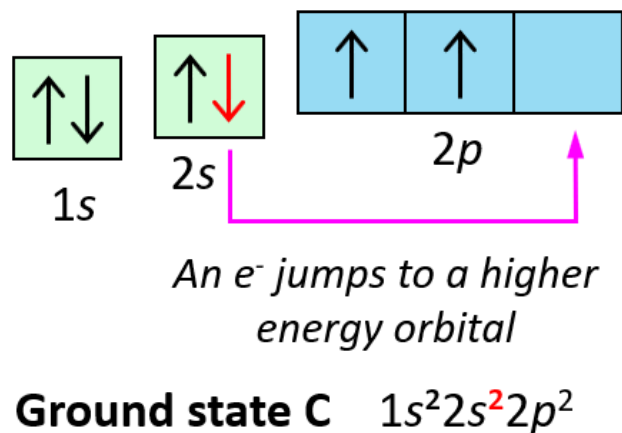
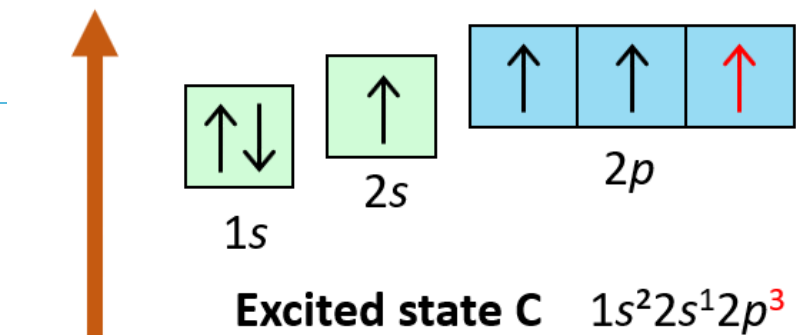
The 2p orbital is the third energy level, and it consists of three sub-levels: $2p_x$, $2p_y$, and $2p_z$.

Each of these sub-levels can hold a maximum of two electrons. In carbon atom two of the 2p orbitals ($2p_x$ and $2p_y$) hold one electron each, while the third 2p orbital ($2p_z$) is empty.



The first wonder of carbon is the electronic structure

Carbon. Electronic structure

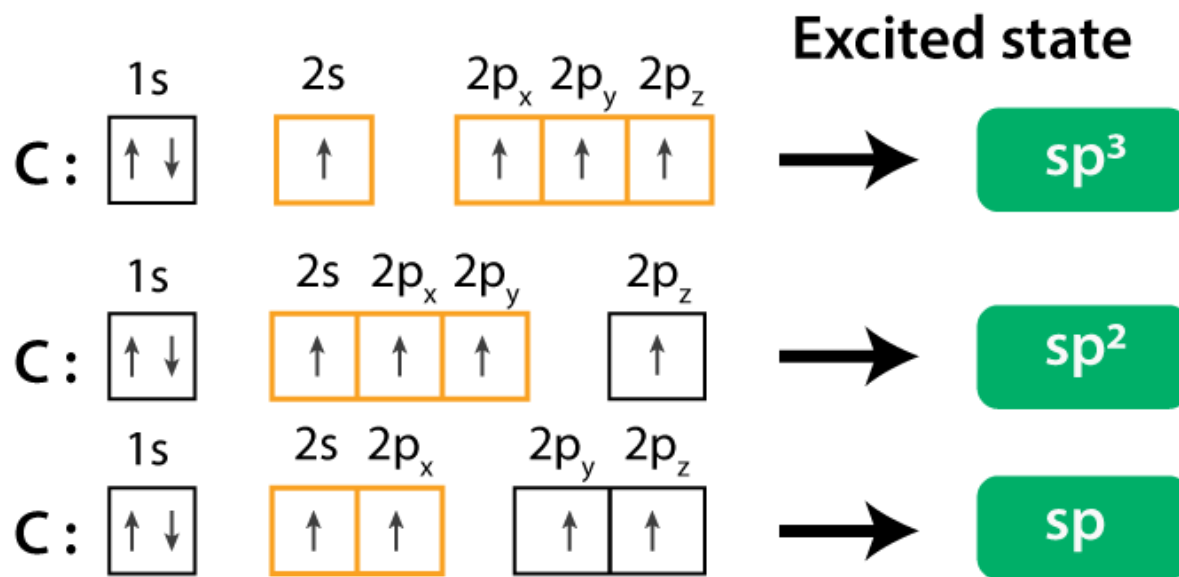


The shape of the single sp -, sp^2 or sp^3 -hybridized orbital

nucleus

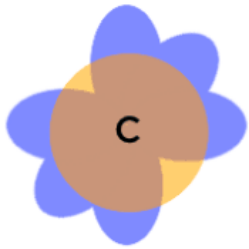
Hybridization is a concept describing the mixing of atomic orbitals to form hybrid orbitals with *different geometries and energies*.

This hybridization occurs when atoms join together to form a more energetically favorable configuration.

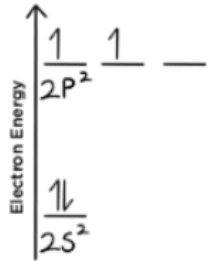


The most common types of hybridization for carbon are:
 sp^3 , sp^2 , sp

Orbital Diagram



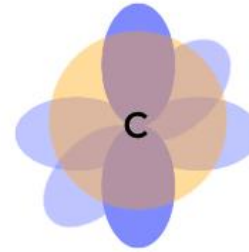
Electronic Configuration



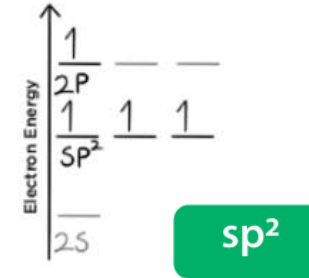
Hybrid Orbital Diagram

No Hybrid Orbitals

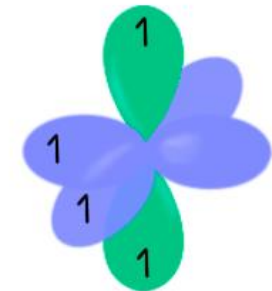
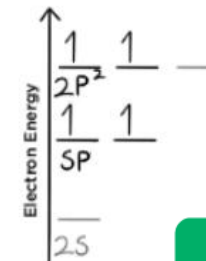
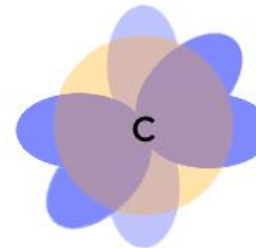
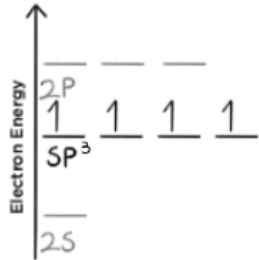
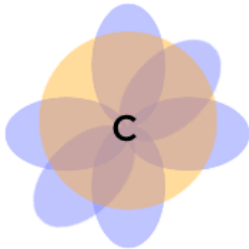
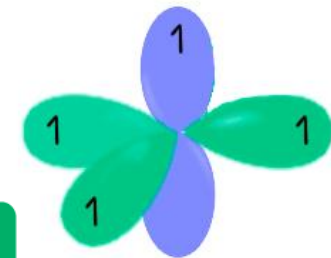
Orbital Diagram



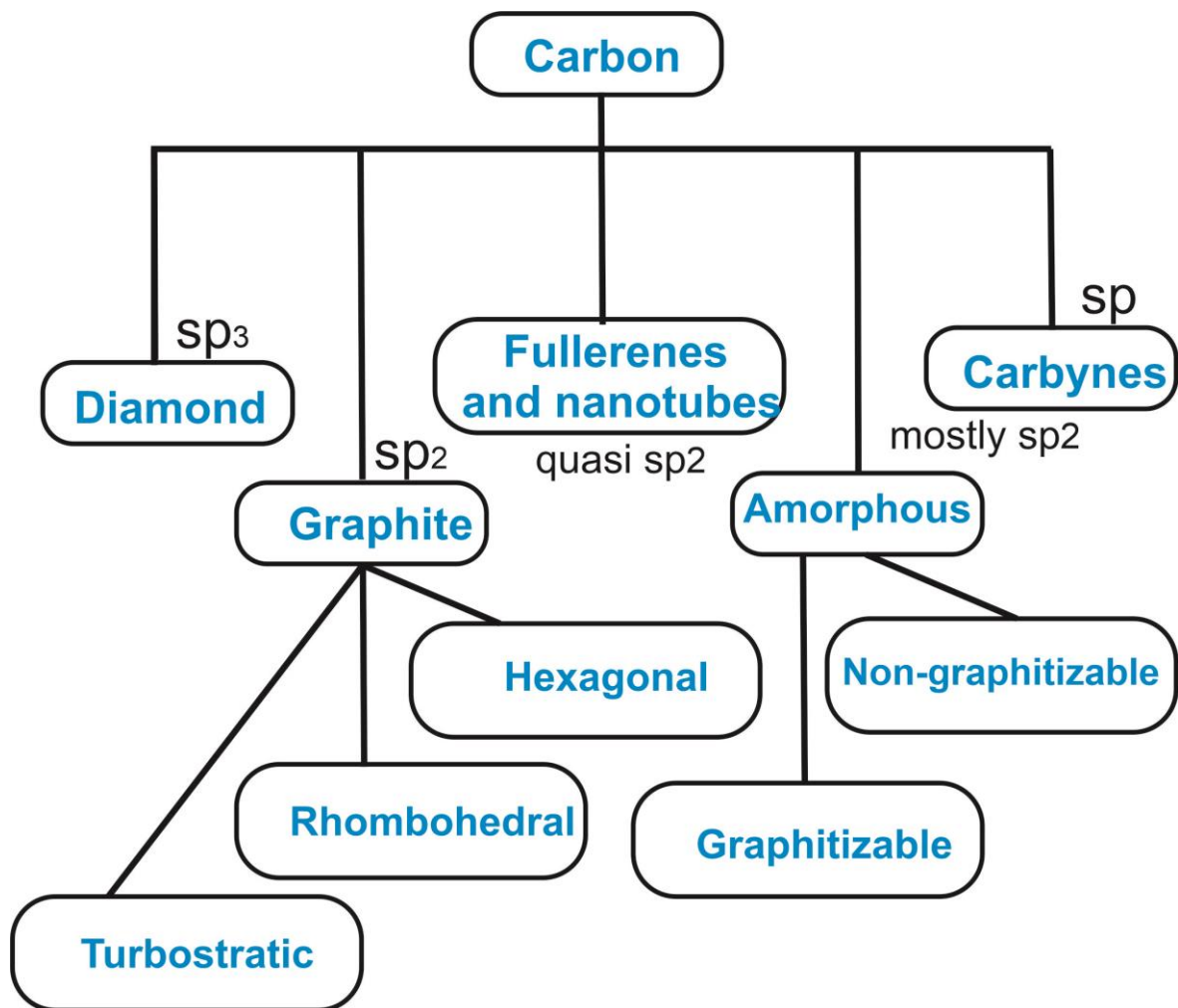
Electronic Configuration



Hybrid Orbital Diagram

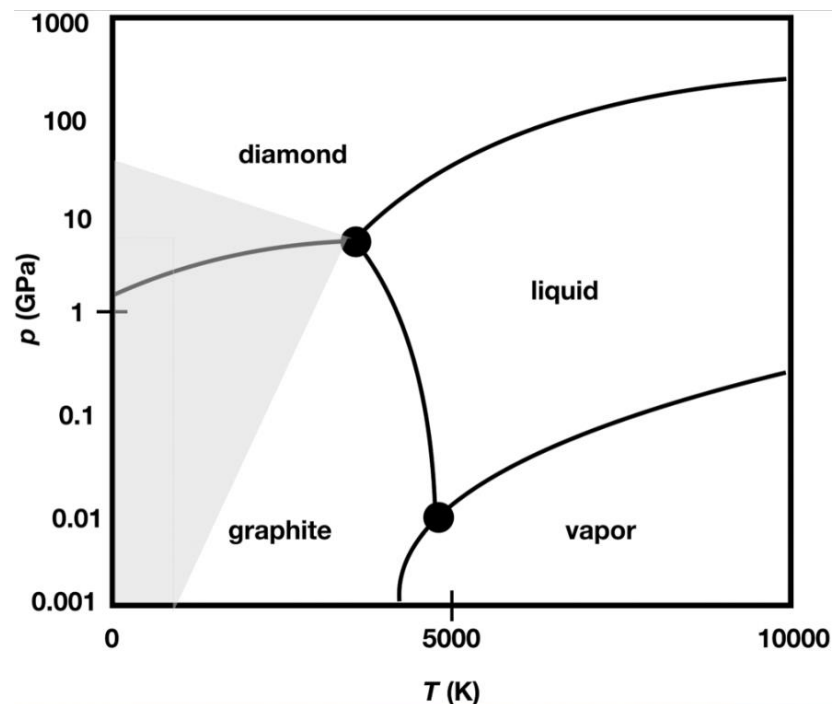


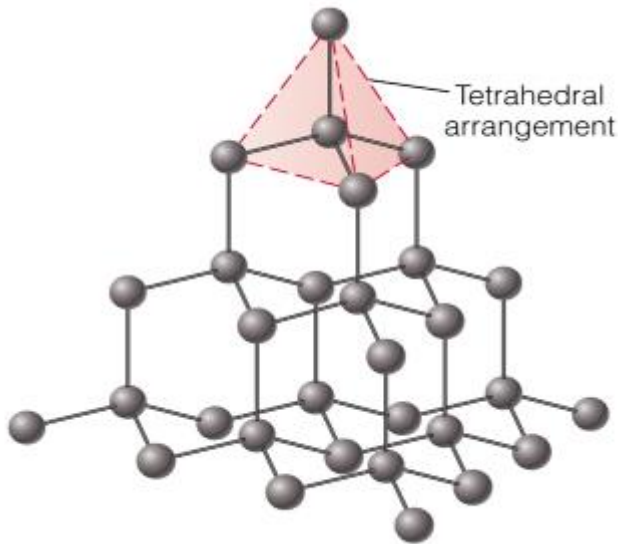
Carbon. Structure of Allotropes



Different geometries of hybridized orbitals result in linear (1-dimensional), planar (2-dimensional), or tetrahedral (3-dimensional) spatial organization of crystalline and amorphous allotropes of carbon.

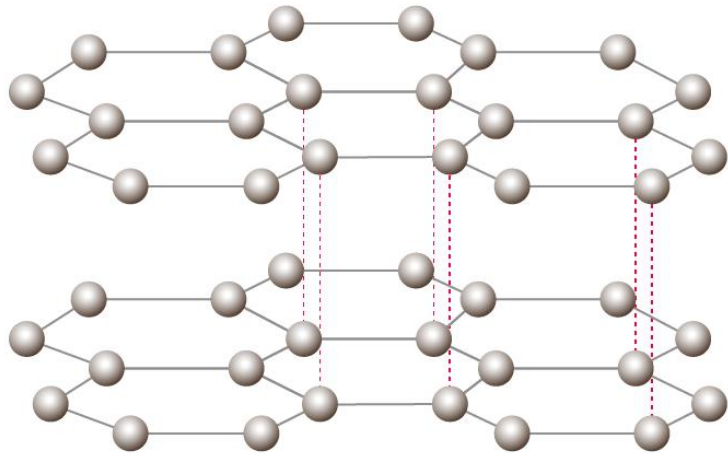
Two main allotropes: graphite and diamond corresponding to sp^2 and sp^3 hybridization





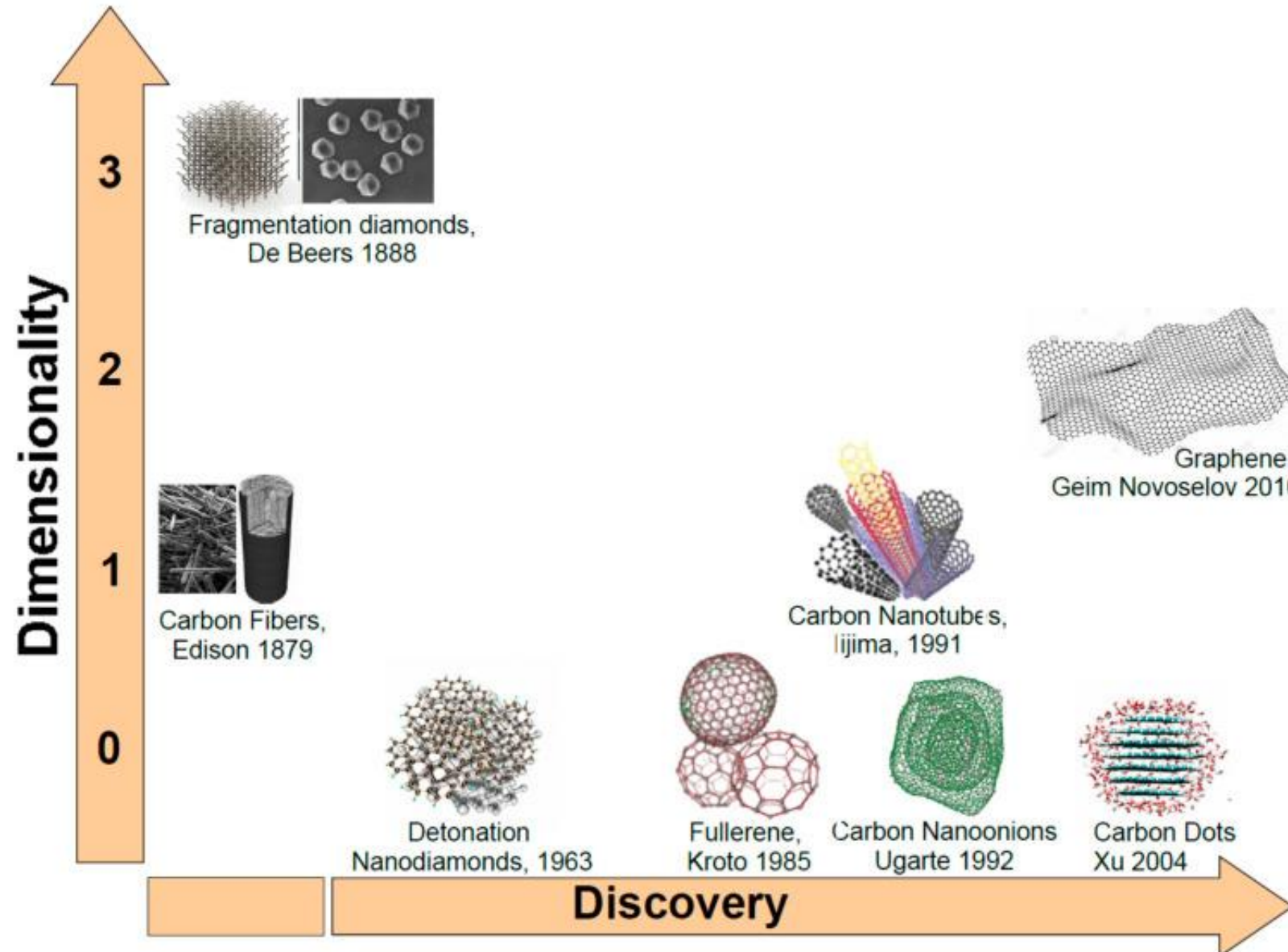
Properties

- ✓ Diamond is the densest form of carbon.
- ✓ Diamond is the hardest natural substance.
- ✓ The melting point of the diamond is 3843K (approx 3570 °C).
- ✓ In diamonds, all valence electrons participate in bond formation. It does not have a lone pair of electrons due to the fact that this diamond has a very low electrical conductivity.
- ✓ Diamond is insoluble in all solvents.
- ✓ When heated at 475K in presence of sulfuric acid and potassium dichromate, diamond is oxidized directly into carbon dioxide, leaving no residue.
- ✓ Diamond has a very high refractive index.
- ✓ It is a good conductor of heat.



Properties of graphite:

- ✓ Graphite is black in color and has a metallic luster.
- ✓ High melting point: The carbon atoms in each layer are bound by strong covalent bonds, so that the melting point of graphite is high, about 3500°C.
- ✓ Conductivity: Every carbon in graphite is sp^2 -hybridized. Thus, one valence electron of each carbon atom can move freely from one point to another. Non-hybrid orbitals containing one electron each overlap in the transverse direction, forming bonds in one layer. These free electrons are delocalized and move under the influence of thermal and electric fields. Thus, it is a good thermal and electrical conductor.
- ✓ Low density of 2.26 g/cm^3 due to large distance between layers.
- ✓ Graphite is slippery because of its characteristic layered structure. It is often used as a dry lubricant.
- ✓ Graphite is chemically stable



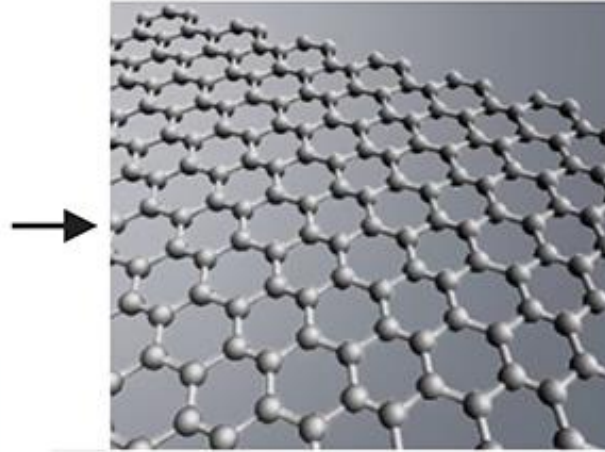
For the discovery of graphene in 2004, the 2010 Nobel Prize in Physics was awarded jointly to Andre Geim and Konstantin Novoselov "for ground-breaking experiments regarding the two-dimensional material graphene"

Carbon nanostructures ordered following dimensionality and discovery time

GRAPHITE



GRAPHENE



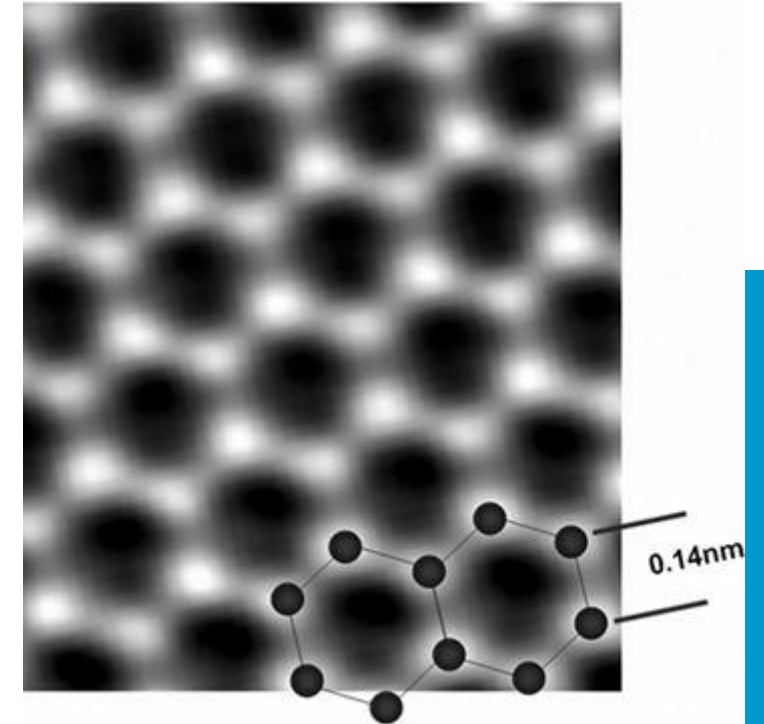
Graphene is a single atom thick sheet of carbon.

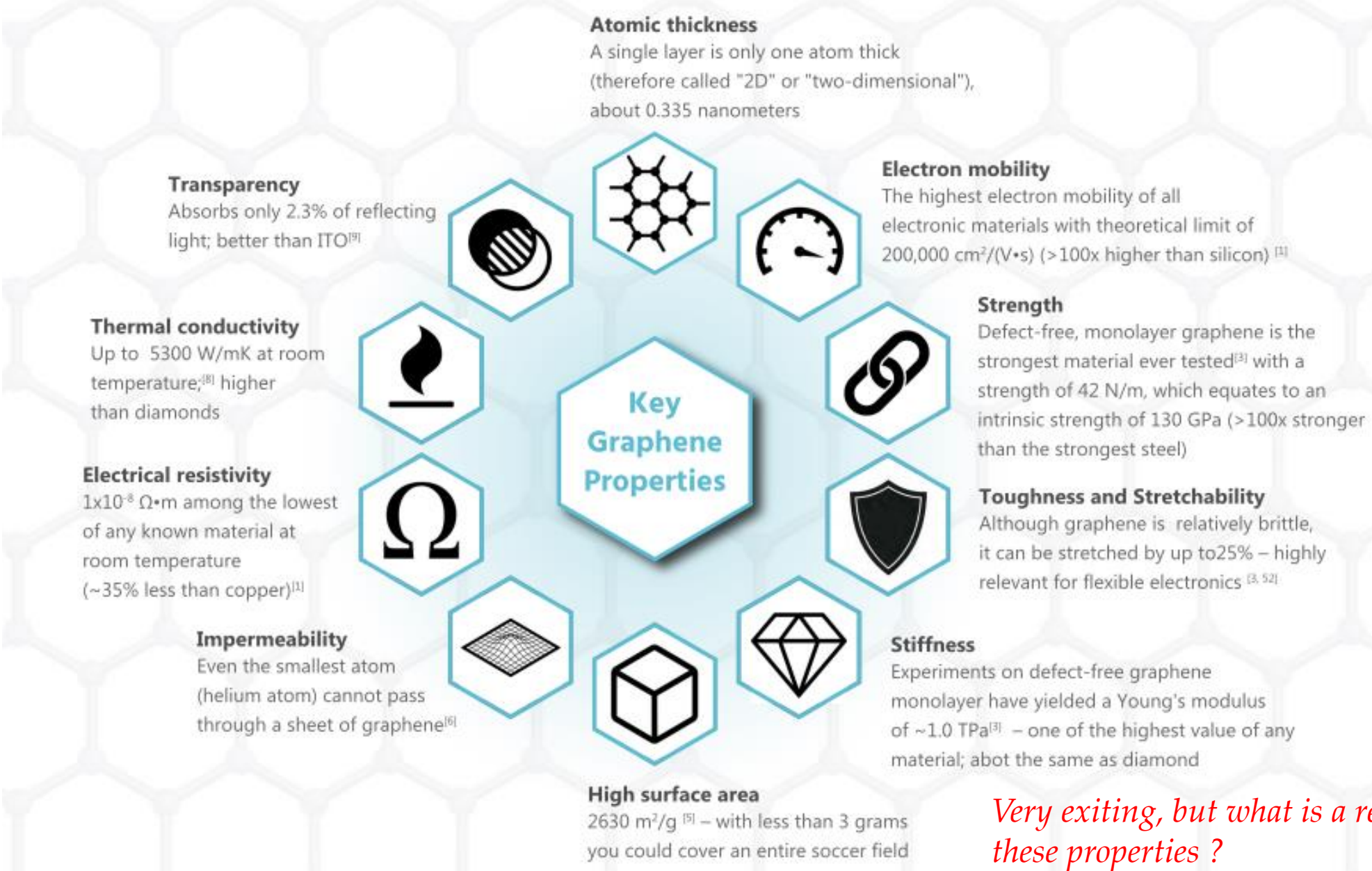
What is the advantage of graphene over graphite?

Strength, flexibility, electrical conductivity, exceptional optical properties have opened up new horizons for research in the field of high energy particle physics, as well as for electronic, optical and energy applications.

In graphene, each carbon atom is covalently bonded to three other carbon atoms. Thanks to the strength of its covalent bonds, graphene boasts great stability and, for example, very high tensile strength (the force you can stretch something before it breaks).

Carbon. Graphene. General





Very exciting, but what is a reason for these properties ?

The electrical and optical properties are due to a very special band structure

Five quite important facts

- 1. There are two types of electrons in solid carbon - electrons in the s- and p-shells associated with the atom, and electrons that have left the shells and transferred to the crystal*
- 2. The energy of all electrons is quantized*
- 3. According to Pauli exclusion principle, only two electrons with opposite spins can have the same energy in the same quantum system; when a system consists of many identical atoms, the individual energy levels of the electrons of individual atoms turn into energy bands*
- 4. The energy of electrons depends on the lattice. The spatial distribution of atoms is the reason for a certain symmetry of the intracrystalline electric field, which determines the allowed energy state of electrons.*
- 5. The grouping of these different energy levels for free and bound electrons forms energy bands.*

Valance band consists of energy levels of valence electrons.

Conduction band consists of energy levels of free electrons.

Forbidden energy gap is the energy gap between the valence band and the conduction band.

The valence band and conduction band overlap in the case of a conductor

There is a small band gap between the valence band and the conduction band in the case of semiconductors.

Between the valence band and the conduction band in the case of insulators there is a large band gap.

The wave vector (k-vector) determines the quasi-momentum and energy of an electron in a crystal

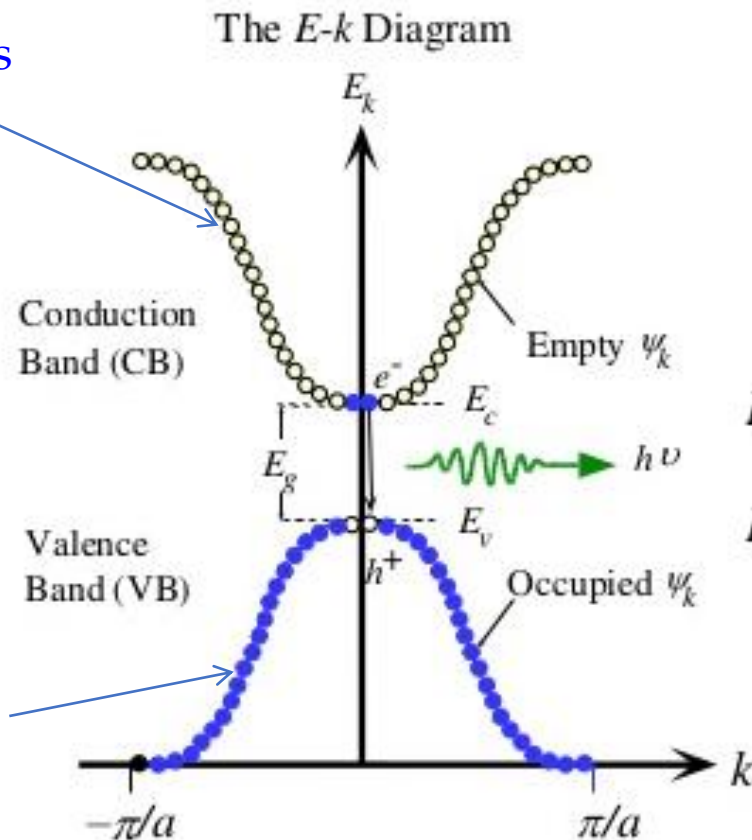
Quasimomentum

Empty cells

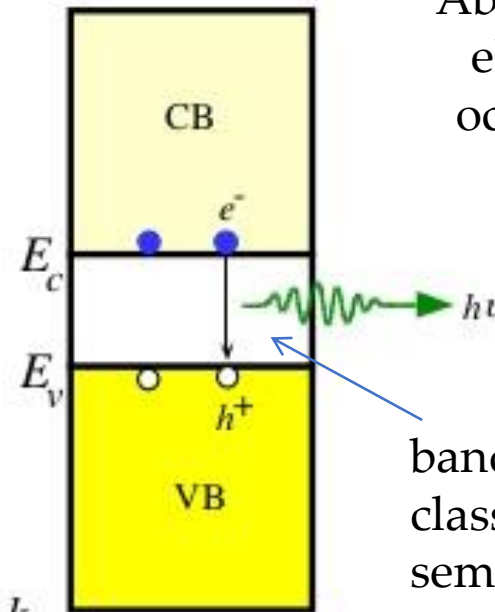
*kinetic energy
of the electron*

The space in which an electron exists is quantized and consists of separate cells, each of which can contain a maximum of two electrons with opposite spins at the same time.

Occupied cells



The Energy Band Diagram



Absorption or emission of electromagnetic waves occurs when an electron passes between cells

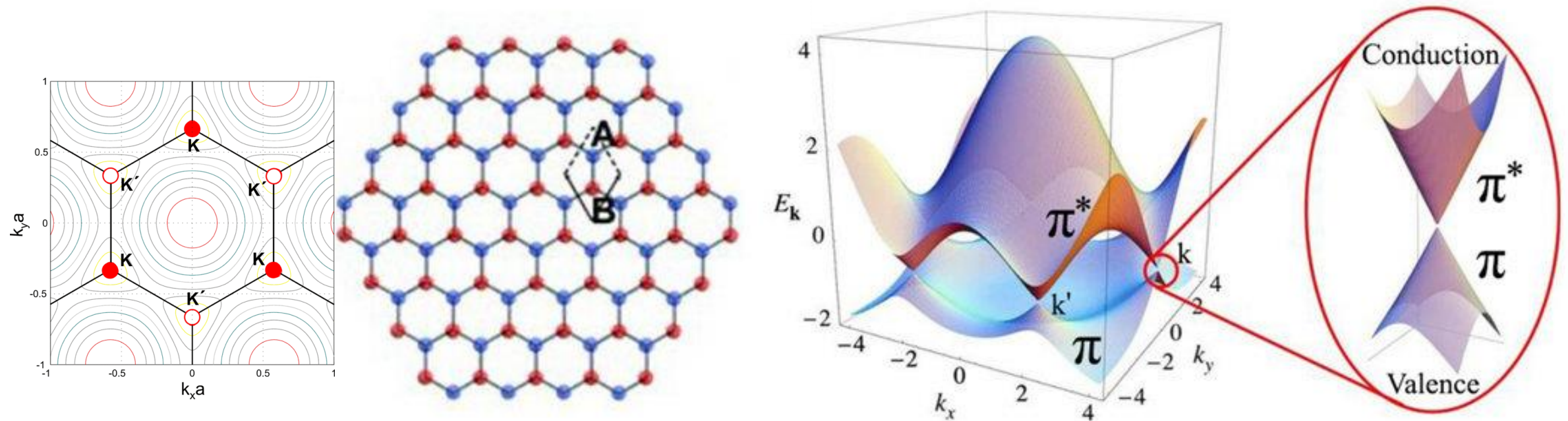
bandgap determines the class of material - conductor, semiconductor or insulator

Graphene has a special electronic structure:

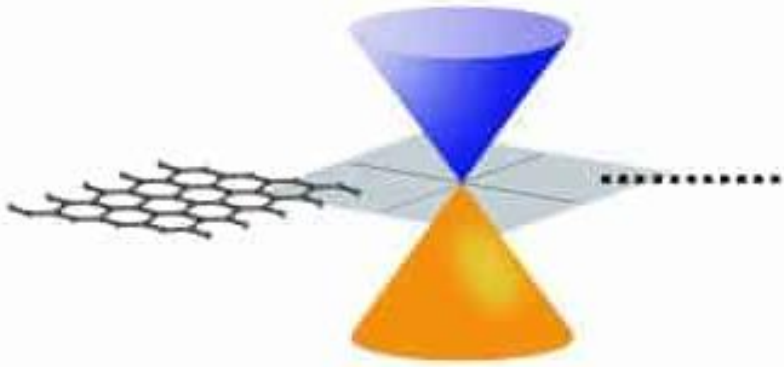
Each atom in a graphene sheet is connected to three nearest neighbors by σ -bonds and a delocalized π -bond, which contributes to the formation of a valence band that spans the entire sheet.

The valence band touches the conduction band, making graphene a semimetal with unusual electronic properties that are best described by massless relativistic particle theories.

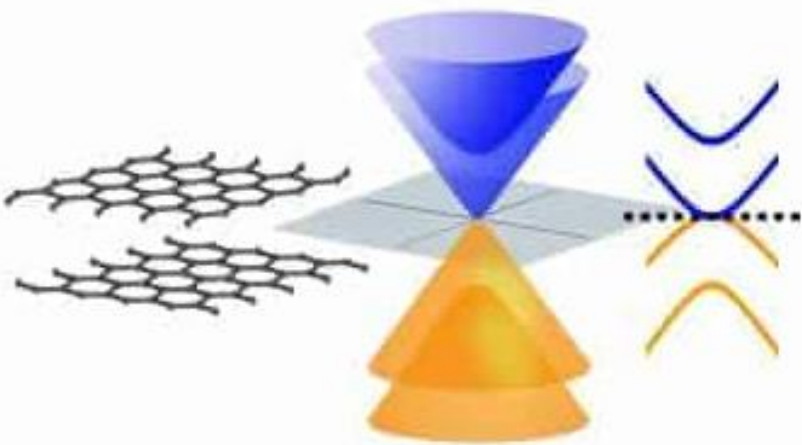
Charge carriers in graphene *show a linear rather than quadratic* dependence of energy on momentum.



Result: Graphene typically has a zero band gap, which is due to its massless electrons.



Quantum confinement effect is a restriction on the movement of an electron in a material to specific discrete energy levels rather than to quasi-continuum of energy bands when the length of a particle is reduced to the same order as *the wave packet (the condition is fulfilled for single and multilayer graphene)*



The difference in energy between filled states and the empty states depends on the size of graphene particles, so the band gap can be tuned (*band gap typically increases with decreasing particle size*)

The transition from single-layer to multilayer graphene makes it possible to vary the electronic properties of the material along with a change in the interlayer distance.

Yes, graphene can be multilayered – up to 10 layers

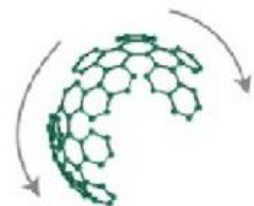
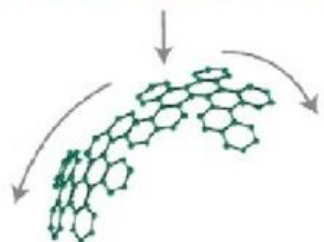
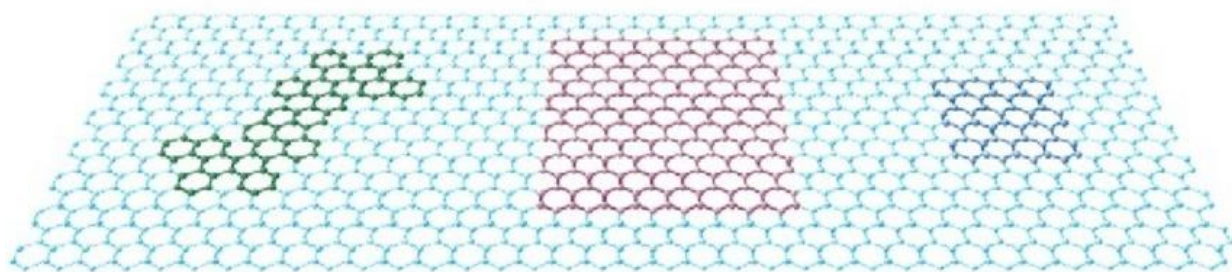
Carbon. Graphene as a building block

Graphene sheets are building blocks for other graphitic materials

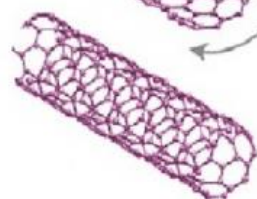
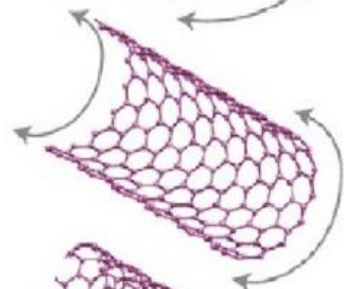
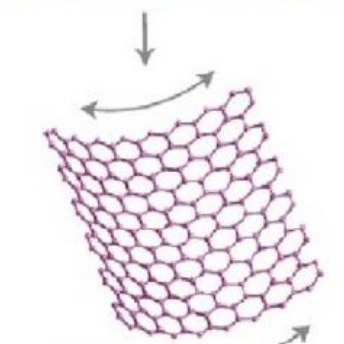
1. Graphene sheets stacked on top of each other make *3D graphite* (1 mm thick graphite contains about 3 million layers of graphene)
2. Graphene sheets rolled up make a *carbon nanotube*
3. Graphene sheets cutting and folding into a spherical shape make a *fullerene*

Yes, the properties of nanotubes and fullerenes are special and size-sensitive

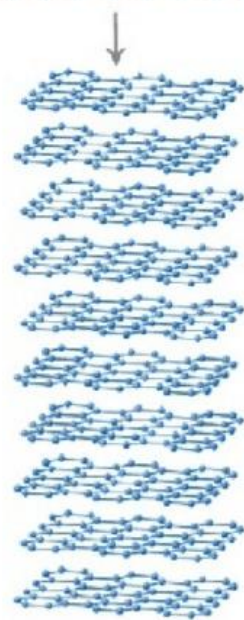
2D Graphene



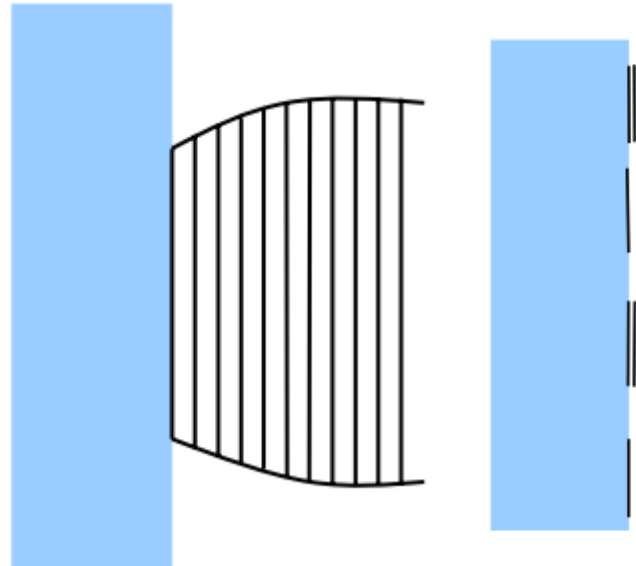
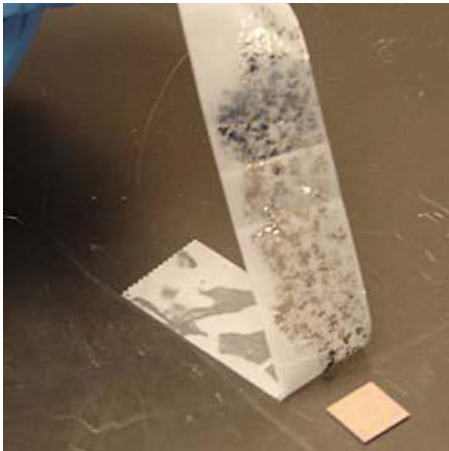
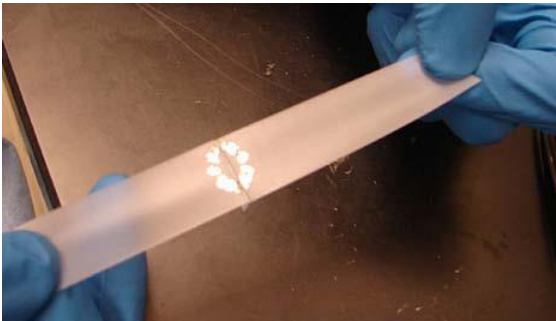
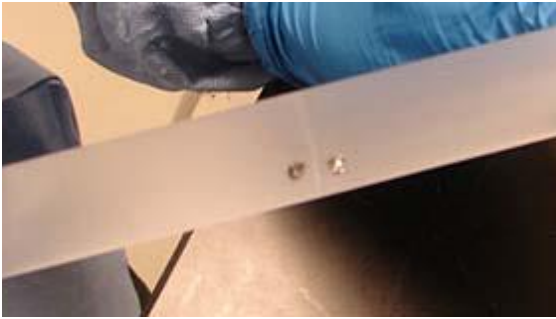
0D Fullerene



1D Carbon nanotubes

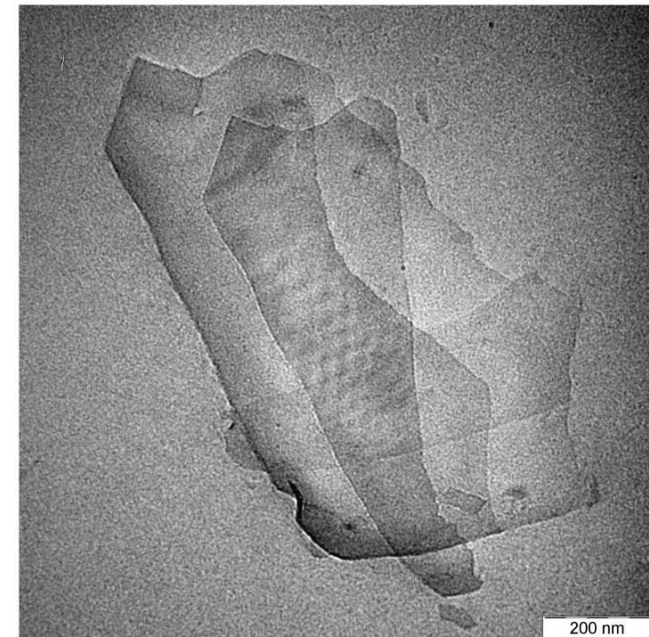


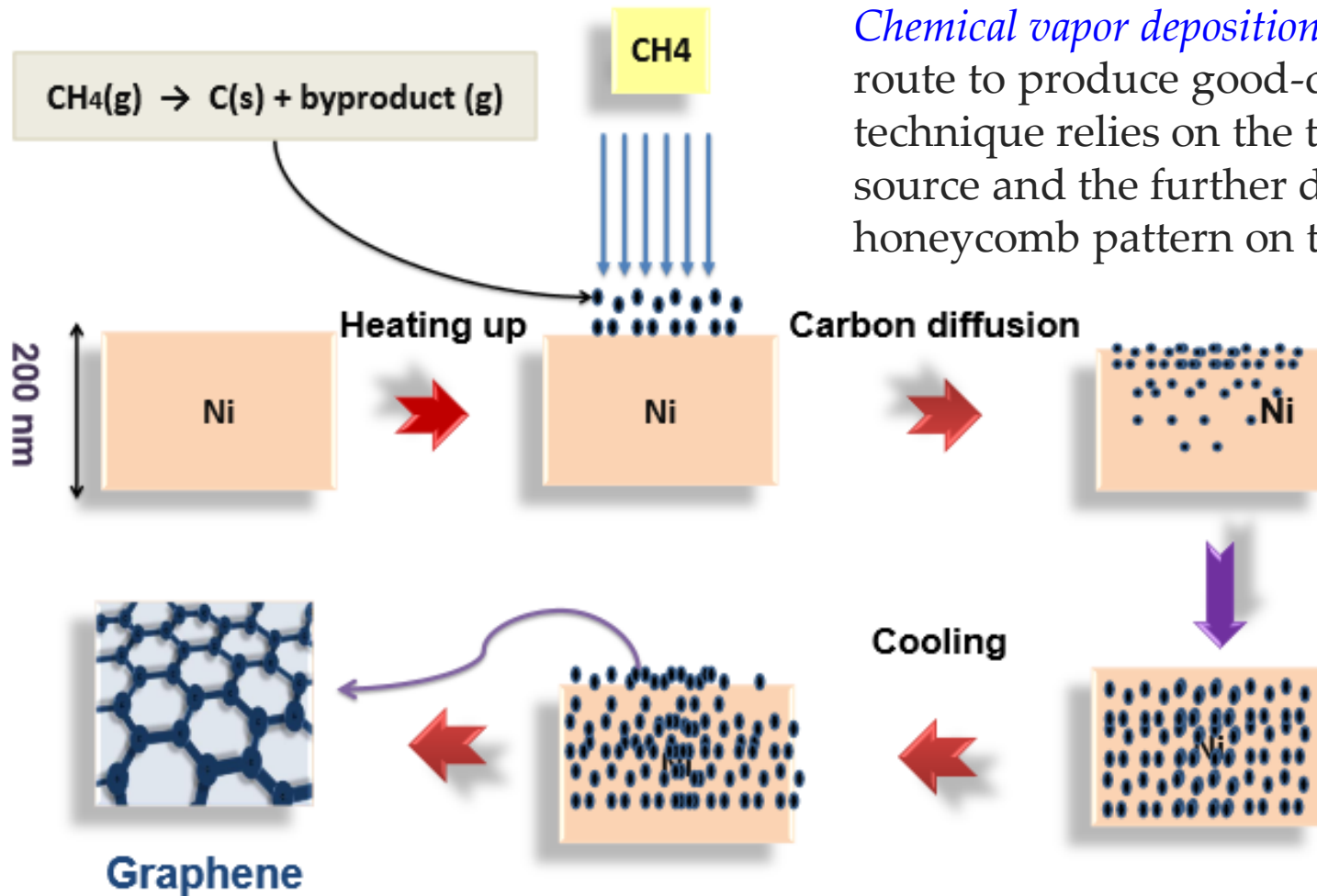
3D Graphite



In *micromechanical exfoliation method*, graphene is separated from a graphite crystal using adhesive tape. After peeling it off the graphite, multiple-layer graphene remains on the tape. By repeated peeling the multiple-layer graphene is cleaved into several flakes of few-layer graphene.

This simple method was applied by graphene inventors

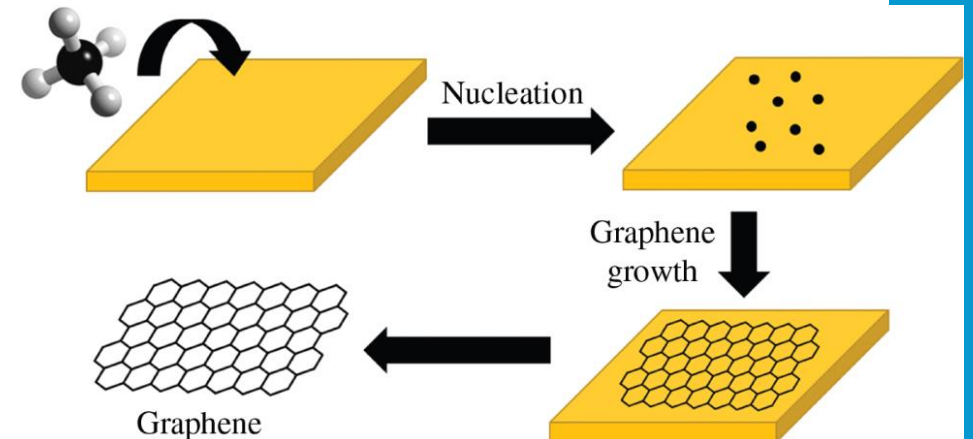




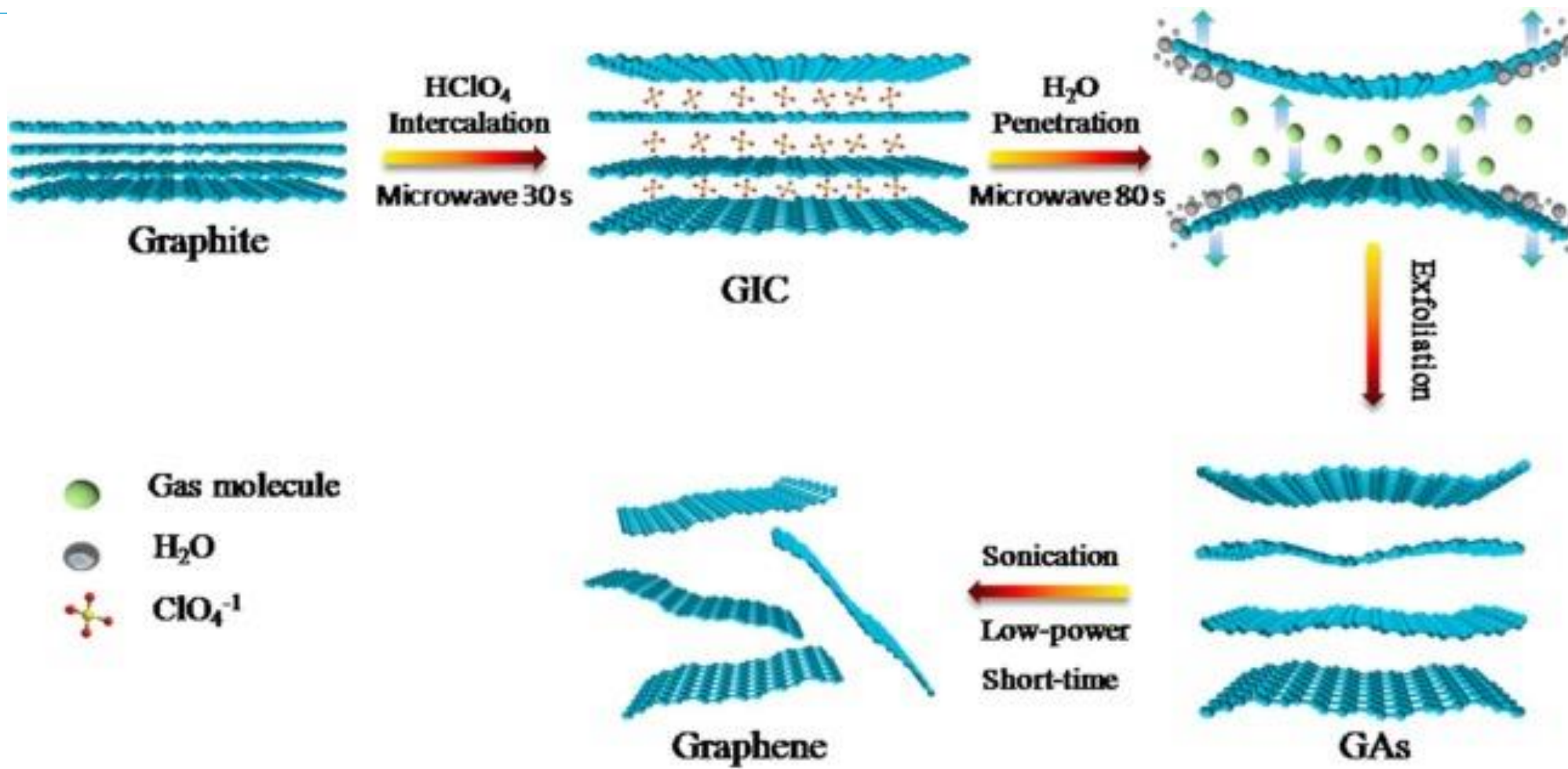
Chemical vapor deposition (CVD) represents a viable synthesis route to produce good-quality, large-area graphene films. The technique relies on the thermal decomposition of a carbon-rich source and the further deposition of carbon atoms in a honeycomb pattern on top of a metallic catalyst film.

Complicated and expensive, but scalable

Meanwhile, graphene costs less than \$0.1 per gram, which can be very profitable, with gold being around \$60-65 per gram.



Carbon. Graphene. Obtaining



Chemical exfoliation is a top-down approach to obtain graphene in dispersion from graphite using stages of intercalation, exfoliation, and sonification. The advantage of this manufacturing route is that the intercalation compounds spontaneously dissolve in polar solvents.

This method is very close to graphene oxide route

Contains epoxy, hydroxyl and carbonyl groups on the basal plane, and carboxylic groups on the edges.

Higher interlayer spacing than graphene, due to sp^3 carbons.

Higher ability to retain compounds.

Lower electron mobility compared to graphene.

Soluble in water.

Amphiphilicity.

Surface-functionalization capability and versatility.

Biocompatibility and ability to interact with biological cells and tissues.

Highly hydrophilic, forming stable aqueous colloids.

Substrate-deposition capability.

Convertible into a conductor.

Really cheap multifunctional material

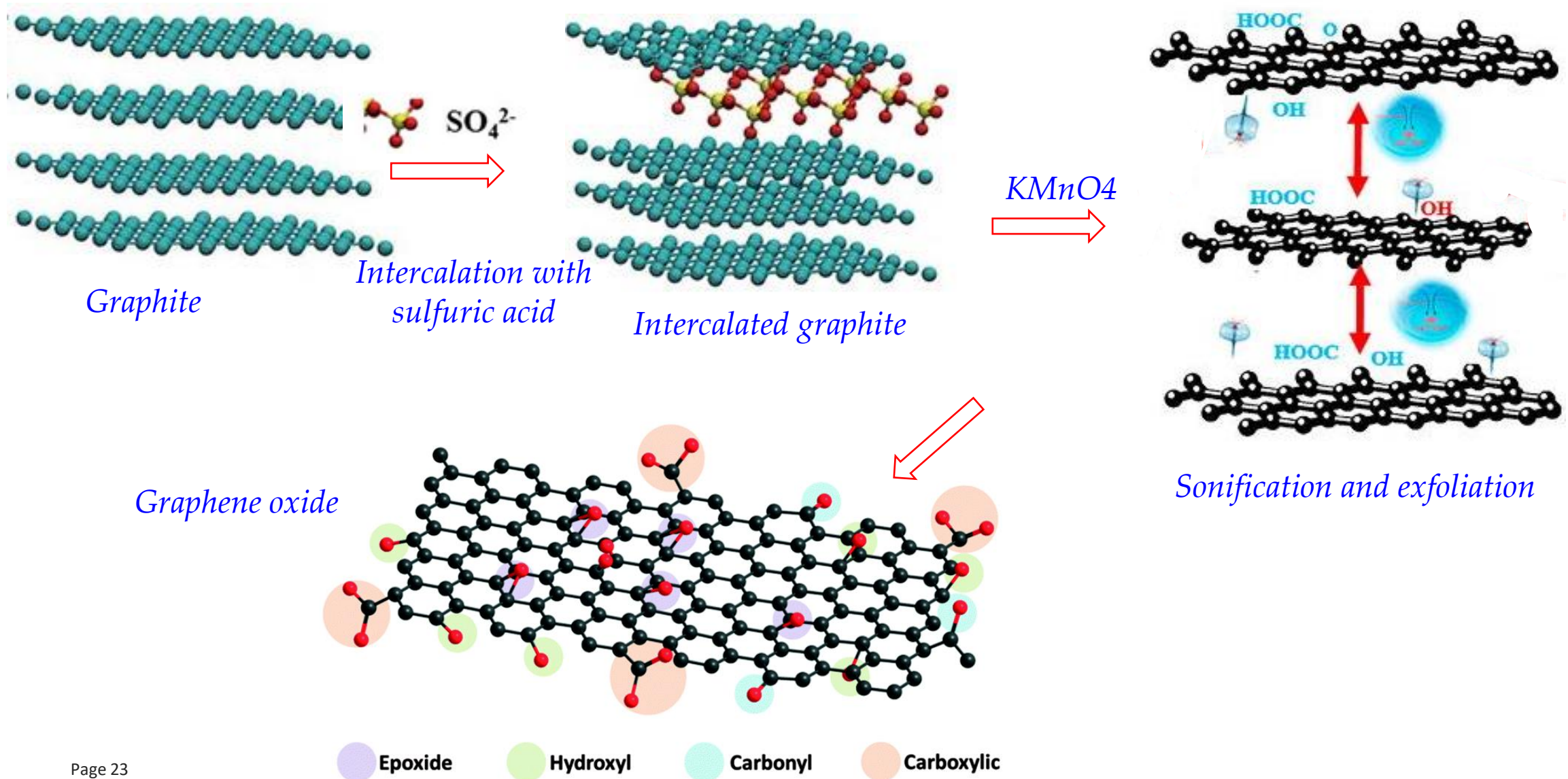
Possible functionalization points:

- Hydroxyl groups*
- Epoxy groups*
- Carboxylic groups*

Functional groups are arbitrarily located and randomly aggregated

Graphene Oxide





Carbon. Graphene Oxide. Obtaining

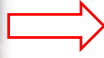
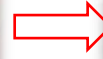
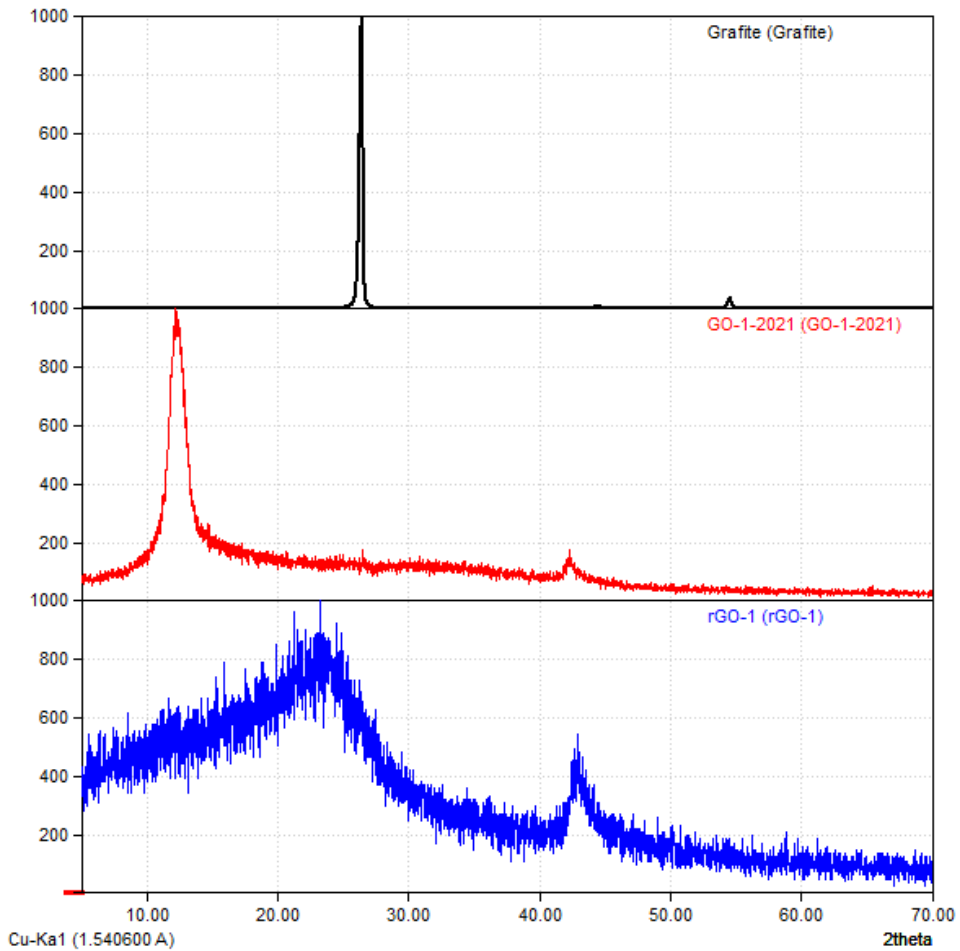
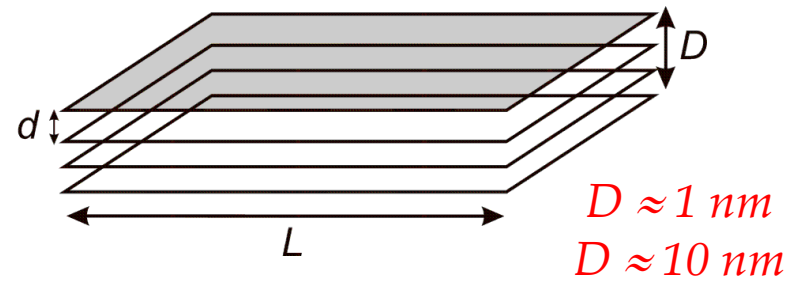
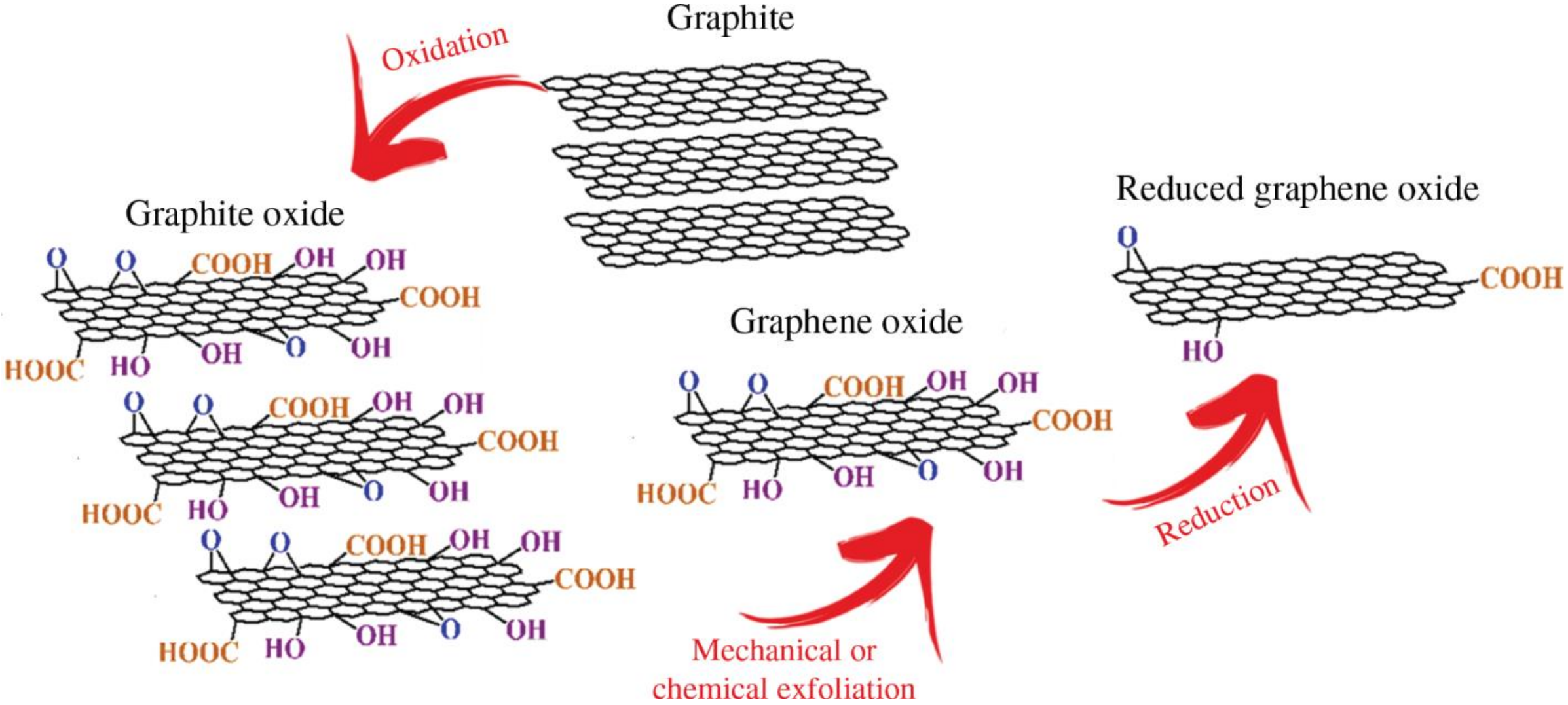


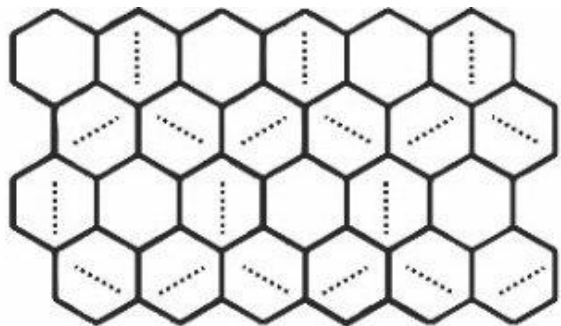
Photo from my student's work



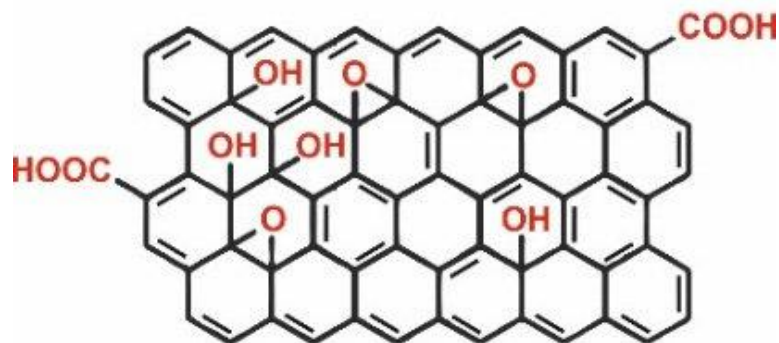


Schematic representation of the reduced graphene oxide production

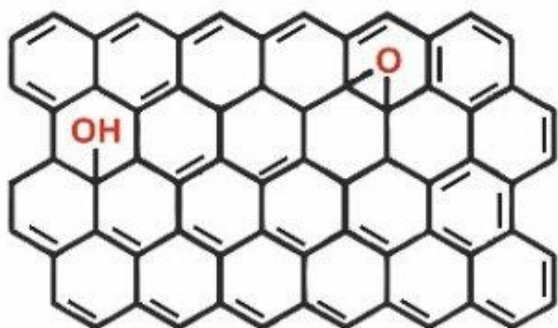
Carbon. Reduced Graphene Oxide



graphene (G)



graphene oxide (GO)



reduced graphene oxide (rGO)

Reduced graphene oxide (rGO) is the form of GO that is processed by chemical, thermal and other methods in order to reduce the oxygen content, while graphite oxide is a material produced by oxidation of graphite which leads to increased interlayer spacing and functionalization of the basal planes of graphite.

One of the most important differences between GO and rGO is the electrical conductivity of these materials. While GO shows insulating or semi-conducting behavior, rGO shows excellent electrical conductivity which is almost as good as graphene.

The main disadvantage of the rGO is defect structure

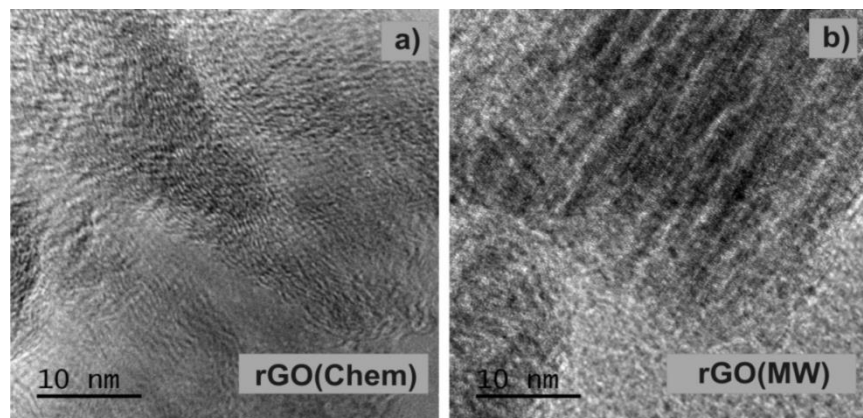
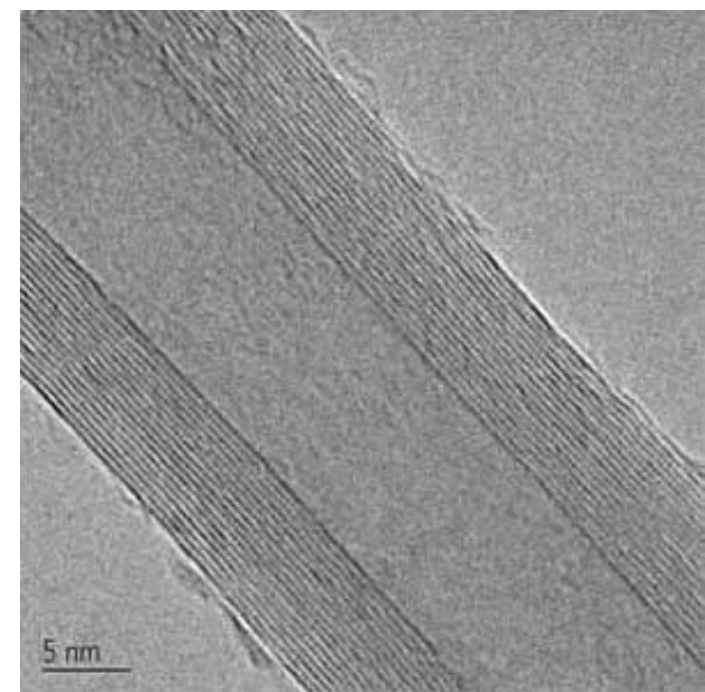
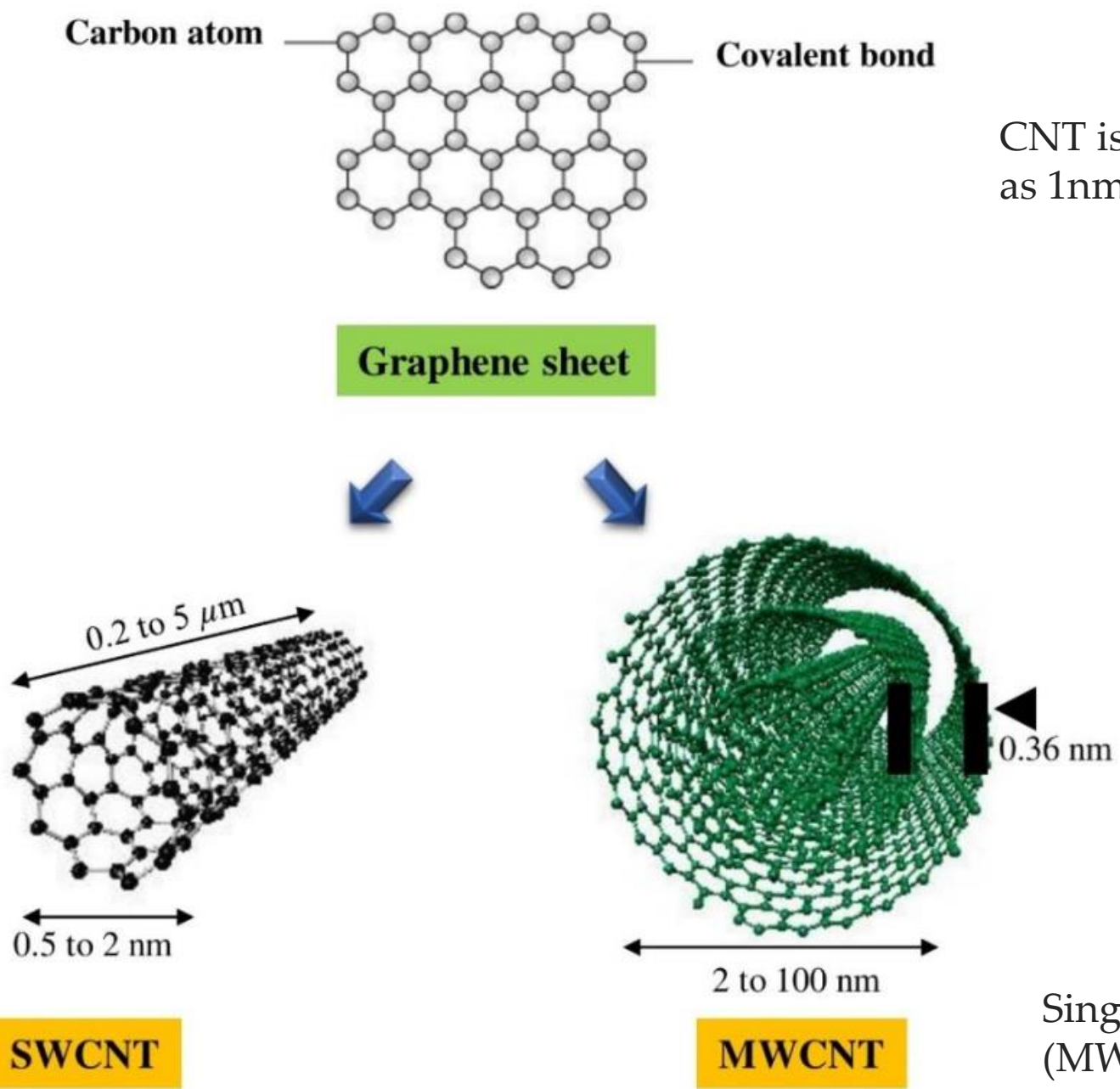


Photo from my student's work

Carbon. Nanotubes. General

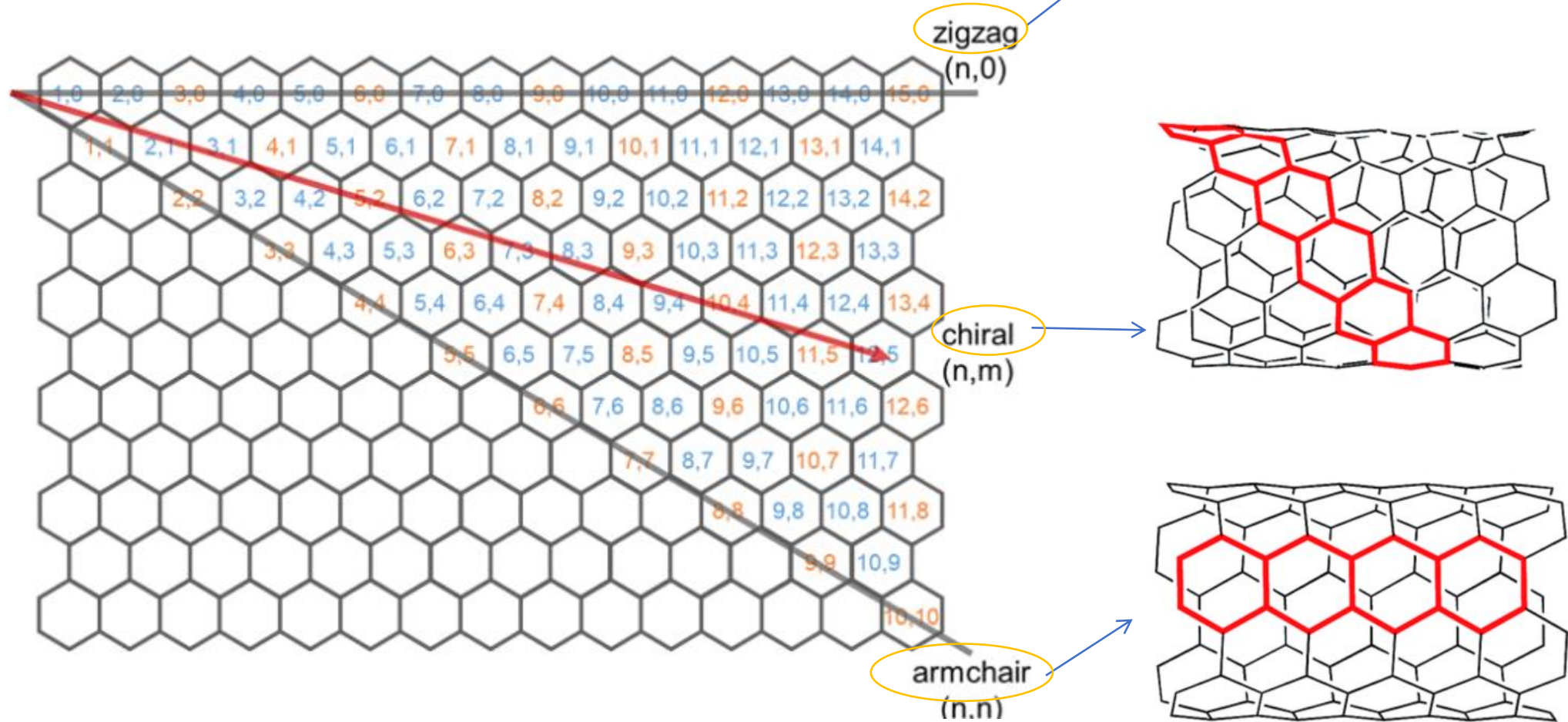
CNT is a tubular form of carbon with diameter as small as 1nm (single wall). Length: few nm to microns.



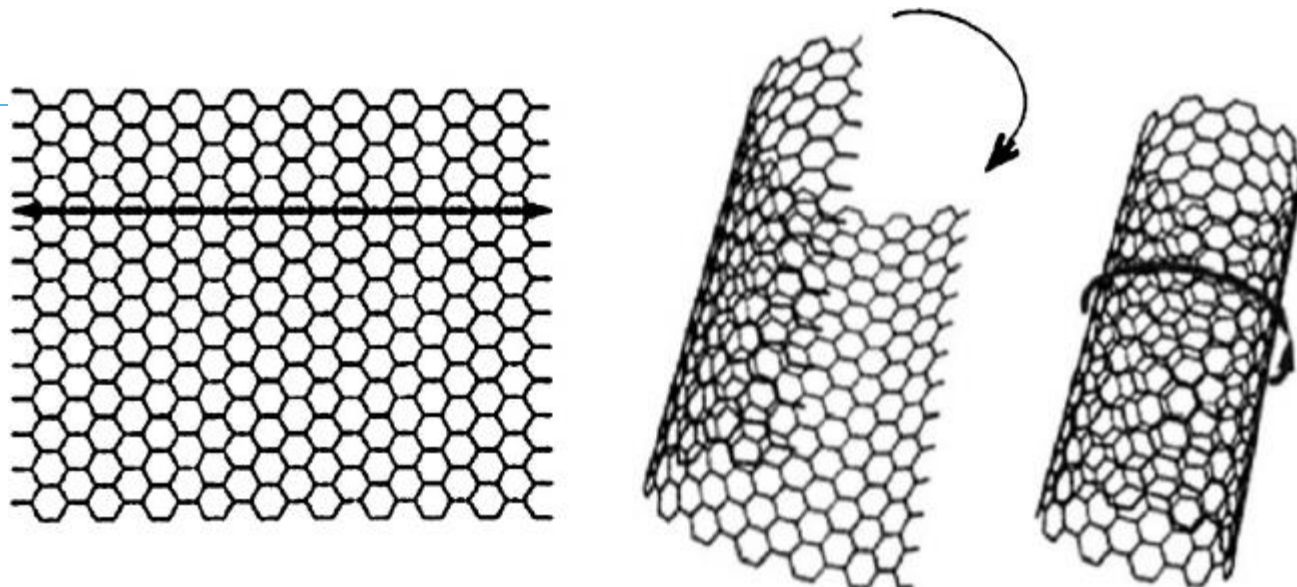
TEM image of an individual MWCNTs

Single Wall CNT (SWCNT) and multiple Wall CNT (MWCNT) can be metallic or semiconducting depending on their geometry

A CNT is characterized by its Chiral Vector: $C_h = na + mb$,



Carbon. Nanotubes. Reason



CNT is configurationally equivalent to a two-dimensional graphene sheet rolled into a tube.

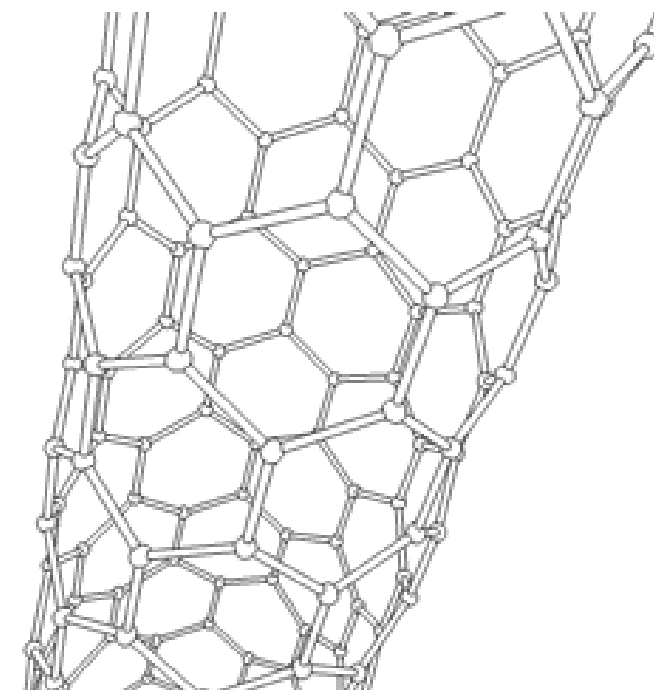
What is the cause of carbon nanotube formation?

Nanotube/Fullerene combines $sp^2 + sp^3$ hybridized carbon atoms. Finite size of graphene layer has dangling bonds (an unsatisfied valence of an immobilized atom).

These dangling bonds correspond to high energy states.

Both the elimination of dangling bonds and an increase in the strain energy lead to a decrease in the total energy.

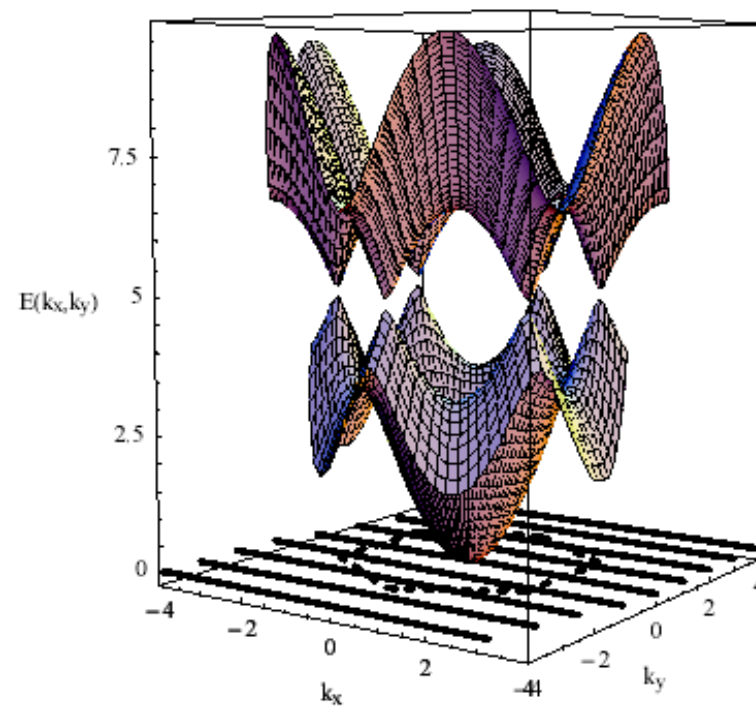
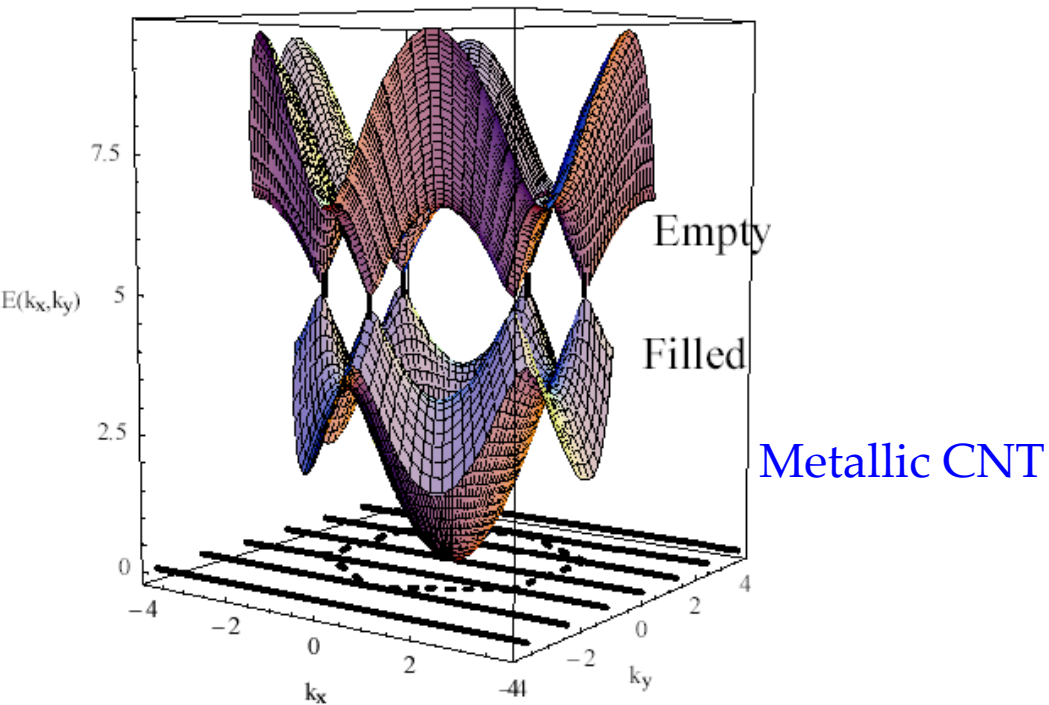
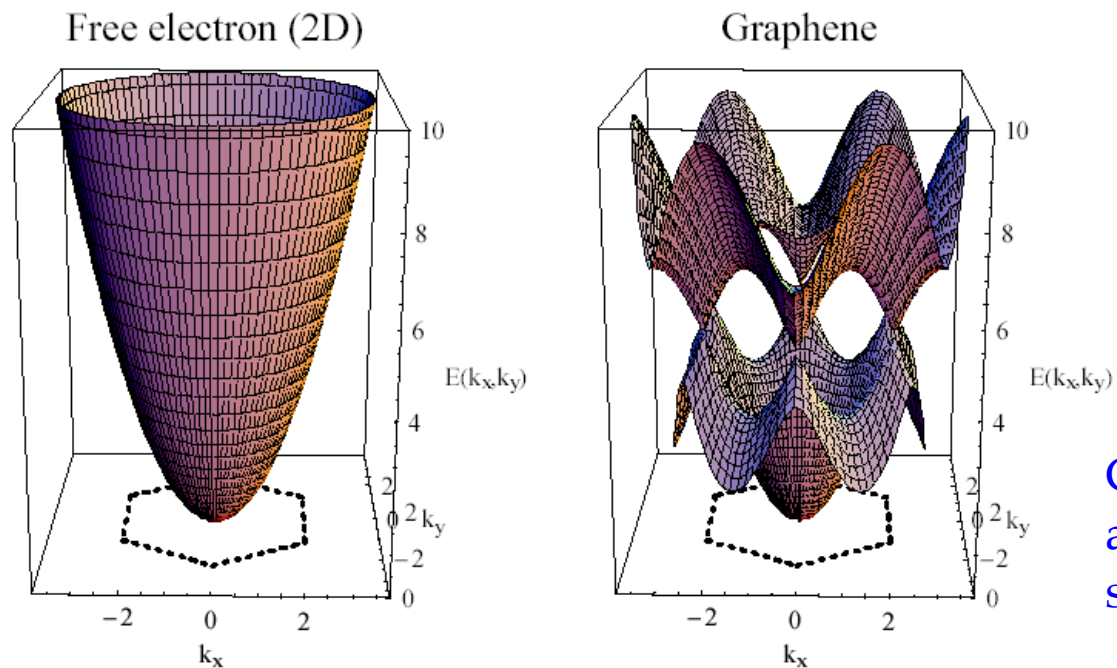
*Why does something happen spontaneously?
Because it leads to a decrease in the energy of the system.*



Carbon. Nanotubes. Electronic properties

Band structure of CNTs in comparison with band structures of free electrons and graphene

Changing the type of geometry (zigzag, chiral, armchair) makes it possible to tune the CNT band structure from metallic to semiconductor.



- The strongest and most flexible molecular material because of C-C covalent bonding and seamless hexagonal network architecture
- Young's modulus of over 1 TPa vs 70 GPa for Aluminum, 700 GPa for C-fiber
 - strength to weight ratio 500 times > for Al; similar improvements over steel and titanium; one order of magnitude improvement over graphite/epoxy
- Maximum strain ~10% much higher than any material
- Thermal conductivity ~ 3000 W/mK in the axial direction with small values in the radial direction

CNT electronics

Carrier transport is 1-D.

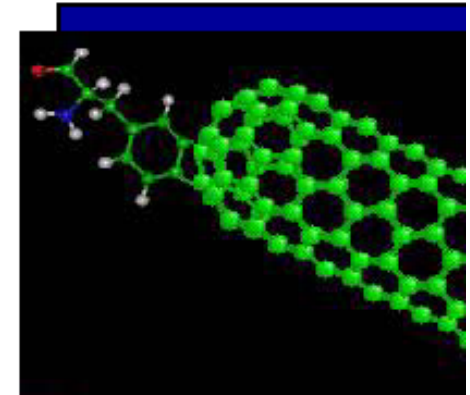
All chemical bonds are satisfied \Rightarrow CNT electronics is not obligated to use SiO_2 as an insulator.

High mechanical and thermal stability and resistance to electromigration \Rightarrow Current densities up to 10^9 A/cm^2 can be sustained.

The diameter is controlled by chemistry, not fabrication.

Both active devices and interconnects can be made from semiconductor and metallic nanotubes.

- Electrical conductivity six orders of magnitude higher than copper
- Can be metallic or semiconducting depending on chirality
 - 'tunable' bandgap
 - electronic properties can be tailored through application of external magnetic field, application of mechanical deformation...
- Very high current carrying capacity
- Excellent field emitter; high aspect ratio and small tip radius of curvature are ideal for field emission
- Can be functionalized



Arc Discharge (close to Laser Ablation method)

Involves condensation of C-atoms generated from evaporation of solid carbon sources.

Temperature ~ 3000-4000K, close to melting point of graphite.

Both produce high-quality SWNTs and MWNTs.

MWNT: 10's of μm long, very straight & has 5-30 nm diameter.

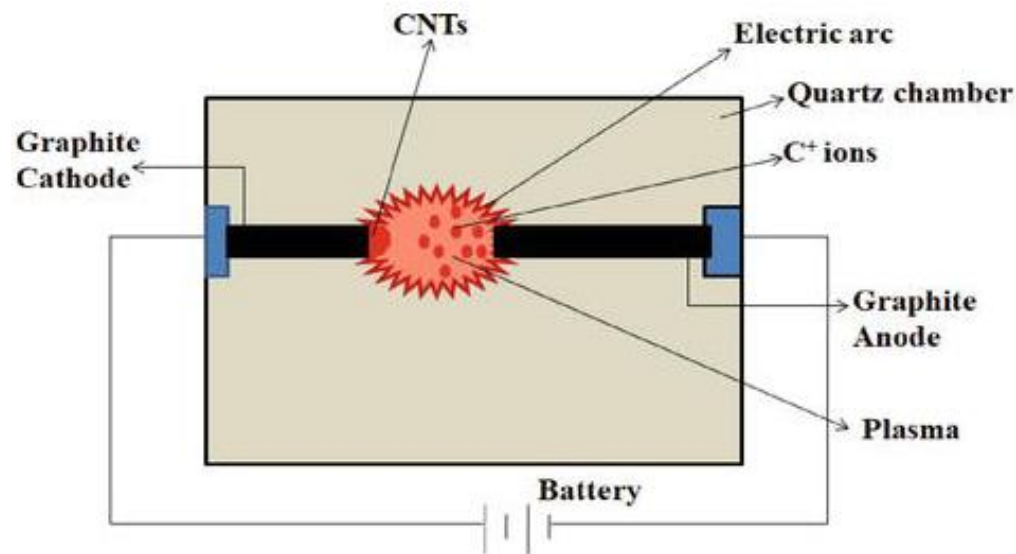
SWNT: needs metal catalyst (Ni, Co, etc.).

Produced in form of ropes consisting of 10's of individual nanotubes close packed in hexagonal crystals.

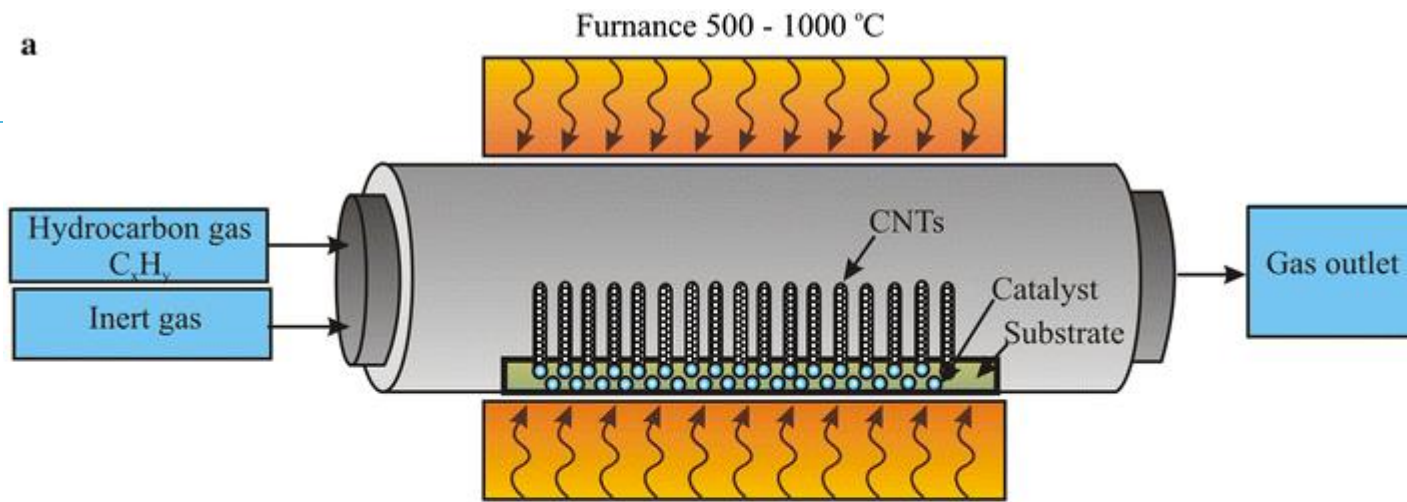
Electric arc discharge method:

A potential of 20–25 V is applied across the pure graphite electrodes separated by 1 mm distance and maintained at 66.7 kPa pressure of flowing helium gas filled inside the quartz chamber. When the electrodes are made to strike each other under these conditions, it produces an electric arc. The energy produced in the arc is transferred to the anode, which ionizes the carbon atoms of pure graphite anode and produces C^+ ions and plasma forms (atoms or molecules in vapor state at high temperature).

These positively charged carbon ions move towards cathode, get reduced and deposited and grow as CNTs on the cathode. As the CNTs grow, the length of the anode decreases, but the electrodes are adjusted and always maintain a gap of 1 mm between the two electrodes.

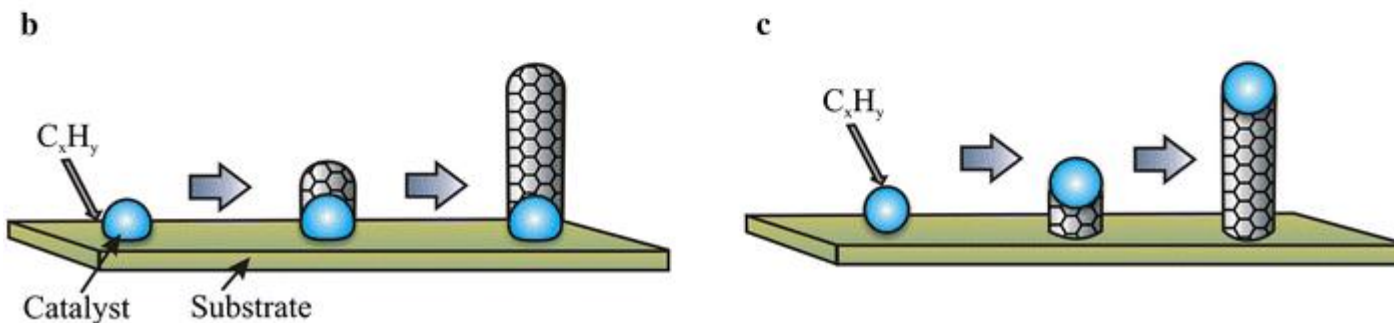


Carbon. Nanotubes. Synthesis



Chemical Vapor Deposition:

The chemical vapor deposition method is to cleave a carbon atom-containing gas continuously flowing through the catalyst nanoparticle to generate carbon atoms and then generate CNTs on the surface of the catalyst or the substrate.



Hydrocarbon gas + Fe/Co/Ni catalyst at 550-1000°C

Steps:

- Dissociation of hydrocarbon.
- Dissolution and saturation of C atoms in metal nanoparticle.
- Precipitation of Carbon.

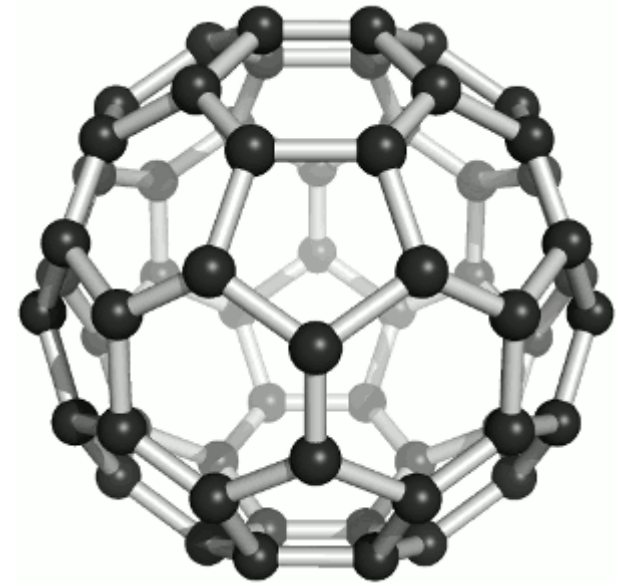
Fullerenes consist of 20 hexagonal and 12 pentagonal rings, which form the basis of a closed framework structure with icosahedral symmetry.

Each carbon atom is bonded to three others and is sp^2 hybridized. The C_{60} molecule has two bond lengths - 6:6 ring bonds can be considered "double bonds" and are shorter than the 6:5 bonds.

C_{60} is not "superaromatic" as it tends to avoid double bonds in the pentagonal rings, resulting in poor electron delocalization.

As a result, C_{60} behaves like an electron deficient alkene, and reacts readily with electron-rich species.

The geometry and electronic bonding factors in the structure determine the stability of the molecule.



Physical properties of C_{60} :

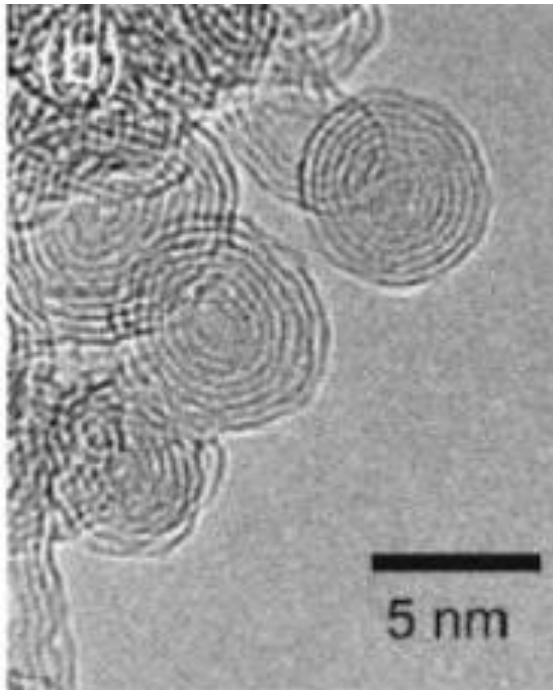
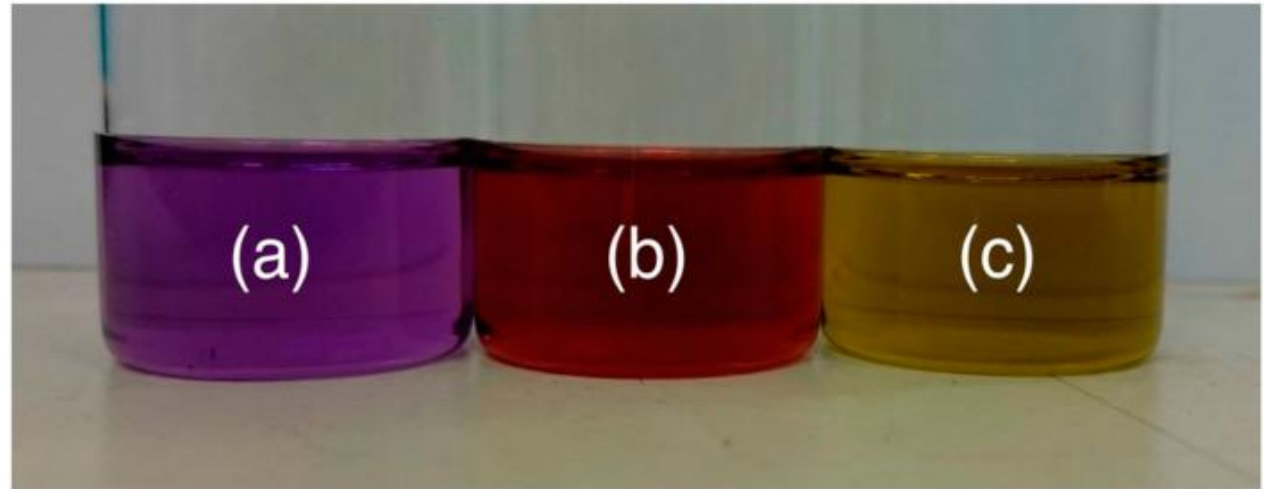
Density: 1.65 g cm^{-3}

Standard heat of formation: $9.08 \text{ kcal mol}^{-1}$

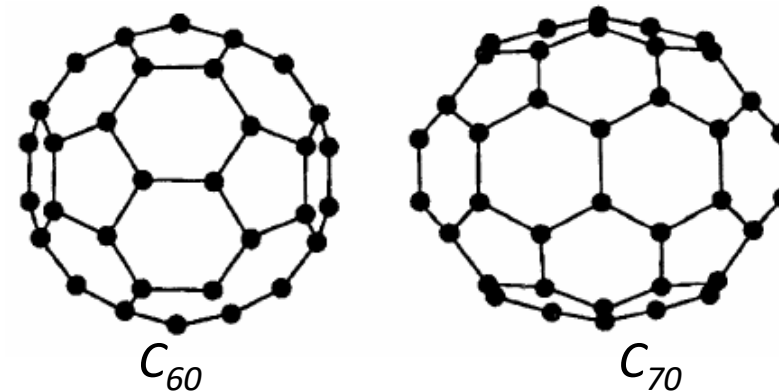
Boiling point: sublimes at 800 K

Resistivity: $10^{14} \text{ ohms m}^{-1}$

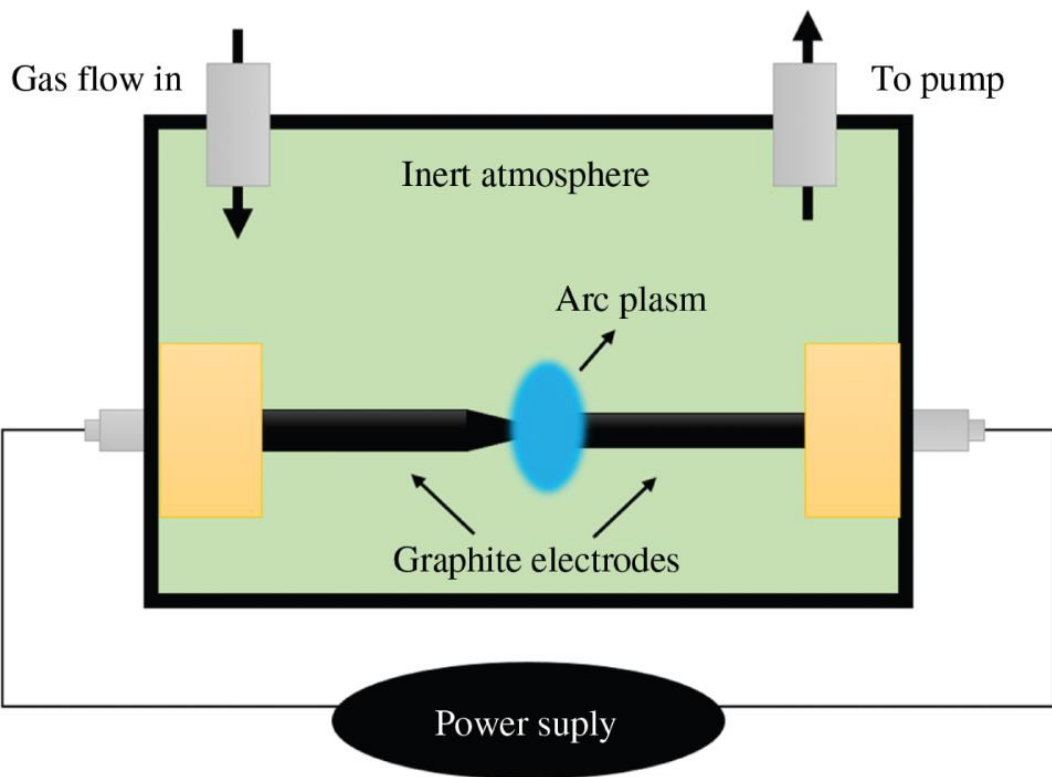
Crystal form: hexagonal cubic



The absorption spectra of the fullerenes change as the size of the conjugated system increases: with slight variations depending on solvent, solutions of C_{60} are an intense purple color (a), C_{70} red like wine (b) and C_{84} a green-yellow (c) (all solutions are in toluene). The gap between the highest occupied molecular orbital and the lowest unoccupied molecular orbital decreases with increasing cage size leading to optical absorptions of lower energy, i.e., longer wavelength

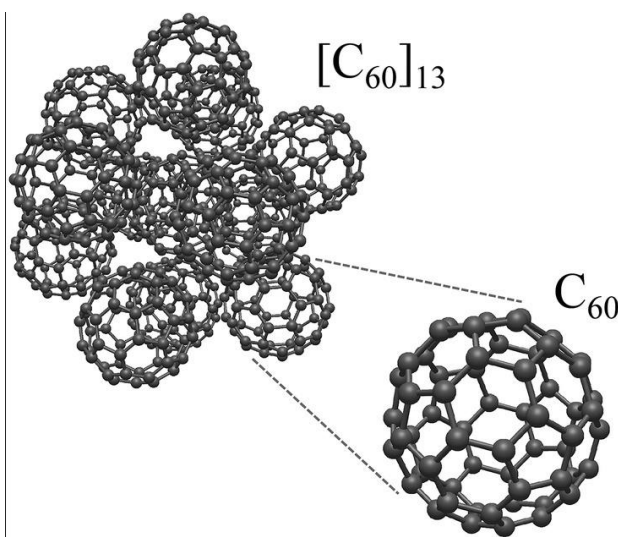


Carbon. Fullerenes. Synthesis



Electric arc discharge method for the production of fullerenes is based on the evaporation of graphite electrodes in a low-pressure helium atmosphere by passing an electrical current through the electrodes, resulting in an arc that produces carbon black containing fullerenes.


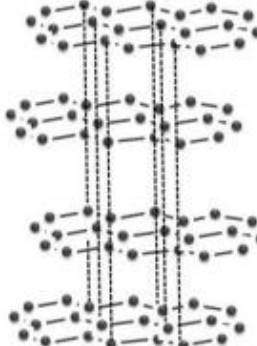


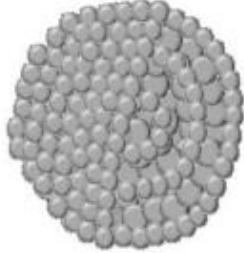
These methods usually lead to the production of various fullerenes in various mixtures and other forms of carbon. So, the fullerenes are mainly extracted from the soot using appropriate organic solvents.



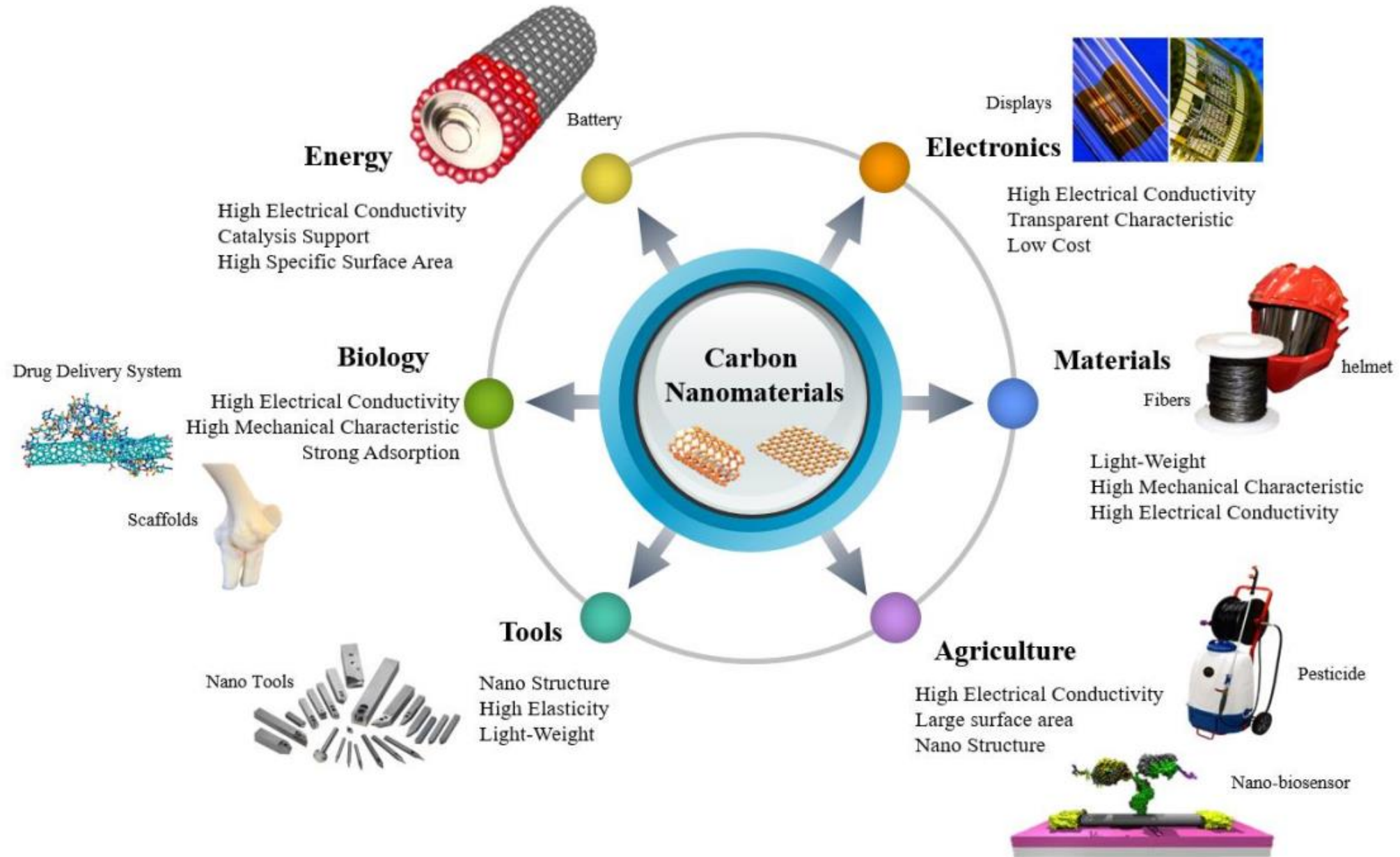
During the synthesis, not individual fullerenes are obtained, but their aggregates.

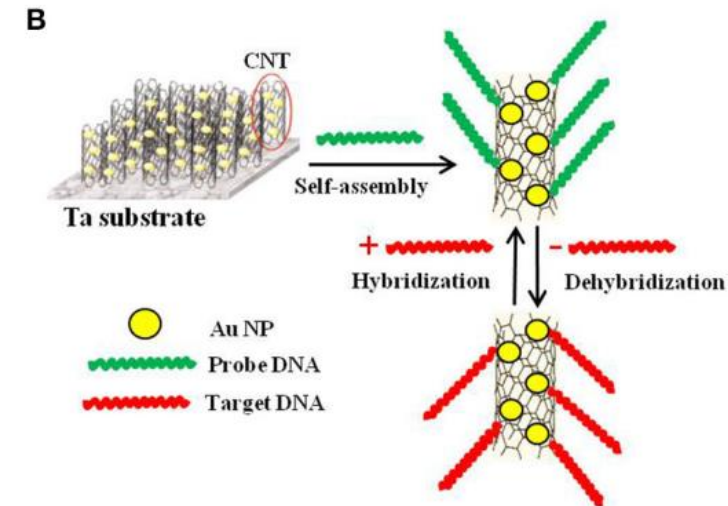
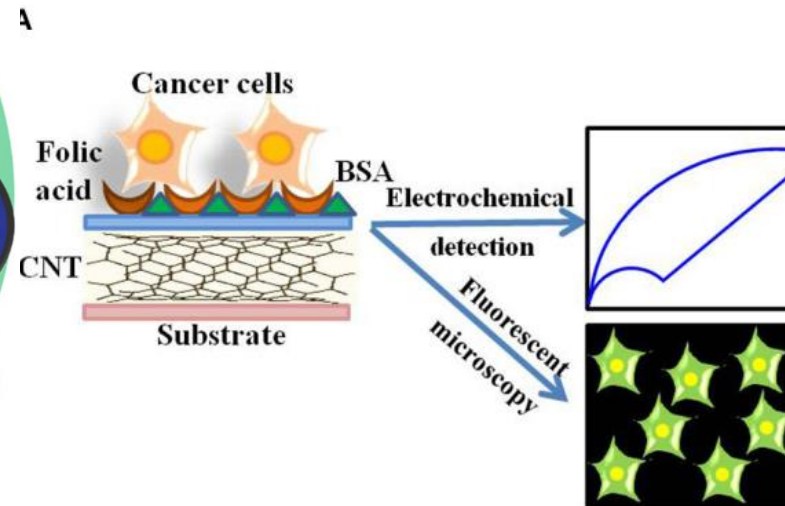
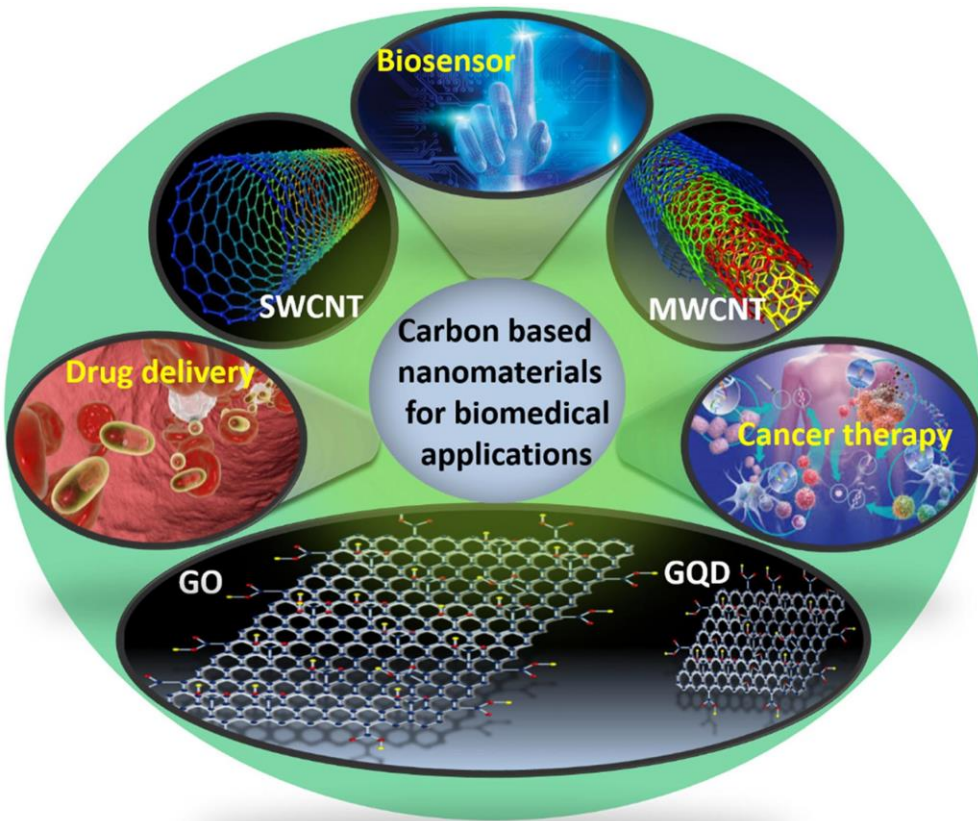
The reason is that the system reduces the excess surface energy, which is proportional to the surface area of the material.

The specific surface area can be very large for carbon nanomaterials - up to 2500 m²/g for *graphene*, up to 700 m²/g for *fullerenes* and up to 3500 m²/g for *microporous* carbon

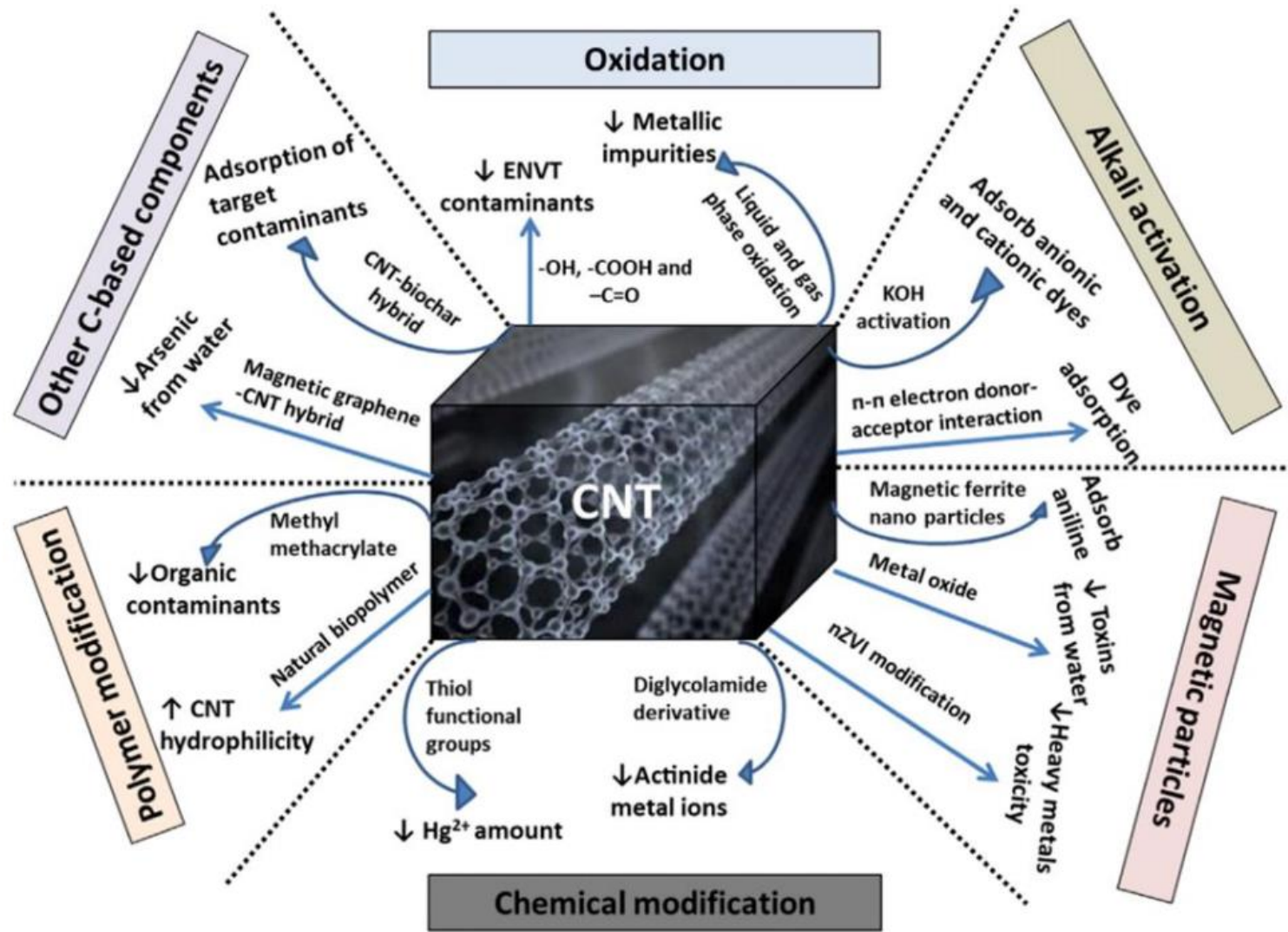
Different types of carbon nanomaterials (CNMs)	Carbon nano tubes	Graphene	Fullerene	Carbon dots	Carbon nano onions
Structure					
Size (nm)	1-100	1-100	0.4-1.6	1-10	3-100
Dimension	1	2	0	0	0
Density (g cm ⁻³)	1.3-2	2.26	1.72	1.0	1.9-3.3
Specific surface area	370-1600	2675	42-85	857	840
Specific capacitance (F g ⁻¹)	2-200	31-1046	—	95	45-334
Key references	34-37	38 and 39	40 and 41	42-44	45-47

Different CNMs and their physical and chemical properties

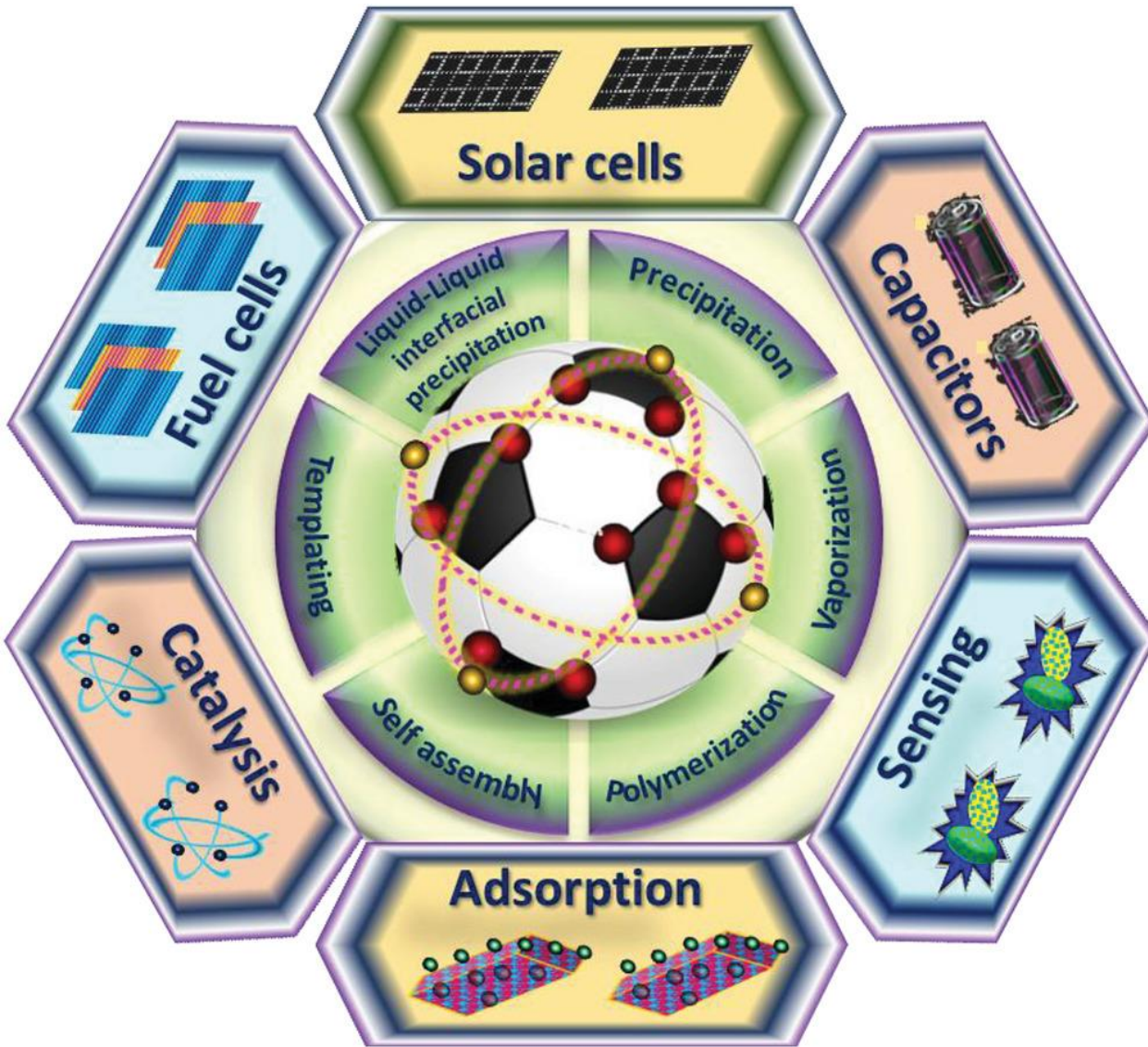




Electrochemical and Electronic CNT biosensors for Cancer Detection. (A) Schematic illustration of the folic acid-targeted cytosensing strategy for an enhanced electrochemical detection of cancer cells using polydopamine-coated carbon nanotubes. (B) Schematic representation of an electrochemical DNA biosensor for cancer detection based on gold nanoparticles/aligned CNTs.

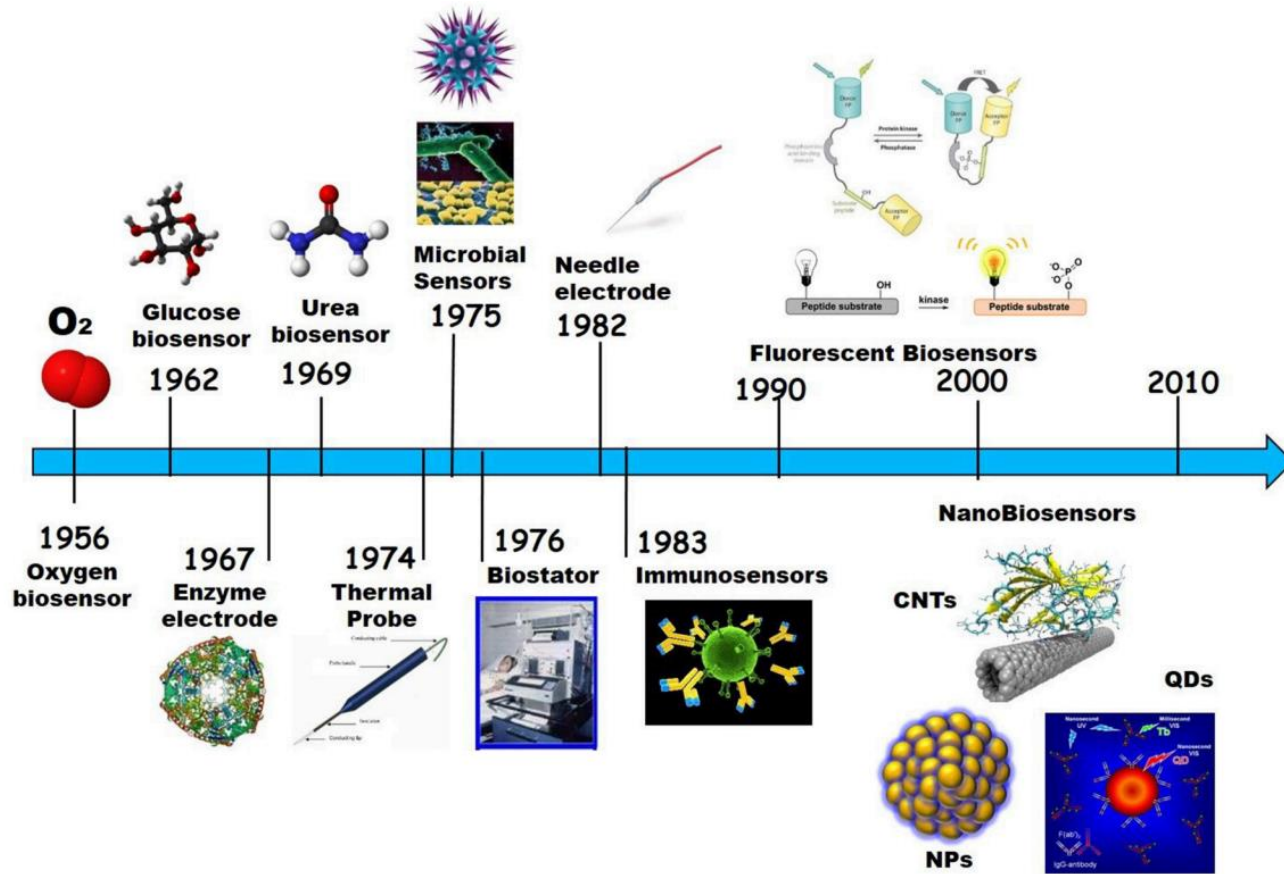


Schematic diagram representing different modification processes of CNTs for contaminant removal from water and wastewater (C: carbon; CNT: carbon nanotube; ENVT: environmental; Hg: mercury; KOH: potassium hydroxide)



There are many fields of practical application of fullerenes, such as materials for solar cells, fuel cells and supercapacitors, materials for adsorption, gas sensors and catalysis. But the most attractive is the biomedical application of fullerenes for drug and gene delivery.

Currently, most of the studies on biological applications of CNTs-based and other carbon allotropes (graphene, fullerene) as biomaterials are focused on an approach to continuous interactions with living cells and tissues.



Biosensor development timeline

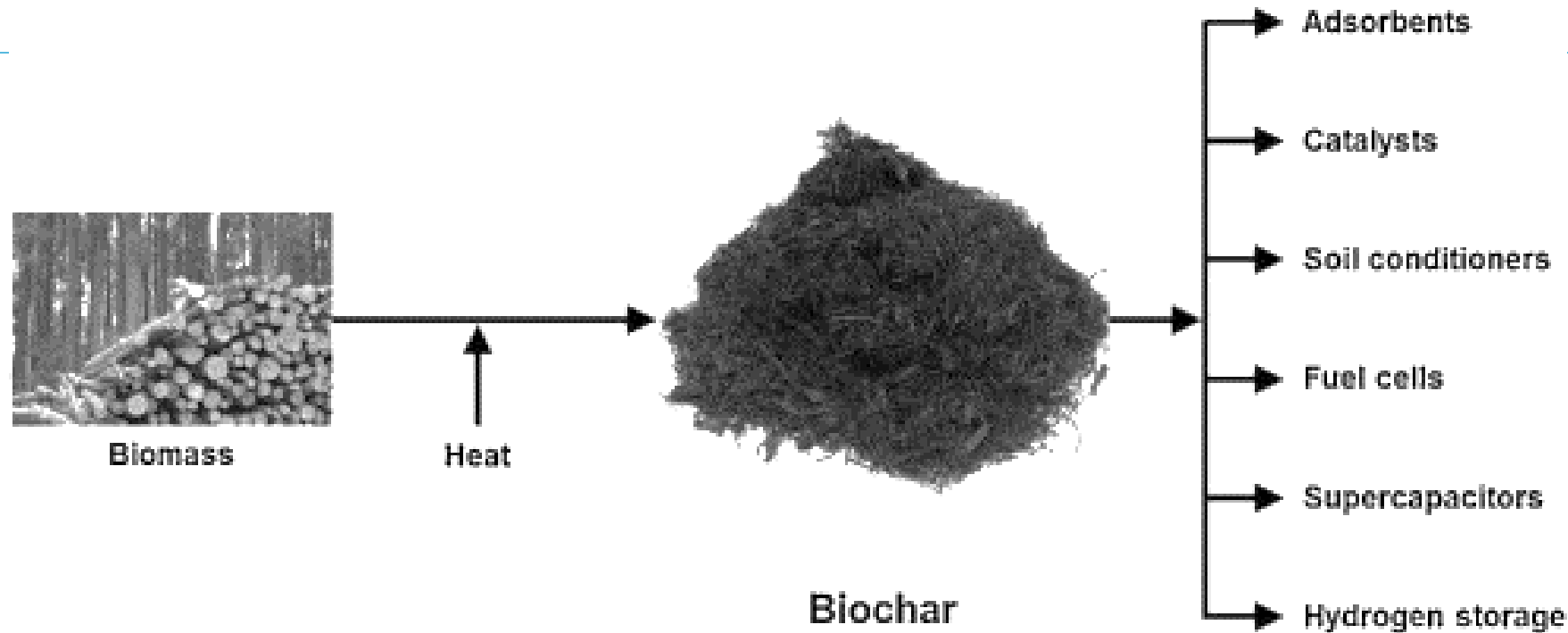




Carbon fiber shield

Besides polyethylene, carbon materials can also be used to provide shielding from cosmic radiation. A full-carbon shield was tested by bombardment with iron nuclei at 1 GeV/nucleon and showed the second-best dose reduction after polyethylene, before aluminum and lead.

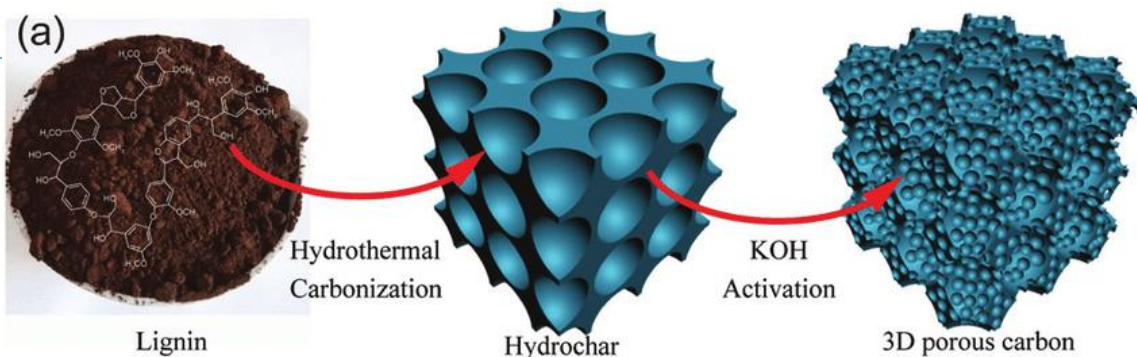
Utilizing polyethylene as the low atomic number (low-Z) substance within this composite necessitates its placement at the outermost layer. This positioning allows it to initially come into contact with incoming radiation. Consequently, a multi-layer composite comprising polyethylene and graphite can be constructed, resulting in enhanced radiation shielding. This assertion is supported by simulations conducted using a particle transport simulation code, which accounts for a variety of particles including solar particles, cosmic rays, protons, and electrons, all within the context of a highly elliptical orbit.



Biochar is produced by burning organic material in a controlled pyrolysis (burning without oxygen). The energy or heat generated during pyrolysis can be collected and used as clean energy. Biochar is much more efficient at converting carbon into a stable form and cleaner than other forms of charcoal.

Biochar is black, very porous, light, fine-grained and has a large surface area. Approximately 70 percent of its composition is carbon. The rest is made up of nitrogen, hydrogen and oxygen among other elements. The chemical composition of Biochar varies depending on the raw materials used to produce it and the methods used to heat it.

Carbon. Porous carbon

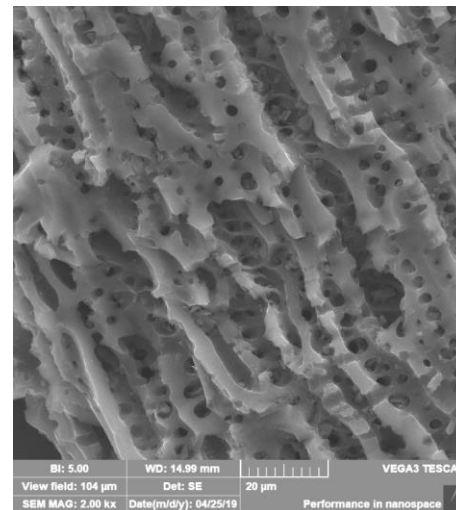
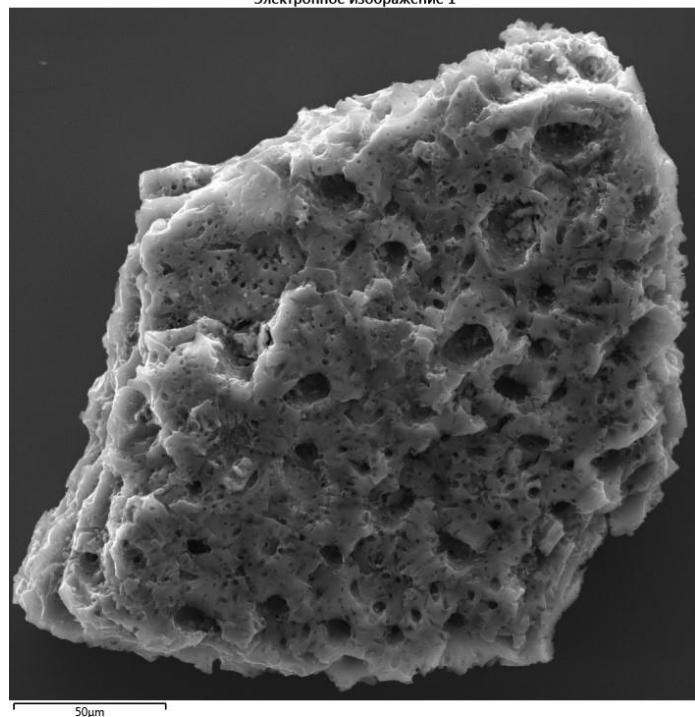


A microporous carbon has most of its porosity in pores less than 2 nm wide and has an apparent surface area that is typically in the range of 1000 to 2000 m² g⁻¹.

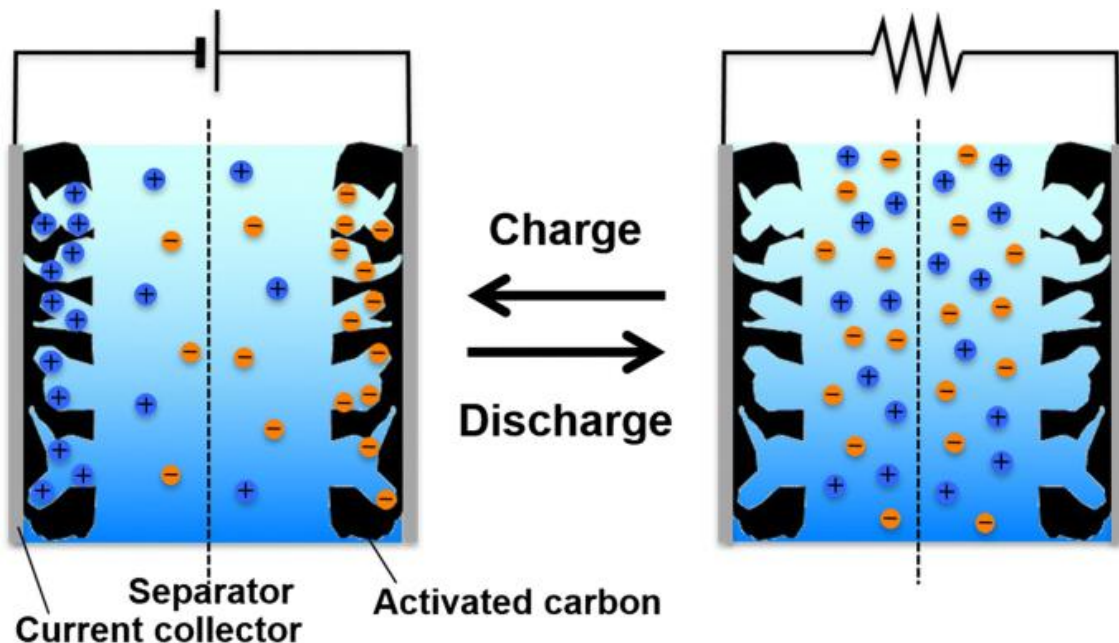
Materials with pore sizes between 2 and 50 nm are called mesoporous, and materials with pores sizes smaller than 2 nm are called microporous. In addition, the term nanoporous material covers materials that have pores up to 100 nm

There are two methods of preparing activated carbons using biochar as raw material: physical (steam) and chemical (bases or acids, carbon dioxide) activation.

Электронное изображение 1



Carbon. Porous carbon. Applications



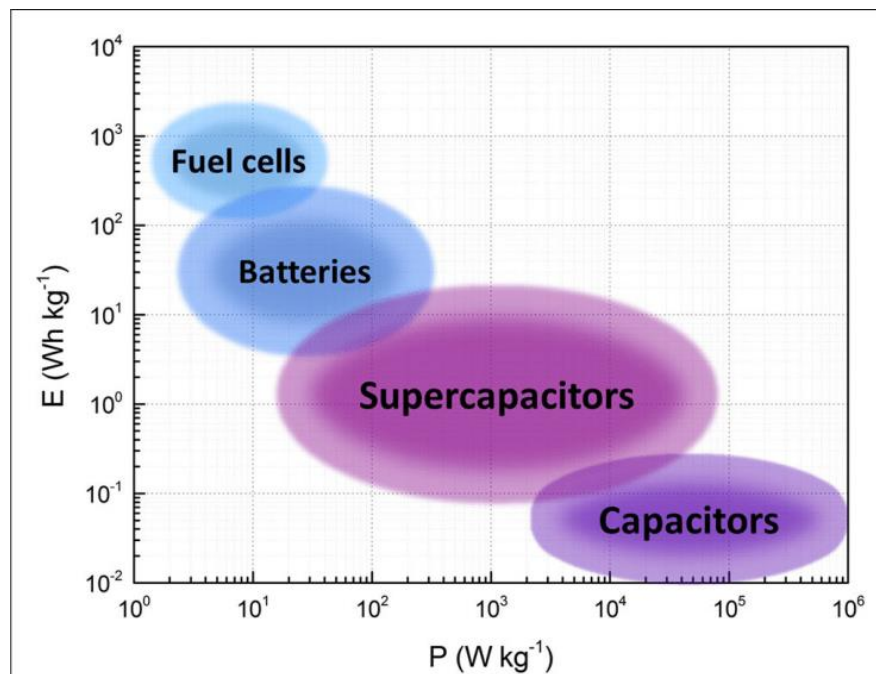
*Extremely wide scope -
from medicine and industry to energy storage*

An Electrochemical Double Layer Capacitor (EDLC) is an energy storage device based on electrostatic effects occurring between two carbon electrodes with a large specific surface areas. The electrodes are immersed in an electrolyte, and a separator is used between the electrodes.

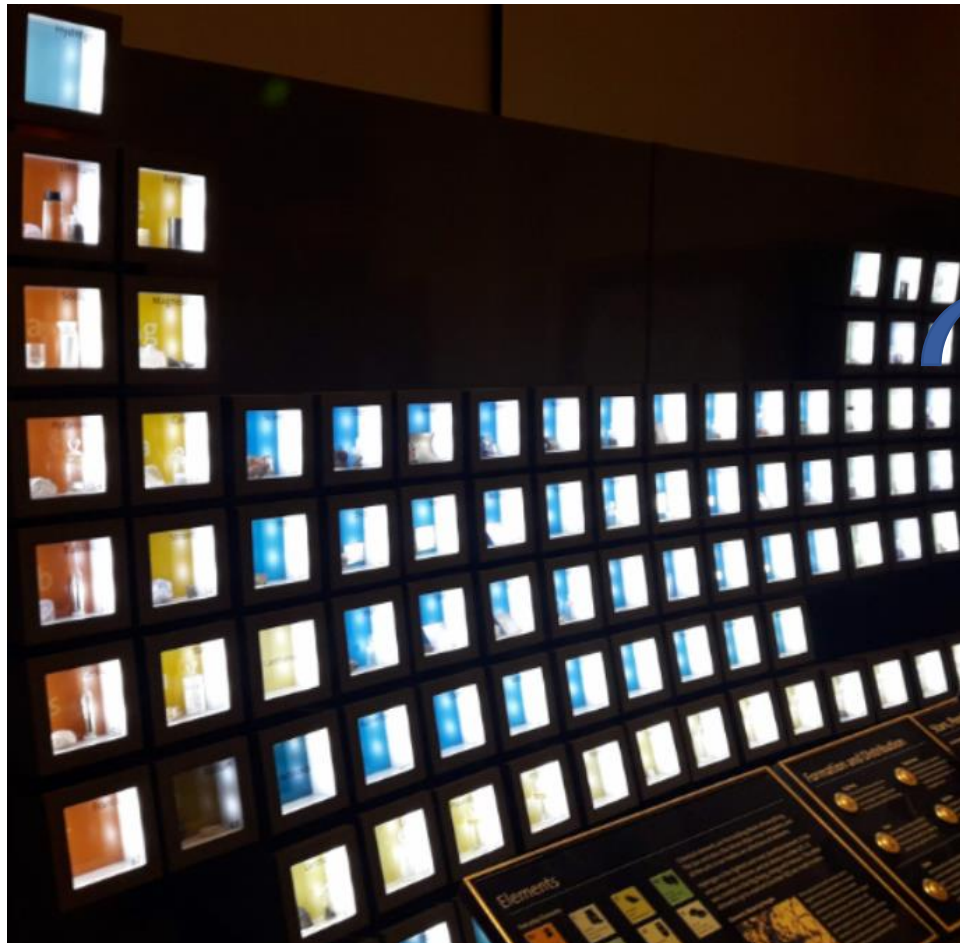
The mechanism of absorption and desorption of ions by an electrical double layer on carbon electrodes promotes charge and discharge. By applying voltage to the facing electrodes, ions are drawn to the surface of the electrical double layer and are charged with electricity.

The main reasons are the extremely high specific surface area, chemical stability and good electrical conductivity.

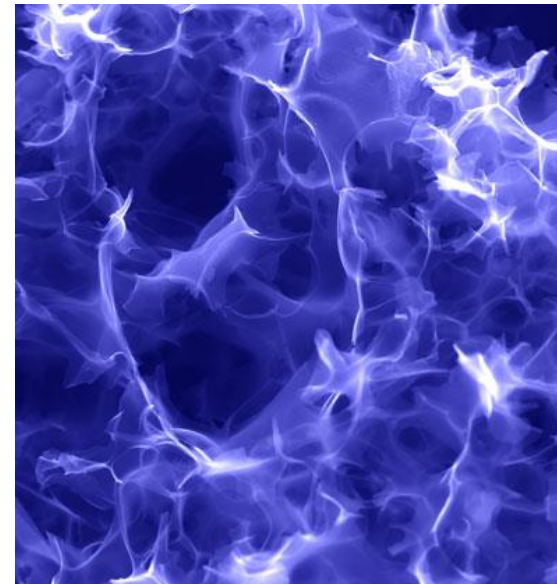
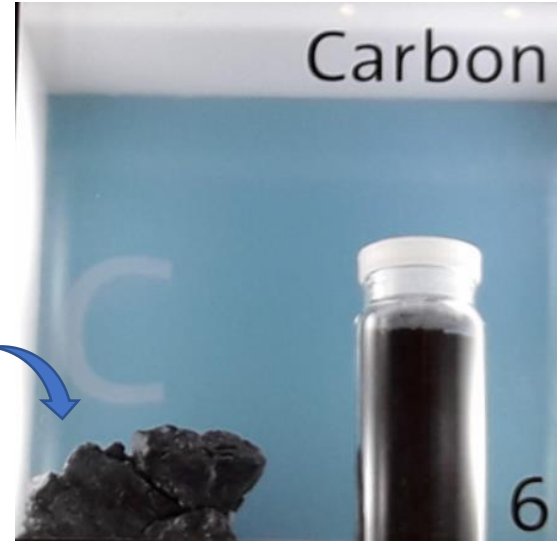
Supercapacitors fill the gap between conventional capacitors and batteries in terms of energy density and power.



Carbon. Porous carbon. Applications



Griffith Observatory



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Volodymyra Boichuk

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