

Applications of carbon nanotubes in energy storage devices

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The development of carbon nanotubes (CNTs) has contributed to the advancement in science and technology. If CNTs could be properly engineered, they can pave the way for the future development of numerous additional materials for a variety of purposes. The open and enriched chiral structure of CNTs enabled improvements in the properties and performances of other materials, when CNTs are incorporated in them. Depending on the structural and morphological requirements, CNTs have been incorporated in energy storage systems either as an additive to improve the electronic conductivity of cathode materials or as an active anode component. Furthermore, they have also been used directly as the electrode material in supercapacitors and fuel cells. Therefore, demand for CNTs is increasing due to their underlying properties and prospective applications in the energy storage research fields. Different types of CNTs are used in energy storage devices such as batteries, supercapacitors, fuel cells, and other devices. This chapter focuses on the review of CNTs in various energy conversion and storage systems, how their morphology and structure affect the electrochemical capabilities and energy storage methods.

Keywords: Carbon nanotubes, Synthesis, Energy storage applications

Energy storage has drawn a lot of interest from researchers and industry for the development of appropriate approaches for storing energy, because of the rapid development of economy and the rise in the use of portable electronic/electronic vehicles. Energy storage can be done in various forms like thermal, chemical, electrical and electrochemical energy¹. Thermal energy utilises electricity to deliver warmth (geezers) or cooling (air conditioners). Renewable natural resources like the sun and wind can be used to generate electricity². Numerous energy-storage technology including batteries, supercapacitors, solar cells, and fuel cells, are being carefully studied for use as primary energy storage devices³⁻⁶. There are wide varieties of energy storage technologies, from complex micro batteries to enormous electric cars. To satisfy the increasing demand, a high-performance, compact, lightweight, adaptive, and more financially viable energy storage device is required in the future. Literature survey depicts a wide range of organic and inorganic materials, or their nanocomposites, are frequently utilised as a material for energy storage systems. Selection of material storage includes metal oxides, conducting polymers, graphene, carbon

nanotubes, metal organic framework etc.⁷⁻¹⁰. However, there are number of energy storage devices have been developed so far like fuel cell, batteries, capacitors, solar cells etc. Among them, a British scientist named William Grove invented the fuel cell in 1839, which was the first energy storage technology capable of producing a significant quantity of energy¹¹. In 1960, National Aeronautics and Space Administration unveiled the first fuel cell for commercial purpose based on Grove's strategy to produce energy¹². After the invention, a number of modifications have been made to improve its energy production ability. Furthermore, various modifications have been done on energy storage devices like batteries, fuel cells and supercapacitors. Gaston Plante introduced the first battery (a lead-acid battery) in the year 1859. However, Emile Alphonse Faure, after developing sticky plates, was the first to commercially launch the lead-acid battery in the late nineteenth century (1880). He improved the storage capacity of the battery by coating lead plates with a paste made of lead powder and sulfuric acid during the development stage. This method has been used repeatedly by other researchers, and it has led to an

increase in storage capacity. For example, Sellon has made breakthrough of employing the Faure's procedure to create batteries. In this work, they placed the same paste on a perforated plate and discovered that the resulting battery has a better storage capacity than the Faure's battery¹³. Late in 1961, American Oil Company and Standard Oil of Ohio developed and patented electronic double layer capacitors, sometimes known as supercapacitors. After that, researchers began to pay close attention to these energy storage devices, and the way they have been used made a notable improvement. The development of solar cell devices created a new revolution in energy storage application and led to an enormous advancement in the field of energy storage. Becquerel built the first solar cell devices and established the idea of a solar cell in the year (1839)¹⁴. To boost the storage capacity and stability of energy storage devices, researchers have investigated a number of materials, including carbon-based materials, metal oxides, composites, and hybrids, which are discussed in the following sections.

Carbon nanotubes

Carbon nanotubes (CNTs) have attracted significant interest due to their distinctive structures and properties. CNTs were considered to be key pioneer and constant driving force towards progress in nanomaterials and nanotechnology over the last three decades¹⁵⁻¹⁶. An individual CNT consists of seamless cylinder formed by rolling up graphene sheets. CNTs can be classified as single-walled CNTs (SWCNTs) or multi-walled CNTs (MWCNTs) depending on number of graphene sheets present in their tube walls. An interlayer spacing of around 0.34 nm separates adjacent graphene layers. CNTs are made of sp^2 bonded carbon atoms and are associated with distinct mechanical, physical, and chemical properties¹⁷. For example, CNTs show electrical conductivity of 10^6 S m^{-1} for SWCNTs or over 10^5 S m^{-1} for MWCNTs at 300 K, thermal conductivity of $3500 \text{ W m}^{-1} \text{ K}^{-1}$, Young's modulus of 1 TPa, tensile strength up to 300 GPa, a large specific surface area up to $1315 \text{ m}^2 \text{ g}^{-1}$, and low mass density¹⁸. CNT's was found to be excellent candidate materials for flexible batteries because of outstanding mechanical, physical property and stability. After tremendous advancements, mass production of CNTs (in powders or liquid dispersions) has now been achieved by chemical vapor deposition (CVD). CNTs finds many applications like energy storage, catalysis, electronics

and functional composites¹⁹⁻²⁰. They have been considered to be essential building block to create various flexible composite structures. There are varieties of several commercial goods available that contain CNTs. For instance, they have replaced conventional conductive additives based on graphite or carbon black particles as conductive additives in many batteries. More intensive application of CNTs in flexible batteries is expected in the near future.

Synthesis of carbon nanotubes

Since the discovery of CNTs by Iijima in the 1990s and they have utilised in various applications like actuators, artificial muscles, and portable electromagnetic shields²¹⁻²³. Carbon nanotubes are made primarily by using energy to break down the carbon source into atomic or ionic forms, which is then condensed into a one-dimensional carbon structure. Large-scale CNTs have currently been produced using the arc discharge method²⁴, laser ablation method²⁵, and chemical vapour deposition (CVD) method²⁶. The CNTs made by various techniques varied greatly in their structure and properties. Arc discharge and laser ablation methods often produce CNTs with great crystallinity and straightness. Various methods of synthesis of CNT is shown in Fig. 1.

CNTs and their composites for alkali metal ion batteries (Li, Na and K) and other batteries

Based on the various methods used for synthesis of CNTs, the one-dimensional CNT can be obtained as SWCNTs and MWCNTs. Due to their superior

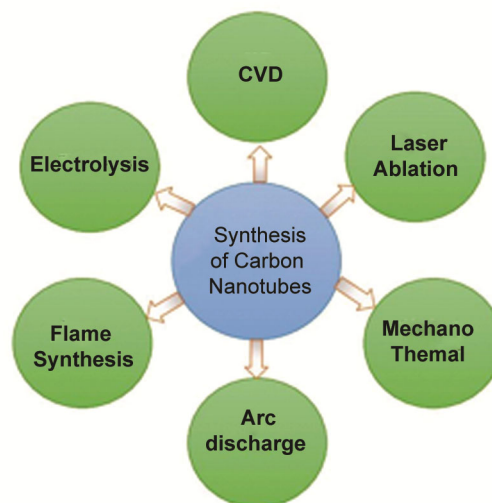


Fig. 1 — Different methods of synthesising carbon nanotubes

electrochemical properties, such as reduced density, stronger tensile strength, and higher stiffness, CNTs have been used in energy storage studies on carbon structures and nano composite materials over the past 20 years. Applications of CNT in energy storage devices are shown in Fig. 2.

Li-ion batteries (LIB)

Recent investigations and development towards the flexible power sources has gained importance in the development of flexible, lightweight, binder-free, and current-collector-free electrodes for LIBs. In conventional methods for fabrication of LIB electrodes, binders may be introduced to inhibit the collapse of the active materials from metal current collectors. In order to maintain electrical conductivity, current collectors are used. The application of binders and current collectors does not contribute to overall working, but only adds dead weight to the lithium storage, which in turn decreases the energy density of LIBs. Both SWCNTs and MWCNTs are used for fabricating CNT papers, and then used as anodes for LIBs. Free-standing paper electrodes have several advantages compared with conventional electrode CNT materials. The dead weight of an electrode is reduced with the removal of the binders and current collector, increasing capacity and specific energy density for the entire battery design. Second, the CNT sheets' ease of handling makes it easy to mould them into the different shapes needed for a variety of flexible and lightweight electrical devices. Literature

survey suggests the applications of carbon nanotubes for Li-Ion batteries²⁷⁻³⁴.

Lithium Sulfur (Li-S) Batteries

Recent applications show considerable promise for lithium-sulfur (Li-S) battery designs based on conversion mechanisms. Theoretically, combination of metallic lithium and elemental sulphur can provide an energy density of 2,500 Wh/kg, which is nearly four times greater than what LIBs can actually achieve. Additionally, Li-S batteries are economical and environmentally friendly substitute for Li-ion batteries due to the natural abundance, superior geographic distribution, and environmental benign nature of sulphur. Furthermore, the closed Li-S system favours an easier and more affordable transition from LIBs to Li-S batteries in terms of battery manufacture, making it more commercially viable than the open lithium-air system. The usage of carbon nanotubes in Li-S batteries are listed below in Table 1³⁵⁻⁴⁴.

Sodium Ion Batteries (SIBs)

The reaction of oxygen with alkali metal to produce alkali oxides underlies the operation of a metal air electrode. Typically, the cathode is comprised of porous carbon or porous metal that serves as both for host for the product and delivers the current needed for oxygen reduction. The discharge process typically involves injecting the oxide products into the voids, and it ends after all of the voids have been filled. The catalyst placed onto the porous matrix

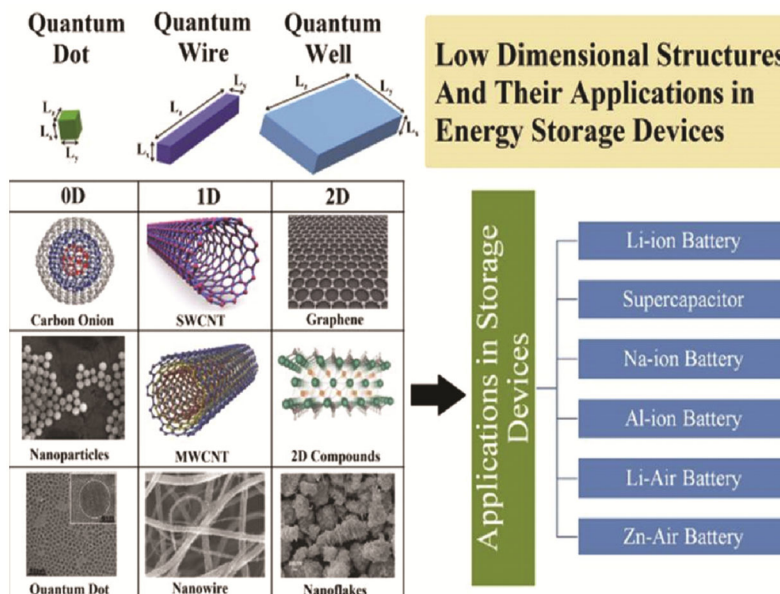


Fig. 2 — Applications of carbon nanotubes in energy storage devices

Table 1 — Applications of carbon nanotubes in Li-S batteries and their performance

S. No	Materials	Mass load (mg cm ⁻²)	Sulfur contents (wt%)/loading (mg cm ⁻²)	Current density	Cycle number	Initial capacity (mA h g ⁻¹)	Post-cycle capacity (mA h g ⁻¹)	References
1	Modify separator MWCNT	0.17	70/2	C/5	150	1324	881	[35]
2	DWCNT	0.00477	60/1.5–2	0.1 C	100	598	508	[36]
3	o-MWCNT	0.4	90/5	0.1 A g ⁻¹	50	1105	925	[37]
4	CNT	–	–/–	1 C	500	684	361	[38]
5	Co-NCNTs	0.23	70/1.8	0.2 C	200	1251.4(20 th)	857.7	[39]
6	SCL	0.25	70/1.35–1.55	1 C	400	1073	700.4	[40]
7	MoS ₂ @CNT	–	77.5/–	1 C	500	~690	~670	[41]
8	Sb ₂ S ₃ /CNT	0.4	65/1.0	1 C	1000	~720	373	[42]
9	SnS ₂ /CNT	0.4	78/1.4	2 C	800	~782	555	[43]
10	NSCNTs/MoS ₂	0.5	88.3/2.2	1 C	1000	1024	814	[44]

Table 2 — Application of carbon nanotubes in Na-ion batteries and their performance

S. No	Material	Current density	Initial discharge capacity (mA h g ⁻¹)	Cyclic number (Cycles)	Final discharge capacity (mA h g ⁻¹)	Rate performance ((mA g ⁻¹)/(mA h g ⁻¹))	Reference
1	Na ₂ FeP ₂₀₇	1 C	90	60	85	2 C/86	[45]
2	NaFe ₂ Mn(PO ₄) ₃	20 mA/g	164.5	50	120.2	80/80	[46]
3	Na ₃ V ²⁰² (PO ₄) ₂ F	0.1 C	110	120	98	1C/42	[47]
4	Na ₃ V ₂ (PO ₄) ₃	10 C	96	150	91	20 C/91.2	[48]
5	NaI/CNT	100 mA/g	170	100	156	2000/65	[49]
6	Na ₂ MnPO ₄ F/C	0.5 C	-	50	80.6	0.05 C/122.4	[50]

is necessary for both oxidation and reduction reactions. The oxygen reduction at the catalyst sites during discharging depends on the reaction between the oxygen, alkali ions, and electrons from the conductive matrix. The accumulation of products such as NaO₂, Na₂O₂, and Na₂O on the cathode has an impact on the battery's electrochemical cyclability performance. The electrode must therefore have a sufficient porous structure with the proper pore volume and pore size to allow for optimal oxygen transport. Large-surface-area porous structured materials were investigated as potential remedies to prevent electrochemical products from accumulating on the electrode's surface. Carbon-based materials have been mostly studied due to their unique properties, such excellent electrical conductivity, high surface area, and controllable structure and porosity and are summarised in Table 2⁴⁵⁻⁵⁰.

Lead acid batteries

The technology that involves higher levels of charge acceptance for system efficiency and also postpones common failure mechanisms like sulfation or dendritic development is required to satisfy the needs of modern lead acid battery applications. For instance, in a modern car, additional features like navigation, heating, and air conditioning can result in higher electrical energy usage than the alternator can supply during regular driving. As a result, the battery

system has a negative energy deficit. Modern applications require improved charge acceptance to keep batteries at higher stages of charge and prevent irreversible sulfation. Another illustration is the need for batteries in hybrid-electric vehicles and some grid storage applications to be able to successfully receive charge in short bursts of high current to avoid negative electrode passivation. Lead acid batteries will continue to be competitive with alternative battery types if these obstacles can be overcome. Carbon has been added to the Negative Active Material (NAM) during paste preparation in a variety of forms including carbon black, activated carbon, and more recently developed varieties including graphite and carbon nanotubes⁵¹⁻⁵⁶.

Metal-air batteries

Because of their comparatively high energy density, metal-air batteries, notably zinc-air and lithium-air, have recently attracted a lot of attention. Making flexible electrodes is crucial to creating flexible metal-air batteries. Due to the possibility of using thin metal foil (such as zinc or lithium foil), the cathode in flexible air batteries has drawn increased attention towards flexible metal-air battery as the anode and it displays a particular degree of flexibility. The cathode should have exceptional air permeability because the oxygen-evolution reaction (OER) and oxygen-reduction reaction are essential for the proper

operation of metal-air batteries (ORR). However, because different flexible substrates have poor electrochemical catalytic activities and limited porosity, the specific capacity drops.

Lithium-Air Batteries

Research has been focused on lithium-air, metal-air, and aluminium-air batteries to increase the energy density, which in turn shows highest energy storage capacity among other batteries and semi fuel cells. Lithium-air battery was considered to be one having highest energy density since it does not store any cathode material. Lithium air batteries provide a number of difficulties, including low round-trip efficiency, cyclic instability, and over voltage. The most crucial element of rechargeable lithium-air batteries is the air cathode, which serves as the sites for the oxidation-reduction of oxygen and also stores the products of reaction. Numerous studies on lithium-air batteries, particularly those focusing on the cathode material have been conducted in the current research. Lithium-air battery design needs actually take into account a variety of factors. Electrical insulator Li_2O_2 is also a powerful oxidizer and is insoluble in aprotic electrolyte. As a result, the cathode should be sufficiently high in electric conductivity and resistant to oxidation by lithium peroxide⁵⁷⁻⁶⁰.

Zn-Air batteries

In comparison with the flexible lithium-air batteries, the flexible Zn-air batteries are more attractive because of eco-friendly nature and of abundant in nature. Just like lithium-air batteries cathodes, excellent electro catalytic activity, high electrical conductivity, air permeability, and remarkable flexibility are needed resources of cathodes in zinc-air batteries. Because of risk associated with electrolyte leakage and evaporation, utilization of flexible Zinc-air batteries is significantly limited by using fluidic liquid electrolytes. It is because of their inherent reliability and safety associated with all-solid-state Zn-air batteries are superior to flexible electronic devices and literature search reveals the usage of CNTs in Zinc air batteries⁶¹.

Fuel-cells (FCs)

In recent years, for the improvement of the efficiency of the fuel cell, a lot of research can be carried out in modification of fuel cells. In recent

years catalyst are added to increase the efficiency of fuel cell. The noble metal platinum (Pt) was considered to have good catalytic efficiency. But the usage of platinum was very expensive and choice as catalyst was huge obstacle for fuel cell to be commercialized. Therefore there is a urgent need to develop new catalysts improving the oxidation-reduction reaction (ORR) of the electrode. Common catalysts support includes carbon, graphene, carbon nanotube (CNT), and other forms of carbon are studied as catalysts⁶²⁻⁷¹.

Supercapacitors

Supercapacitors are considered as bridging between batteries and conventional capacitors, covering several orders of magnitude both in energy and in power densities. Two non reactive porous plates, or electrodes, submerged in an electrolyte with a voltage potential supplied across the collectors can be thought of as an ultracapacitor or supercapacitor. The charge is prevented from passing between the two electrodes by a porous dielectric separator. Pseudocapacitor and electric-double-layer capacitor are the two categories into which supercapacitors are typically divided. CNTs feature a distinctive structure, a narrow size distribution in the nanometer range, a large amount of surface area that is easily accessible, low resistance, and great stability. These characteristics imply that CNTs are appropriate for use as polarizable electrode materials. Due to their distinct characteristics, MWCNTs and SWCNTs have both been investigated for use as electrochemical electrodes. As opposed to this, composites with a nano tubular backbone coated in an active phase with pseudo capacitive properties like CNT/oxide composite, represent a substantial step for developing a new generation of supercapacitors for three key reasons. (1) Compared to conventional carbon materials, nanotubes are more effective at allowing the active particles to percolate.(2) Ions can easily diffuse to the active surface of the composite components and to the open mesoporous network created by the entangled nanotubes (3) The nanotubular materials are characterized by a high resiliency and the composite electrodes can easily adapt to the volumetric changes during charge and discharge, which improves drastically the cycling performance of supercapacitors. The applications of capacitors in supercapacitors are shown in Table 3⁷²⁻⁸⁰.

Table 3 — Applications of carbon nanotubes in supercapacitors and their performance

S. No	Electrodes	Synthesis method	Specific capacitance	Electrolytes	Retention after bending/twisting	Retention rate (Cycles)	Energy density	Power density	Ref.
1	GR-MnO ₂ /CNT	electrochemical deposition	486.6 F g ⁻¹	Na ₂ SO ₄ /PVP	NA	92.8% (800)	24.8 Whkg ⁻¹	NA	[72]
2	CoO/CNT	hydrothermal method	17.4 F cm ⁻³	PVA-KOH	NA	85% (1700)	Whcm	NA	[73]
3	MVNN/CNT	hydrothermal method	7.9 F cm ⁻³	PVA/H ₃ PO ₄	NA	82% (10,000)	0.00054 Whcm ⁻³	0.0004 mWcm ⁻³	[74]
4	CNT	chemical vapor deposition	135 F g ⁻¹	PVA/H ₂ SO ₄	5% (100)	97% (3000)	41 Whkg ⁻¹	16,400 Wkg ⁻¹	[75]
5	MnO ₂ @MXene/CNT	hydrothermal method	181.8 F g ⁻¹	1 M Na ₂ SO ₄	95% (1000)	91% (5000)	NA	NA	[76]
6	PVA/CNT/PANI	in situ polymerization	196.5 F cm ⁻³	PVA/H ₂ SO ₄	NA	71.4% (5000)	NA	NA	[77]
7	HPCF	ageing followed by calcination	194.8 F g ⁻¹	6 M KOH	NA	95% (10,000)	9.1 Whkg ⁻¹	3500 Wkg ⁻¹	[78]
8	CNT/Fe ₃ O ₄ /PANI	chemical vapour deposition method	201 F g ⁻¹	NA	PVA/H ₂ SO ₄	96.4% (10,000)	28 Whkg ⁻¹	5300 Wkg ⁻¹	[79]
9	CNT/Cu/PET method	electroplating	4.312 × 10 ⁻³ F cm ⁻²	NA	PVA/H ₃ PO ₄	88% (2500)	NA	NA	[80]
10	CNT/PANI	In situ chemical solution method	NA	NA	1 M H ₂ SO ₄	96.5% (1000)	NA	9000 Wkg ⁻¹	[80]

Conclusion

Nanomaterials and nanotechnologies have been extensively used for energy storage devices over the past years, the performance of the devices based on nanomaterials has attracted significant improvement. Carbon nanotubes is one dimensional nanomaterial functioning as conductive component and also as electroactive material, which makes CNTs an ideal nanomaterial for energy storage devices. CNTs have outstanding benefits over other carbon materials used in conventional Li ion batteries and supercapacitors, such as amorphous carbon, carbon black, hard carbon, and others, including light weight, strong electrical conductivity, exceptional physical robustness, and improved electrochemical performance. This chapter highlights that usage of carbon nanotubes in Li ion batteries can be also applied to other material based energy storage devices, like lithium-sulfur batteries, sodium-ion batteries, lithium-air batteries and so on. Additionally, the employment of CNTs could make the energy storage devices flexible and stretchable, an important feature for wearable applications.

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