Green synthesis of carbon nanotubes to address the water-energy-food nexus: A critical review

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ABSTRACT

Carbon nanotubes are valuable materials that possess excellent physicochemical properties such as high electrical conductivity, thermal stability, mechanical strength, aspect ratio and surface area. These unique properties of CNTs make it possible to develop innovative technologies and materials that can be used in various applications such as agriculture (food), energy and water industries. Regardless of the high demand of CNTs, their cost remains the major problem due to their synthesis procedures that are high temperature dependent and require high capital costs to set-up. CNT costs only become cheaper when bought in large volumes e.g., tonnes. This paper reviews some of the greener methods used for the sustainable synthesis of CNTs in order to address the water-energy-food nexus. It also reports the challenges on green production processes and application of these nanomaterials. The intricate relationship between these resources (water, energy and food) requires a sustainably integrated approach in ensuring water and food security as well as sustainable energy production. CNTs are advanced materials with the capability of solving these challenges and their application in the water, energy and food industry is reviewed.

1. Introduction

Owing to their excellent physicochemical properties, carbon nanotubes (CNTs) have received greater attention in various fields and applications compared to other allotropes of carbon such as amorphous carbon, graphite and diamond [1]. Graphite and diamond are considered the two natural crystalline forms of carbon. In diamond, carbon atoms are sp^3 hybridized, where four bonds are directed towards the corners of a regular tetrahedron resulting in a three-dimensional network [2]. In graphite, sp^2 hybridization occurs in which each atom is connected to three carbons in the xy plane and a weak π bond in the z plane resulting in the hexagonal arrangement of carbon atoms [2]. Carbon atoms can also exist as sp hybridized such as in carbene, which is not very stable [3].

Notably, CNTs are carbon-based materials characterized by excellent mechanical, electrical, thermal and optical properties [4–6]. They are described as nanoscale graphene cylinders that are formed by rolled-up graphene sheets. Depending on the graphene layers, the cylinder could be made up of one, two or multiple graphene sheets resulting in the formation of single-walled, double-walled or multi-walled carbon nanotubes (MWCNTs) respectively [7]. CNTs are a kind of tubular carbon molecule, with each carbon atom being sp^2 hybridized on the tube. The carbon-carbon σ bonds form a hexagonal honeycomb-like structure on the skeleton of the tubes. On each of the carbon atoms, there is a pair of π electrons not involved in the bonding that forms a conjugated π electron cloud across the entire CNT [8]. Raman spectroscopy is the often-used characterization technique to determine the hybridization of the C-atoms in CNTs. The major Raman features of graphene and CNTs are the graphitic (G) band ($\sim$1580 cm^{-1}) and 2D band ($\sim$2670 cm^{-1}). The G band originates from in-plane vibration of sp^2 carbon atoms. This technique gives information about the hybridization state of carbon atoms in a two-dimensional hexagonal lattice [9]. Additional techniques used to determine hybridization in carbon atoms in CNTs include X-ray photoelectron spectroscopy (XPS) and electron energy loss spectroscopy

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(EELS) [10–12]. Normally, XPS spectra is deconvoluted, giving rise to two contributions which are associated with sp^2 and sp^3 hybridized atoms. The sp^3 hybridized atoms obtained from XPS are typically compared to atomic density obtained from the EELS spectrum [10–12].

CNTs exist in different forms and these are presented in Fig. 1. Single-walled carbon nanotubes (SWCNTs) are classified according to their chirality leading to crystallographic configurations such as zig-zag, armchair and chiral [13,14]. The difference in SWCNTs depends on how the graphene is rolled up during synthesis. The choice of rolling relative to the hexagonal network of the graphene sheet and the radius of the closing cylinder allows for various types of SWCNTs to be formed [15]. The chiral vector is represented by a pair of indices, n and m, where these two integers correspond to the number of unit vectors along the two directions in the honeycomb crystal lattice of graphene. When $m = 0$ the nanotube is called "zig-zag", when $n=m$ the nanotube is called "armchair" and all other configurations are designated as chiral [15] (Fig. 1).

A novel carbon nanostructured material named few-walled carbon nanotubes (FWCNTs) was discovered, consisting of 2–6 layers of side walls with few tens of micrometers in length and diameters ranging between 2–8 nm [16]. Due to their excellent electronic properties and high mechanical strength, FWCNTs find extensive applications in field emission and are also believed to be the next generation CNT-based reinforcement composite materials [17]. MWCNTs can have outer diameters of 2–100 nm and the length can reach several hundreds of microns. On the other hand, SWCNTs have diameters of $1-2$ nm and their length is typically in the micrometer range [7]. SWCNTs like DWCNTs, usually cluster together to form bundles. In a bundle structure, the SWCNTs are hexagonally organized to form a crystal-like structure [18].

CNTs find major applications in field-effect transistors, energy conversion, and conducting films due to their remarkable electrical characteristics [19]. MWCNTs and their hybrid nanocomposites are widely developed for energy storage, transistors, photocatalytic application, additives in polymers, drug delivery, sensors, coating, films for aerospace applications and flat panel display [20–24]. Furthermore, the effect of MWCNTs on nanofluids as future thermal devices with an improved heat transfer rate is being investigated [25]. Due to the advanced properties of CNTs, researchers have shown a great interest in these materials for next generation applications. Consequently, the rate of research output relating to CNTs and their application in the water-energy-food sector has grown remarkably over the past 10 years from 2010 (Fig. 2). The energy sector has shown a greater rate of CNT application compared to the water and food sector. Remarkably, CNTs are extensively used in the development of energy storage and
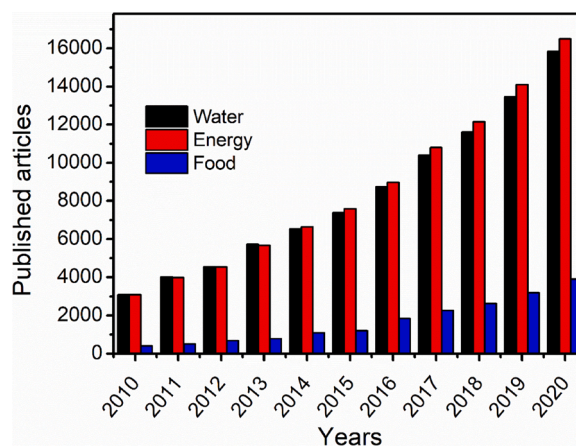

Fig. 2. Number of articles published in CNTs in the water-food-energy sector over 10 years obtained from Science Direct.
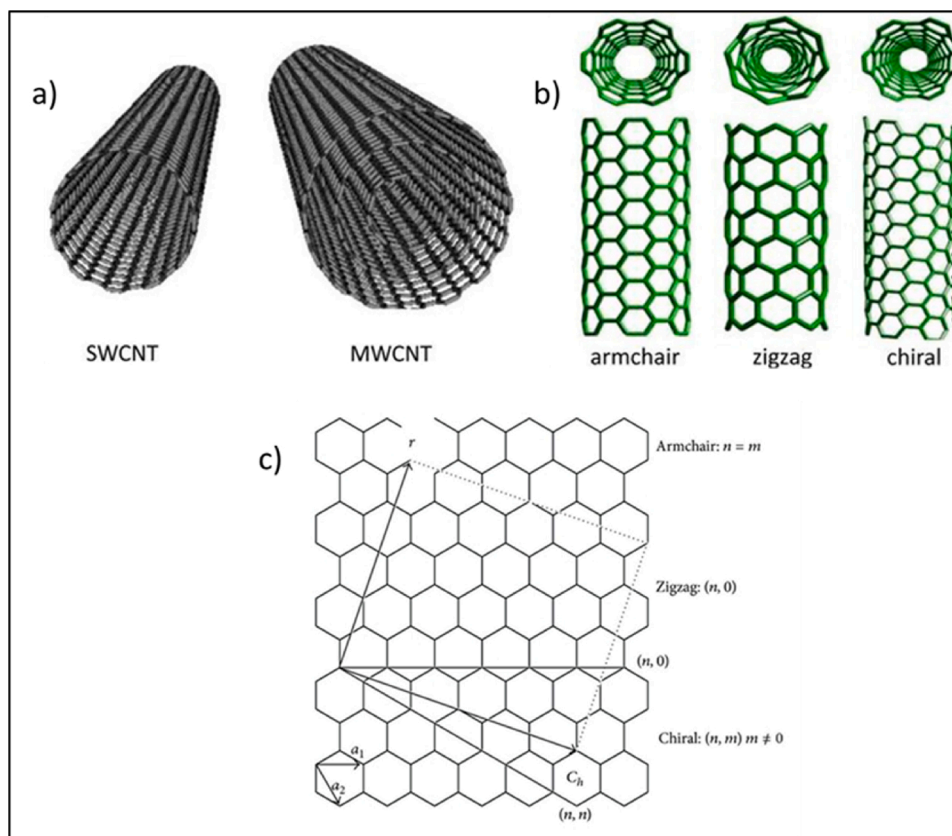

Fig. 1. a) Models of single-walled and multi-walled CNTs, b) the crystallographic configurations of SWCNTs (armchair, zig-zag and chiral) and c) the formation of SWCNTs [14,15].

conversion devices such as solar cells [26]. The slow rate of CNT application in the food sector is attributed to the uncertain toxicological effects of CNTs. However, undertaken toxicological studies are opening research directions towards further applications of CNTs in the food sector.

CNTs are synthesized using different methods leading to the formation of different types of CNTs (i.e. SWCNTs, DWCNTs, FWCNTs and MWCNTs) with distinct properties such as size and purity [27]. Table 1 provides a comparison between SWCNTs and MWCNTs. The focus of this review is on MWCNTs because they are highly studied due to their ease of synthesis and vast applications [28]. Typically, laser ablation, arc discharge, high pressure carbon monoxide (HiPco) and chemical vapor deposition (CVD) are the commonly used methods for synthesis of CNTs, with the latter being the most preferred [29–31]. The laser ablation method involves the vaporization of a graphite target using a laser, usually in a quartz tube furnace in an inert environment of Ar or He at a pressure of 500 Torr [6]. This is followed by collection of produced CNTs on a water-cooled copper target. Both SWCNTs and MWCNTs can be produced at varied reaction conditions. A high purity graphite anode and cathode under an inert environment of He are utilized in the arc discharge method, where a stable arc is achieved at a voltage of $12-25$ V [10]. The CNTs deposit on the cathode as the anode is consumed during synthesis process. The method can produce both SWCNTs and MWCNTs, where doping the electrodes with a metallic catalyst produces SWCNTs and unmodified electrodes produce MWCNTs [27].

HiPco is the prevalent method for SWCNT synthesis. It involves mixing high pressure CO at about 30 atm that has been preheated at 1000 °C [28]. The precursor decomposes under these conditions forming metal particle clusters on which nucleation and growth of CNTs takes place. The catalyst used is in a gas phase from the volatile organometallic catalyst precursor [29]. The laser deposition and arc discharge methods have been replaced by the CVD method due to low yields and the high temperature requirements of these processes, which increase the production costs.

There are many different types of CVD methods such as catalytic chemical vapor deposition (CCVD), either thermal or plasma enhanced (PE), oxygen assisted CVD, water assisted CVD, microwave plasma, radiofrequency CVD (RF-CVD) or hot-filament (HFCVD) [32–34]. The CCVD is currently the standard technique for CNT synthesis, which involves the decomposition of a hydrocarbon gas or carbon containing solvent in an inert environment leading to the formation of CNTs on a substrate/support or reactor [35]. It is facile and produces high yields of CNTs. Furthermore, the morphology, particle size, length, alignment, density and orientation of the CNTs produced under this method are controllable [36]. The CVD method is currently the most widely to produce both MWCNTs and SWCNTs of high yield and quality. Aligned SWCNTs on silica substrates for electronic applications can be attained easily using the CVD method [37]. The drawback associated with the CNTs produced using these methods is the requirement of a purification step (removal of impurities) prior to their application. The impurities are typically amorphous carbon, graphene flakes, and the metal catalyst

[38].

The growth of the CNTs occurs in two different ways depending on the metal-support interaction that exists. When there is a weak interaction, a tip growth mechanism is observed [37]. Here, the hydrocarbon decomposes on top of the metal and the carbon diffuses through the metal [38]. SWCNT growth has been shown to be inhibited by weak interactions due to the large catalyst particles that form. Thus, a moderate interaction is preferred for the right particle size and shape for SWCNT growth. When a strong interaction exists between the catalyst and the support, a root/base growth is observed (Fig. 3) [39]. Like the tip-growth mechanism, the hydrocarbon decomposes on top of the metal while the carbon starts to diffuse through the metal (Fig. 3a(i)). However, due to strong catalyst-substrate interaction, the metal particle is not pushed higher, and the CNT grows on top of the metal. Similarly, the catalytic activity terminates upon metal coverage by the excess carbon, leading to stoppage of CNT growth (Fig. 3a(iii)).

There are various factors that influence the growth of SWCNTs or MWCNTs. The size and properties of the catalyst play a significant role, where smaller particle size (a few nm) leads to the growth of SWCNTs, whereas MWCNTs are formed when the particle size is larger (tens of nm) [39]. The SWCNTs have a high formation energy than MWCNTs. Hence, MWCNTs are easily grown even at low temperatures (~600 °C), while SWCNTs require high temperatures (~900−1200 °C) [40]. Therefore, SWCNTs are produced from hydrocarbons such as benzene and carbon monoxide, which are stable at high temperatures. Furthermore, the type of hydrocarbon used influences the shape of CNTs produced. For example, acetylene and methane being linear hydrocarbons produce hallow CNTs while cyclic hydrocarbons such as benzene and fullerene produce curved CNTs [41].

The diameter and length of CNTs are mainly influenced by the catalyst, gas flow rate, reaction temperature and time. Increasing the catalyst concentration leads to an increase in the diameter of the CNTs. This is believed to be due to the formation of larger particles at a higher metal concentration that produces CNTs with large diameters [44]. With an increase in temperature, the CNT growth rate increases leading to an increase in their density and length [45] while their diameter and wall thickness decreases at high temperatures [46,47]. Although, higher temperature increases the CNT growth rate, the morphological evolution deprives the adsorption of carbon onto the catalysts, thus, forming thinner nanotubes [48]. The length, quantity and crystallinity (quality) of CNTs increases with increasing reaction time [49]. This is attributed to the longer resident time, which increases the concentration of active carbon precursor in the gas phase. CNTs formed under high flow rate appear to be shorter and more entangled. Similarly, higher flow rate causes higher carbon diffusion rate, thus, causing the carbon precursor to react with the catalysts and form more carbon species [50,51]. However, when the flow rate continues to increase, the contact time between the carbon precursor and catalysts decreases significantly, resulting in insufficient decomposition rate of the hydrocarbon and a decline in the CNT yield [52].

Although, CNTs possess excellent properties, purification and functionalization is required to further enhance their properties for a desired application. The semiconducting or metallic properties of CNTs resulting from the different structure and chirality brings challenges to the further development of CNTs in various fields. Functionalization of CNTs leads to the alteration of their electronic properties, introduction of some functional groups, and increase in hydrophilicity or adhesion of tubes to a matrix. The attachment of various functional groups enhances their solubility, chemical reactivity and other physicochemical properties [41].

Recently, various heteroatoms have been used to dope and decorate the sidewalls of CNTs and their effect in application studied. Heteroatom-doping is a process in which carbon atoms in the graphitic structure of CNTs are replaced by heteroatoms [43]. The dopant (heteroatom) such as nitrogen (N), fluorine (F) or boron (B), phosphorus (P) and sulfur (S) can either have a higher or lower electronegativity than

Table 1
Differences between SWCNTs and MWCNTs [39].

| Single-walled carbon nanotubes (SWCNTs)	Multi-walled carbon nanotubes (MWCNTs)
• A single layer of graphene.	• Multiple layers of graphene.
• Bulk synthesis is difficult due to the requirement of controlled growth and atmospheric conditions.	• Bulk synthesis is easier.
• Purity is often poor.	• High purity.
• Chances of defects are high during functionalization.	• Chances of defects are less and could be improved if they occur.
• Easily twisted due to a single layer of graphene.	• Difficult to be twisted due to multiple layers of graphene.
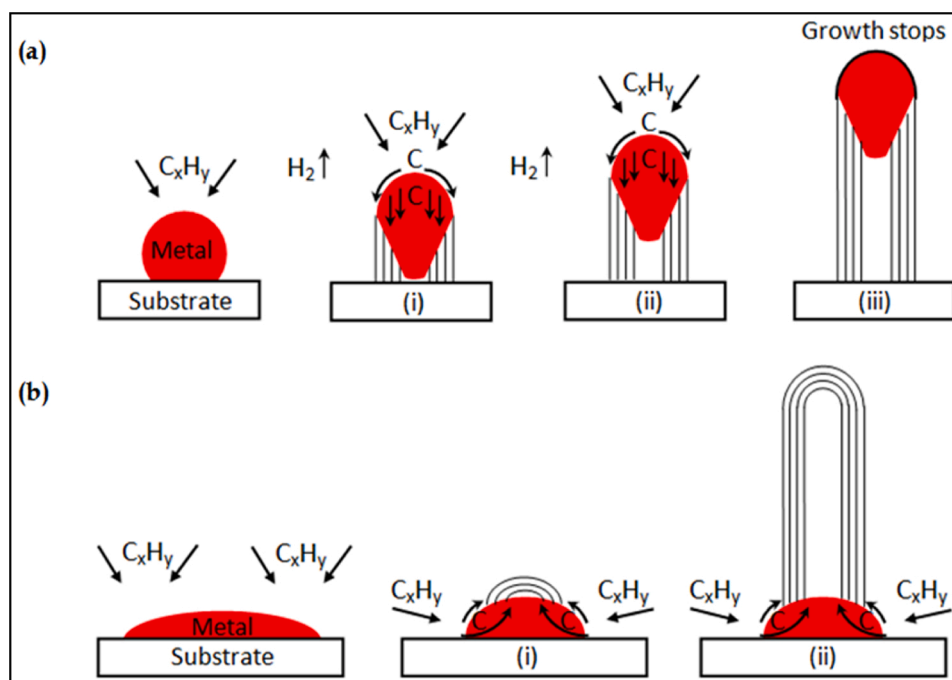
Fig. 3. Growth mechanism of CNTs during CVD: (a) tip-growth mechanism and (b) root-growth mechanism [40].

that of carbon [44,45]. The N and F atoms (higher electronegativity) draws electrons from carbon and lead to the delocalization of π-electrons, which improves the electrical conductivity of CNTs compared to the pristine CNTs. Even though, low electronegative atoms lead to defects on the CNTs surface, their bigger atom size (P, B) compared to C can behave as n-type donors and alter optical and electronic properties of CNTs [46]. Heteroatom-doped CNTs are promising substituents of oxygen reduction reactions (ORR) metal-based catalysts owing to their inherently excellent physicochemical properties and lower cost compared to the precious metal (i.e. Au and Pt) catalysts [47]. N-doped

MWCNTs have also been reported to have extraordinary activity towards both oxygen and hydrogen evolution reactions (OER and HER) [48]. Moreover, recent findings show that dual-doping and multi-doping of CNTs with heteroatoms can further improve their electrocatalytic activity [49]. This is due to the synergistic effect arising from other heteroatoms, resulting in higher charge density and asymmetric spin compared to single heteroatom doping [50]. A new avenue has been opened by heteroatom doped CNTs for metal-free catalysts, energy conversion and storage.

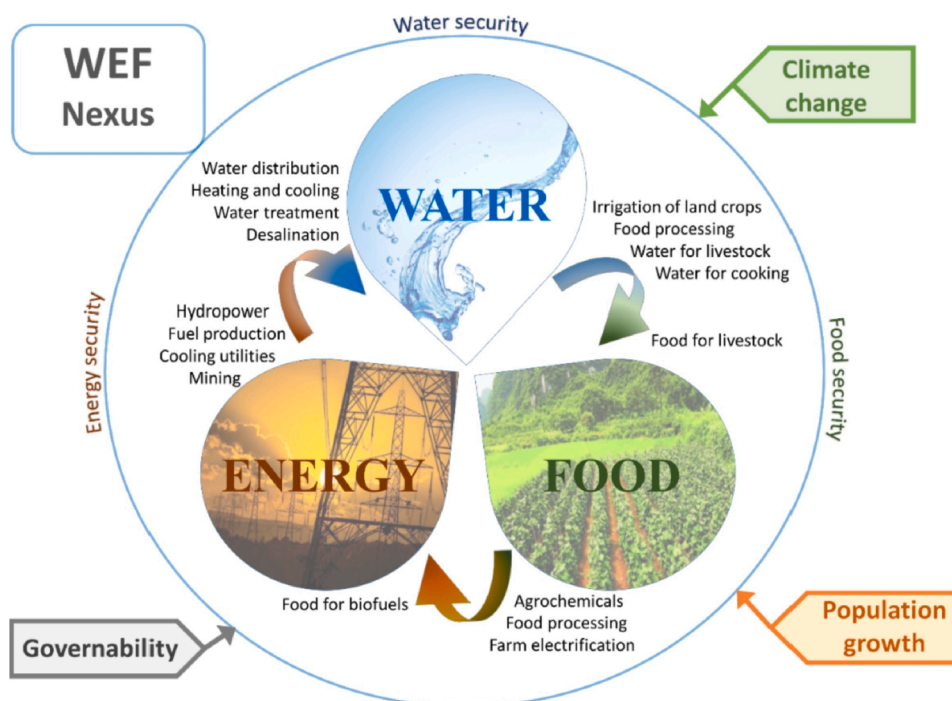The interdependency of water, energy and food is referred to as the

Fig. 4. A diagram depicting the water-energy-food nexus [46].

water-energy-food-nexus (WEF-nexus) [42], which seeks to ensure the security and availability of these resources in the future [43]. The link between these resources is presented in Fig. 4. Typically, water is needed for production of both energy and food. Likewise, energy is a key requirement for the purification and distribution of water. Energy is also required for agricultural activities as well as the distribution of food. Furthermore, food can also be used to generate energy in the form of biofuels. Energy is considered the driving force for production and economic growth. It has a great value and affects the commodity prices. An increase in oil prices (energy) results to an increase in all other products from different sectors such as the food and clothing [44]. It is evident that the shortage of one resource has a negative bearing on the availability of another. Moreover, the demand of the three resources (water, energy and food) is constantly increasing due to urbanization, population growth and climate change, thus, putting further strain on these limited resources [45].

The intricate relationship between these resources requires a sustainably integrated approach in ensuring water and food security and sustainable energy production [44,46]. Nanotechnology has allowed the development of engineered nanomaterials (ENMs) characterized by superior properties compared to their bulk counterparts for application in the WEF nexus. Due to their small sizes and large surface area, ENMs are very reactive and possess a variety of properties that allows them to provide sustainable solutions [62]. The application of ENMs to detect both long-standing and emerging contaminants in water at low concentrations is crucial for treating diverse water pollutants which conventional treatment technologies struggle to manage [63]. A range of ENMs have been applied for accurate, sensitive and reliable detection of organic and inorganic molecules, metals and nonmetal ions and bacteria [64]. Interestingly, ENMs have enabled technologies such as desalination of seawater to increase water supply [65]. In the energy sector, ENMs have been reported to improve energy storage. Additionally, they are electrically insulating and therefore used in batteries and supercapacitors. Novel high-performance supercapacitors and batteries derived from nanocellulose have been developed with possibilities of developing flexible substrates [66]. Notably, the use of clean and energy efficient methods to treat water is among the research topics that have gained attention.

ENMs such as titanium dioxide (TiO_2) remain the leading photocatalytic material used in solar-driven water treatment [67]. Furthermore, high amounts of water and energy are consumed to meet the increasing demand of food. The ENMs have been found to enhance cation exchange capacity, long-lasting nutrient release and nutrient delivery which improves the quality of soils and subsequently ensure sustainable agriculture. In soybeans, iron oxide nanoparticles were observed to improve chlorophyll content and influenced biochemical and enzymatic efficiency in different stages of the photosynthesis reactions [68]. Zinc oxide (ZnO) nanoparticles were found to increase root and shoot growth in chickpea and mung beans [69]. The ENMs have emerged as innovative materials in the agricultural sector to address food security and challenges in the water and energy sector.

CNTs are among the countless ENMs with advanced properties. Since they are a man-made raw material produced via the principles of nanoscience and nanotechnology, it is possible to control their thermal stability, electric conductivity, mechanical strength, aspect ratio and surface area to make them applicable in the water-energy-food sectors. CNTs have surfaces that can easily be modified. As such, their hybrids and composites are developed for water treatment. CNT-based composites have superior adsorption capacity and are used for the removal of multiple organic (e.g., dyes), inorganic (e.g., toxic metals) and biological water contaminants [70]. CNTs are used to improve the electrical conductivity of transition metal oxides/sulfides and hydroxides which are currently used as electrode material in energy storage equipment [71]. Moreover, the application of CNTs in the agricultural sector is increasing due to their plant growth regulation, nano-transport, capabilities to cross plant cell walls, agricultural smart delivery capabilities

and providing a medium for nano-pesticides [72]. The application of CNTs within the WEF sectors is more detailed in the upcoming sections.

The motivation of the study was towards understanding greener techniques and nanotechnology "green nanotechnology" in solving emerging and existing challenges within the WEF nexus. Green nanotechnology has the potential of providing sustainable solutions at an affordable production cost for industrial application. This is paramount especially for CNTs. This is achieved using conventional synthesis methods and understanding that their applicability at an industrial scale is highly dependent on their accessibility, quality and affordability. This review reports on the greener approaches for the synthesis of CNTs focusing on the natural/waste precursors and cleaner energies (e.g., microwaves). Their advanced applications to address the WEF nexus and the current global market of CNTs is discussed. Lastly, challenges presented by the green synthesis methods and application of the CNTs within the WEF nexus is deliberated and perspective is detailed.

2. Green production of carbon nanotubes

The large-scale production of CNTs requires establishment of greener precursors and methods to minimize costs and the negative impacts on the environment [53,72]. Emphasis will be on the CCVD process, which is widely used to produce MWCNTs in large quantities. This method is extensively studied to improve the yield, reduce reaction temperature and time in order to meet the high market demand [7,27,38,47–52]. This increasing demand of CNTs places a strain on the non-renewable resources used in their production and increases the carbon footprint. Thus, methods employed for synthesis of CNTs need to be green to ensure a sustainable and environmentally benign production process. This follows a reduction in the usage of solid-state synthesis which was extensively used previously. The CVD method involves the decomposition of a hydrocarbon gas or organic solvent in the presence of nanosized catalyst particles in a heated furnace [53,54]. Solid-state synthesis is a facile method that employs a non-specialized reactor and utilizes a solid hydrocarbon that is initially vaporized through pyrolysis at elevated temperatures. CVD methods have become popular due to the ability to optimize reaction conditions such that CNTs of various properties are produced [55].

The growth mechanism of CNTs synthesized from green precursors is not thoroughly understood. It is believed to follow a mechanism like that of conventional precursors where the active part/ingredient in the natural hydrocarbon precursor (e.g., cellulose) decomposes to form carbon at a specific temperature in an inert environment, followed by deposition on the surface of the natural/ waste catalyst (having an active catalyst source) which provides nucleating sites for the growth of CNTs [74]. In a study performed using bamboo charcoal as a catalyst/ substrate for the synthesis of CNTs, it was observed that during the pyrolysis at high temperatures, minerals in the bamboo charcoal melted. The melt consisted of mainly calcium silicate which was found to exhibit catalytic effect like that of transition metals for the nucleation of CNTs [75]. In another study, plant extract used as a catalyst, was shown to convert into porous/activated carbon. These pores of activated carbon provided the nucleation sites for CNT growth. Due to the porous surface, activated carbon has a large surface area as compared to a transition metal, so nucleation sites for CNT growth were larger. Activated carbon was also observed to decompose hydrocarbons at low temperatures. Thus, ensuring growth of CNTs with plant extract at lower temperatures of 575 °C, compared to transition metal catalyst reactions performed at 700−1200 °C [73].

In order to make the production processes green, the carbon feedstock, catalysts used, purification process and gas emissions must meet the Principles of Green Chemistry and Engineering [56]. As such, the use of natural carbon sources/feedstocks, microwave-assisted methods, green catalysts and support materials are imperative and therefore discussed.

2.1. Natural and waste hydrocarbon precursors

The choice of a carbon precursor is primarily based on its ease of availability as stated in the Principles of Green Chemistry, as well as on the amount of carbon content present in the precursor [57]. The selection of a carbon source is a crucial part in the synthesis of CNTs as it affects the morphology and properties of the final product. Furthermore, the growth rate of CNTs and the life-span of the catalyst are significantly dependent on the carbon precursor [58]. Despite their environmental challenges including their carbon footprint and being non-renewable, the current literature is dominated by fossil fuel-based carbon sources such as methane, natural gas, acetylene and benzene for synthesis of the CNTs [59–62]. Natural precursors such as plant-based hydrocarbons have only recently gained popularity in the synthesis of CNTs. While, almost all parts of plants (stem, leaves, seeds and roots) can be used as CNT precursors [57,63,64]. Moreover, plant derivatives such as camphor, turpentine oil, palm oil, sesame oil, olive oil and coconut oil have been explored as precursors of CNTs [59,65,66]. These plant-derived oils are eco-friendly, renewable, and readily available in large quantities. They have carbon content in the range of 73.8–77.2 % [65], making them ideal carbon precursors and a sustainable approach for CNT synthesis. The use of various natural/waste feedstock in the synthesis of CNTs using different preparation methods and conditions is demonstrated in Table 2.

The use of waste hydrocarbons as alternative carbon precursors for CNT synthesis is another greener approach for reducing the carbon footprint and environmental impact. Among other utilization and recycling approaches, plastic waste which is an environmental challenge, has been used a carbon feedstock for CNT synthesis [79,80]. Furthermore, the high production of by-products remains a financial and environmental challenge for many industries. Glycerol, a by-product produced by the biodiesel industry, with around 10 % of the weight of the feedstock amounted for about 0.96 million tonnes of waste generated in Europe in 2010 [81]. Interestingly, Wu et al. [81], synthesized CNTs from glycerol and estimated that the reforming of 1 tonne of glycerol would produce 2.8 kg CNTs in addition to 500 Nm^3 of hydrogen which could have a considerable impact on the economics of biodiesel production [81]. CNTs were synthesized during combustion in a diesel engine using biodiesel as a carbon source. This process simultaneously provided power generation and produced MWCNTs in the diesel engine which was used as a combustion reactor [66]. Sagu et al. [82], showed that pollutant blast furnace gases from the iron and steel industry, which consist mainly of CO_2, CO, N_2 and H_2, can directly be used to grow CNTs on steel substrates at atmospheric pressure with no modification to the gas composition [82]. Another study conducted by Johnson et al. [83], provided a climate mitigation solution for the synthesis of CNTs from a greenhouse gas (CO_2) by molten carbonate electrolysis [83]. These studies show that natural and waste hydrocarbons have sufficient carbon content to produce CNTs.

2.2. Microwave-assisted methods

The microwave-assisted synthesis technique has been used to produce CNTs at shorter reaction time. The ability of this technique to be used in mass production of CNTs would be a significant contribution towards realizing greener ways of making CNTs [84]. The method has attracted more attention over conventional techniques due to its advantages such as direct, internal and volumetric heating, rapid reaction time, relatively lower cost, and environmental friendliness [85]. Post synthesis processes of the microwave generated CNTs such as purification and functionalization are also quite simple and timesaving. The principle of microwave irradiation is the conversion of electromagnetic energy to thermal energy within the material used [86]. It is worth noting that carbon materials can be easily heated in the presence of a microwave field, defined as dielectric loss tangent. This characteristic enables them to be transformed by microwave heating, giving rise to

Table 2
Synthesis of CNTs using natural/waste feedstock.

Feedstock	Preparation method	Catalyst/ support	Reaction conditions	Properties of CNTs	Ref
Rice straw	Pyrolysis	Fe-Ni/Al_2O_3	T: 830 °C t: 30 min Carrier gas: N_2	MWCNTs Surface area: 188 m^2/g Diameter: 15-40 nm Length: - Purity: - Yield: -	[67]
Olive oil	Pyrolysis	$NiCl_2$/Si wafer	T: 900 °C t: 60 min Carrier gas: Ar	SWCNTs Surface area: - Diameter: 27-31 nm Length: - Purity: - Yield: -	[65]
Turpentine oil	Spray pyrolysis	Ferrocene	T: 700-900 °C t: 15 min Carrier gas: Ar at 100 ml/min	Aligned MWCNTs Surface area: - Diameter: 15-40 nm Length: 70–130 μm Purity: - Yield: -	[68]
Turpentine oil	Spray pyrolysis	Fe-Co/ zeolite	T: 850 °C t: 25 min Carrier gas: N_2 at 100 ml/min	SWCNTs Surface area: - Diameter: 7-20 nm Length: - Purity: 58% Yield: -	[69]
Coconut oil	CVD	Fe	T: 850 °C t: 60 min Carrier gas: N_2 at 100 ml/min	MWCNTs Surface area: - Diameter: 80-100 nm Length: 3-4 μm Purity: 58% Yield: -	[70]
Sesame oil	Spray pyrolysis assisted CVD	Ferrocene Nitrogen source: Acetonitrile	T: 900 °C t: 15 min Carrier gas: Ar at 120 ml/min	N-doped MWCNTs Surface area: - Diameter: 30-60 nm Length: 3-4 μm Purity: 58% Yield: -	[71]
Palm oil	CVD	Ferrocene/ silicon	T: 750 °C t: 30 min Carrier gas: Ar at 150 ml/min	Mixed MWCNTs & SWCNTs Surface area: - Diameter: 0.6-1.2 nm Length: ca. 110 μm Purity: 90% Yield: -	[72]
Camphor	Pyrolysis	Ferrocene	T: 900 °C t: 15 min	Aligned MWCNTs Surface area: -	[73]

(continued on next page)

Table 2 (*continued*)

Feedstock	Preparation method	Catalyst/support	Reaction conditions	Properties of CNTs	Ref
Camphor	CVD	Fe-Co/zeolite	Carrier gas: Ar T: 650 °C t: 60 min	Dimeter: 20–40 nm Length: 200 μm Purity: - Yield: 90% MWCNTs Surface area: -	[74]
			Carrier gas: Ar at 50 ml/min T: 700 °C t: 60 min	Dimeter: ca. 10 nm Length: - Purity: 88% Yield: over 50% MWCNTs Surface area: -	
Petroleum coke	CVD	Fe/silica	Carrier gas: He at 185 ml/min T: 800 °C t: 30 min	Dimeter: 18 nm Length: - Purity: - Yield: - MWCNTs Surface area: -	[75]
Plastic waste (LDPE and PP)	Pyrolysis assisted CVD	Ni	Carrier gas: N$_2$ at 50 ml/min T: 900 °C t: 60 min	Diameter: 40–50 nm Length: - Purity: - Yield: 31% MWCNTs Surface area: -	[76]
Plastic waste centrifuge tubes (PP)	CVD	Steel reactor (contains Fe and Ni)	Carrier gas: N$_2$ T: 750 °C t: 60 min	Diameter: ca. 50 nm Length: 5–8 μm Purity: - Yield: 42% MWCNTs Surface area: -	[77]
Plastic waste (PP)	Pyrolysis	Ni-Mg/cordierite)	Carrier gas: N$_2$ at 100 ml/min	Diameter: 30-50 nm Length: 5–8 μm Purity: - Yield: 93%	[78]

*- data unavailable, T: Temperature, t: time.

new carbon materials such as CNTs [87].

As opposed to the slow cooling steps involved in CVD due to long time required for the heat dissipation of the furnace at elevated synthesis temperatures, cooling of the CNTs synthesized by microwave radiation can be instantaneously triggered as the microwave power ceases [88]. Furthermore, microwave-assisted methods have been reported to produce CNTs that have less defects compared to the conventional heating methods [89]. The microwave technique is applied at various stages of CNT production such as synthesis, purification and functionalization and these are discussed in this section.

2.2.1. Carbon nanotube synthesis using microwave energy

The use of microwave energy in the synthesis of CNTs provides a rapid synthesis process compared to conventional methods. The special heating mechanism of microwaves can accomplish CNT growth in seconds [90]. In the studies discussed below, a rapid growth of CNTs was reported using microwave irradiation within 5 s - 30 min reaction time. This is a great improvement from the conventional pyrolysis and CVD methods discussed in Table 2, where CNTs were formed at 15–60 min

reaction time.

Burakova et al., [85] modified nickel magnesium oxide catalyst with microwave irradiation, which led to an increase in catalyst specific surface area from 5.2 m^2/g to 9.1 m^2/g and a 40–45 % increase in the yield of CNTs. In another study, Guo et al., [91] synthesized MWCNTs on a graphite surface with ferrocene as a catalyst. It was observed that the high temperature generated by microwave irradiation decomposed the ferrocene powder into iron and hydrocarbons, which served as the catalyst and carbon source for the CNTs growth, respectively [91]. This method did not only utilize low temperatures of microwaves at shorter reaction times but also reduced the number of precursors used, which is one of the important factors in green chemistry principles.

Ferrocene and carbon fibers were also studied by Bajpai et al., [92] as precursor materials for CNT synthesis. A rapid CNT growth was observed in a microwave-assisted heating method where graphite was used as a support material. The choice of graphite was based on its strong capability to absorb microwave radiation, thus, reduced the reaction time [92]. Algadri et al. [93], also observed CNT growth using graphite/ferrocene mixture catalyst inside the microwave oven under ambient conditions [93]. Furthermore, Sridhar et al. [94], developed a novel single precursor microwave synthesis of nitrogen doped-CNTs using cobalt-zeolitic imidazolate frameworks. Bamboo-shaped CNTs were grown on graphene substrates to give unique mesoporous three-dimensional carbon nanostructures [94].

Cellulose from biomass is another promising precursor for CNT synthesis using microwave techniques. Zhang et al., [95] successfully synthesized MWCNTs on biochar substrates using nickel as a catalyst. A microwave-assisted CVD method was used with a self-designed quartz reactor using a microwave oven with an output power of 2 kW. The reaction was performed at 600 °C which is significantly lower than conventional CVD method. The carbon-containing species detected in the volatiles were assumed to act as a carbon source initiating the growth of CNTs on the surface of the biochar surfaces, while inorganic species in biomass (i.e. Ca, K, and Fe) structure were assumed to act as catalysts [95]. A similar observation was made by Shi et al. [96], using gunwood biomass via a microwave-induced synthesis of MWCNTs at 500 °C [96].

Hong et al., [97] reported a microwave irradiation method for CNT synthesis on carbon black, silica powder or organic polymer substrates (Teflon and polycarbonate). It was observed that CNTs can be potentially grown on any substrate provided its absorption of microwave energy is small [97]. Another study was performed with a 1000 W, 2.45 GHz domestic microwave oven for the synthesis of MWCNT sponge. MWCNTs having diameters in the 20–50 nm range were obtained under microwave exposure [98]. These studies show that even domestic microwaves can be used as convenient plasma reactors to grow CNTs in a fast, simple and energy efficient way. Moreover, eliminate the need for a flammable gaseous carbon source and an expensive furnace.

2.2.2. Carbon nanotube purification and functionalization using microwave energy

Purification and functionalization are important post synthesis processes of CNTs which can also be performed using microwave techniques to attain desired properties of CNTs. The conventional methods of CNT purification involve sonication, thermal and chemical treatments [38]. In a study performed to determine the effect of microwave irradiation-based purification process toward the physical properties of MWCNTs, it was observed that the thermal conductivity, thermal diffusivity and heat transfer coefficient enhancement ratio of the MWCNTs were significantly improved [99]. Zheng et al. [100], purified MWCNTs using a microwave oxidation method. The process resulted in almost no structural defect and improved the dispersity of the CNTs. It was therefore noted that microwave purification provides higher purification efficiency compared to normal oxidation method [100]. Gomez et al. [101], employed a two-step microwave treatment followed by chlorine gas phase purification process. The MWCNTs were subjected to

1 min microwave irradiation at 1000 W power. The significant advantage of this method over conventional CNT purification procedures is that under microwave treatment in air, the carbon shells that encase the residual metal catalyst particles are removed and the metallic iron is exposed and subsequently oxidized making it assessable for chemical removal [101].

Notably, the functionalization of CNTs is a critically required process since it modifies the CNT surfaces towards the desired application. It typically involves either oxidation using acids or oxidants, causing carboxyl groups (−COOH) to functionalize the defects of CNTs or the addition of a C=C double bond by alkylation, arylation, oxycarbonyl nitrene, and 1,3 dipolar cyclo-addition [102–104]. The oxidation of carbon surfaces is known to generate not only more hydrophilic surface, but also more oxygen containing functional groups to increase the ion-exchange capacity [102]. The oxidized CNTs can be further treated with modifying and reducing agents to accomplish amination functionalization (−CONHR) [122]. The amine functionalized CNTs are reported to possess high solubility properties in water and organic solvents [123]. Fluorides (-FH) [8] and thiol-functional groups (-SH) have also been used to modify CNTs [124]. Thiol-functionalization is believed to be a promising alternative to oxidization in many applications such as composites and metal hybrids [124]. Furthermore, polymer functionalization of CNTs by non-covalent "grafting to" has also gained interest. Contrary to covalent functionalization, non-covalent functionalization ensures the intrinsic properties of CNTs are not altered [125].

Microwave-assisted functionalization of CNTs reduces the reaction time while increasing the degree of functionalization compared to the conventional method [84]. Conventional methods require absorption of thermal energy on the surface of the material and transfer the heat energy through thermal conductivity which is a very slow process [87,99]. CNTs were functionalized with a 1:1 nitric acid and sulfuric acid solution under microwave irradiation. −COOH groups and high concentration of acid sulfonated groups was present in the CNTs. Thus, exhibited excellent solubility, producing long term stable suspensions in deionized water and anhydrous alcohol with higher solubilities of 20 mg/mL observed in acidified water [105]. Mallakpour et al. [106], performed a surface modification of carboxylated-MWCNTs by 5-aminoisophthalic acid under microwave irradiation. The reaction was performed 120 °C for 15 min with a microwave output power of 700 W. The functionalized MWCNTs showed excellent dispersion and were used to prepare reinforced polymer nanocomposites [106]. In another study, it was observed that 5–15 min was sufficient to functionalize the SWCNTs. Longer microwave exposure reduced the functionalization yield and lead to the removal of functional groups that were bonded [107]. Microwave-assisted CNT purification and functionalization processes offer efficiently modified CNT that are highly dispersible and thus, applicable in various fields.

2.3. Natural/waste catalysts and support materials

Conventional metal catalysts have been studied with the quest of finding a catalyst that results in higher CNT yields using minimal amounts [108–110]. Nano-sized transition metal particles such as copper, iron, nickel, cobalt and molybdenum have been successfully used in CCVD methods either in oxide or metallic forms as well as alloys [50]. The bimetallic and trimetallic alloys have shown improved performance compared to the single metal catalyst form [50]. However, these metal catalysts are not environmentally friendly. It was reported that the metal catalysts used for CNT growth portray serious concerns towards the living organisms [111]. Specifically, the catalyst particles remain within the grown CNTs, and these could potentially be transferred to human body in medical treatment where the CNT-based materials are used. Furthermore, unmanaged disposal of these products to the environment, could affect the growth and purity of the plants [56]. In production of CNTs, the catalyst is considered as one of the most expensive components [112]. Therefore, finding an alternative low-cost and effective

catalyst for producing high-quality CNTs remains a challenge for the mass production. Green catalysts derived from plant extracts and natural minerals offer a sustainable approach for CNT synthesis due to their cost-effectiveness and abundant availability.

Minerals such as garnet sand, lava, wollastonites, magnesite, montmorillonite, vermiculite, sepiolite, and biomass-based activated carbon, have been used as catalysts and/or catalyst supports for the synthesis of CNTs [113–116]. Most of these minerals have iron oxide particles in their structures which act as the potential active catalyst source for CNT growth [113]. Endo et al. [112], reported an environmentally friendly synthesis of highly ordered MWCNTs using stone garnet sand as a catalyst and support, and city gas (natural gas collected from gas fields) as the carbon source. It was assumed that iron species embedded in the garnet particles diffused through the material upon heating and precipitated on the surface of the sand to act as a catalyst [112]. Vermiculite, which is a kind of expansion clay was used as a catalyst for the synthesis of vertically aligned CNTs (VACNTs) [117]. In another study, VACNTs were fabricated on fibrous natural wollastonite (type of calcium inosilicate ($CaSiO_3$) found in mines). The VACNTs were in the diameter of 4−30 nm and yields of 0.86 g/h were obtained. High yields of 3 kg/h VACNTs were achieved using Fe/vermiculite catalyst on a pilot plant fluidized bed reactor CVD for mass production [118]. These type of CNTs are not abundantly availability due to small yields obtained during their synthesis leading to high selling prices [118]. Other factors leading to high CNT costs are the production process, product purification, high equipment cost, and limited scalability of manufacturing methods [119]. However, this study shows a feasible large-scale production of VACNTs at low-cost using a natural mineral catalyst, which can potentially meet the increasing demand.

Sepiolite, a natural clay was used to synthesized SWCNTs, which was used as both a catalyst and support [120]. Natural clays are attractive as catalysts due to their large surface area, strong acidity, and strong adsorption and ion-exchange ability [121]. Another type of clay, bentonite was used directly to catalyze the growth of MWCNTs and produce MWCNTs/bentonite composites for styrene synthesis [122]. Interestingly, red mud, a toxic waste product from bauxite processing, was used as a catalyst for the synthesis of MWCNTs by fluidized bed CVD. The reaction was performed at 650 °C and a MWCNT yield of 375 % was achieved [123].

The use of plant extracts as catalyst is advantageous as they are organic compounds that can produce CNTs that are free from metal impurities. Triphati et al., [56] evaluated garden grass (Cynodon dactylon), rose (Rosa), neem (Azadirachta indica) and wall-nut (Juglans regia) plant extracts to synthesize catalysts for MWCNTs growth using the CVD method [56]. The wall-nut extract produced higher yields of CNTs compared to the plant extract catalysts at CVD temperature of 575 °C [56]. Remarkably, the temperatures (575 °C) are lower than the optimum temperatures (≥600 °C) which are typical required for conventional catalysts. Kumar et al. [124], synthesized CNTs using siliceous breccia (natural rock powder composed essentially of SiO_2 α-quartz) as a catalyst source without any pretreatment *via* the CVD method at 750 °C for 30 min. Siliceous breccia contains iron oxide-hydroxides, goethite or hematite, which act as catalysts to dissociate the hydrocarbon precursors and form CNTs [124]. Another natural rock containing an iron silicate skeleton (such as fayalite, Fe_2SiO_4) was used to synthesize CNTs. The iron silicate was etched with HF, releasing iron from the silicate skeleton surface to form iron oxides on the surface of the silicate. The iron oxide was reduced by H_2 to form metallic iron clusters which also acted as a substrate for CNT growth [125]. Furthermore, natural catalysts such as pumice (dust from volcanic eruption) and laterite (clay material) have been studied for synthesis of graphitized CNTs for application as reinforcement materials in composite materials [62,126].

Similarly, the substrate (support) materials also play a critical role in the growth of CNTs and influences the activity of the catalyst. It has been established that the chemical and/or physical interactions occur between the catalyst and support during the CNTs production [127]. Thus,

support materials are required to be resistant towards thermal and chemical process conditions. Typically, the Van der Waals and electrostatic forces are some of the physical interactions that reduce the thermally driven diffusion of metal particles on the support surface. Furthermore, particle mobility can be decreased, causing limited CNT growth as a result of chemical interactions between the catalyst and support surface. Additional properties required for the efficient CNT growth are but not limited to surface area and porosity of the support materials [27].

Generally, substrates such as silica, activated carbon and alumina are used for CNT growth. Aluminum hydroxide and calcium carbonate have recently been shown to influence the CNT growth process during their decomposition [35,47,50]. However, these substrates and supports are non-renewable and costly compared to plant-based substrates. Zhang et al. [95], synthesized CNTs using biochar as a renewable substrate in microwave-assisted CVD method. The porous nature of biochar allowed effective dispersion of catalyst nanoparticles on its surface, thus promoting the formation of CNTs. The biochar also acted as a microwave absorber and raised the temperature on the surface to the required amount. A high degree of graphitization was achieved at 600 °C [95].

Coal fly ash, a noxious coal combustion byproduct regenerated in large quantities by thermal power has been used as a substrate/support material for growing CNTs [128–130]. Dunens et al., [129] used coal fly ash as a support material in the synthesis of MWCNTs. They used Fe as a catalyst and observed that the use of coal fly ash with high Fe content reduced the amount of virgin Fe used, thus lowering costs associated with CNT catalyst manufacturing [129]. Furthermore, VACNTs were grown on a lamellar clay support [131]. An inherent disadvantage of VACNTs is the required use of low surface area flat catalyst supports during synthesis that are not easily amenable to scale up. Lamellar clay support allowed VAWNT growth rates of 160 mm/h with lengths over 80 microns [131]. This shows a promising catalyst support material for VACNT growth that can potentially produce scaled up synthesis.

3. Application of carbon nanotubes

3.1. Carbon nanotubes in water treatment

Access to clean water for the domestic, agricultural, and industrial sectors remains a challenge in many parts of the world. To make it worse, there is pollution coming from these sectors which further threatens the availability and quality of water [132]. Advanced water treatment processes remain indispensable to ensure safe water for consumption and protection of the ecosystem. To further elaborate on the need of advanced water treatment processes, the type of pollutants in water systems are becoming more complex to be treated conventionally. For example, some pollutants exist as soluble compounds at μg/L and ng/L concentrations and are hardly treated using conventional methods. Nanomaterials including CNTs are reported to be effective in treating water polluted by trace compounds. As such, they can be used as independent treatment processes or incorporated into the existing conventional treatment methods. Furthermore, nanomaterial-based sensors have been developed to detect these contaminates at low concentrations to ensure improved water treatment and thus quality. These detectors have given rise to the estimation of the levels of nutrients and water required by the soil, as well as the potential threats posed by the presence of chemical or biological contaminants in water used for food production [62]. Thus, significantly improving precision farming practices. This shows an important linkage to the WEF-nexus, where CNT based water sensors have the ability to ensure food security and safety.

Among the several technologies proposed, CNT-based water treatment technologies are the most promising due to their large surface area, high aspect ratio, high chemical reactivity, cost-effectiveness, and energy efficient [132]. Specifically, CNTs can be incorporated into conventional water treatment processes, thus, opening new avenues towards more efficient water purification processes. Water treatment

processes that involve the use of CNTs and CNT-based materials include adsorption, membrane technology for nanofiltration and desalination, hybrid catalysis, sensing and monitoring of organic, inorganic and biological pollutants [133]. CNT and CNT-based adsorbents are effective in removing various pollutants in aqueous solution (Table 3). The adsorption process is envisaged as a cost-effective and efficient technology for the cleaning of hydrocarbon spillages. The advantage of using CNT-based adsorbents relies on their high hydrocarbon retention, adsorbent reusability and recovery of the adsorbate [134]. In Table 3, a selection of papers based on heavy metal removal by CNTs was made. This was due to the great demand of effective treatment solutions towards emerging pollutants in water bodies because of industrial discharges. Heavy metals are toxic and a serious threat to the environment and human health. Thus, their potential treatment using CNTs and their hybrid is imperative.

Wastewater remains a complex matrix of various pollutants which has fatal effects on human health if consumed. However, the use of CNT-based materials is a one-step solution to alleviate challenges associated with wastewater treatment [143]. In a recent comprehensive review, it was reported that the use of surface-modified CNTs demonstrated a great potential in the removal of heavy metals from wastewater [144]. These CNTs are normally functionalized through oxidative processes where carboxylic (−COOH) and hydroxyl (−OH) functional groups are introduced to the surface of the CNTs. However, this functionalization

Table 3
Application of CNTs in aqueous solutions for the removal of heavy metals.

Nanomaterial	Pollutant	Adsorption experimental conditions	Adsorption capacity (mg/g)	Ref.
MWCNTs composite	Copper ions	D: 0.2 g/L T: 25 °C pH: 7.0 t: 180 min Co: 100 mg/L	189	[135]
Deep eutectic solvent functionalized CNTs	Hg^{2+}	D: 0.12 g/L T: 25 °C pH: 6.7 t: 45 min Co: 5 mg/L	177.76	[136]
MWCNTs/ Polyethyleneimine	Cr^{6+}	D: 0.5 g/L T: 25 °C t: 60 min pH:4 Co: 120 mg/L	45	[137]
MWCNTs/ Selenophosphoryl	Pb^{2+}	D: 0.75 g/L T: 39.85 °C pH 5.0 t: 60 min Co: 20 mg/L	43	[138]
Carboxylic functionalized MWCNTs	Mn^{7+}	D: 0.3 g/L T: 50 °C pH 5.0 t: 60 min Co: -	238	[139]
Amine-MWCNTs/ ALIQUAT336	As^{5+}	D: 0.3 g/L T: 25 °C pH: 12 t: 5 min Co: 1 mg/L	150	[140]
CNTs/activated carbon	Chromium (VI)	D: 0.04 g/L T: 25 °C pH: 4 t: 60 min Co: 0. 5 mg/L	9	[141]
CNTs/mimosa tannin	Pb^{2+}	D: 5 g/L T: 20 °C pH: 4.5 t: 1440 min Co: 10 mg/L	13.8	[142]

*D: dosage, T: temperature, t: equilibrium time, Co: initial pollutant concentration.

remains critically challenged due to the use of strong oxidizing agents such as nitric and sulfuric acid and reducing agents such as KMnO$_4$ which are toxic. Thus, deep eutectic solvents (DESs) have been studied as alternative green functionalization solvents. The advantage of DESs include lower costs, availability, biodegradability, and they are relatively simple to synthesize [144].

Among the heavy metals mentioned in Table 3, a range of other pollutants have been successfully removed by CNTs. These include organic dyes, dissolved organic matter, aromatic compounds, phenolic derivatives, pharmaceuticals, and antibiotics [134,145]. Arabi et al., studied the removal of Congo red dye using ZnO-MWCNTs composite. A maximum adsorption capacity of 249.51 mg/g was achieved at optimum conditions [146]. NH$_2$-MWCNTs also showed high adsorption capacity (178.5 mg/g) towards the removal of methylene blue dye [147]. The adsorption of aspirin, paracetamol, ketoprofen and tonalide by CNTs and their hybrids was investigated [148,149]. The CNTs showed an adsorption capacity in the range of $71-90$ mg/g towards these commonly used drugs. Notably, the hydrophobicity of the organic pollutants governs their adsorption removal from aqueous solutions. CNTs usually offer strong adsorption affinities towards more hydrophobic molecules [150]. For example, 2-naphthol (more hydrophobic) demonstrated a higher adsorption capacity compared to 4-chlorophenol (less hydrophobic) [151,152]. Furthermore, the adsorption capacities of organic contaminants are also affected by their uptake mechanism by the surface of CNTs. Xiong et al., observed that the π-π adsorbate-adsorbent interactions played an important role during the adsorption process of tetracycline antibiotics by MWCNT/MIL-53(Fe) [153]. Interestingly surface modification of CNTs even with low crystallinity has been proven to improve their adsorption capacity towards organic pollutants from wastewater [143].

Along with the adsorption processes earlier discussed, membrane technology has demonstrated effective water purification and has been relatively tested for the desalination of sea and brackish water [154]. However, the processes are energy intensive and thus costly. Additionally, pressure-driven membrane technologies require self-cleaning mechanisms to reduce fouling propensities for long-term operations [155]. Oxidized CNTs enhance the hydrophilic nature of the membranes, thus reducing the interactions between the hydrophobic foulants and the surface of the membrane [156]. The hydrophobic hollow structure of CNTs also aids frictionless fast transport of polar water molecules as shown in Fig. 5 [157].

Ragunath et al., presented an optimized method for CNT immobilization on membrane surface for seawater desalination *via* direct contact membrane distillation (DCMD). A higher water vapor flux of 51.4 L/m^{2}h was observed on CNT immobilized membrane (CNIM), which was 76 % higher than the unmodified support membrane [158]. The CNIM membranes were also observed by Gupta et al., to be effective in the separation of organic solvents from aqueous mixture via sweep gas membrane distillation [159]. Polyvinylidene fluoride (PVDF) membranes were blended with various amounts MWCNTs to fabricate membrane composite with enhanced properties for vacuum membrane distillation. The MWCNT-fabricated membranes exhibited desalination potential with salt rejection $\geq$ 99.9 % [160]. Nthunya et al. [154], have also demonstrated the importance of the oxidized MWCNTs to fabricate fouling-resistant PVDF nanofibre membranes for use in direct contact membrane distillation. CNTs can be incorporated in various water treatment processes as new technologies or in conjunction with existing conventional treatment methods. They are highly effective in removing various water contaminates in a wide concentration range.

3.2. Carbon nanotubes in energy storage

The rise in global energy demand calls for clean sustainable energy production, especially for developing countries like South Africa where the primary source of energy is fossil fuels [161]. Fossils fuels are a non-renewable resource and the leading contributor to the emission of greenhouse gases. Although cleaner technologies are encouraged for sustainable growth and development; they have a short fall of limited energy storage capacity. Once the energy is produced, if not safely stored it can be lost. Thus, affecting food and water security which are highly dependent on energy.

Nanotechnology plays a vital role in developing devices for energy conversion and storage and green engineering of environmentally friendly materials, Fig. 6 [162]. CNTs have high mechanical and electrical properties that makes them suited energy storage devices. The storage performance for both gas adsorption and ion insertion are relatively influenced by the surface and pore structures of CNTs. Three types of intrinsic pore structures exist for CNTs. The first being the hallow tube cavity of CNTs. The inner tube diameter of SWCNTs, DWCNTs and MWCNTs is usually in the range of 1–2 nm, 2–3 nm and 2–30 nm, respectively [163]. The as-synthesized CNTs are usually capped with spheres of fullerene molecules. Thus, decapping is crucial for the access of the adsorbent into the inner tube. The high aspect ratio (ca. 1000) also mean the tubes are long and limits the absorbent from
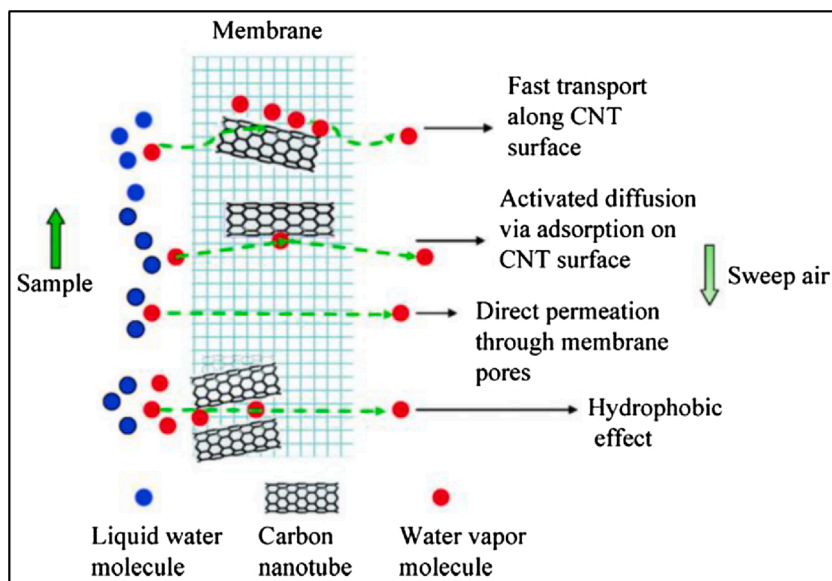

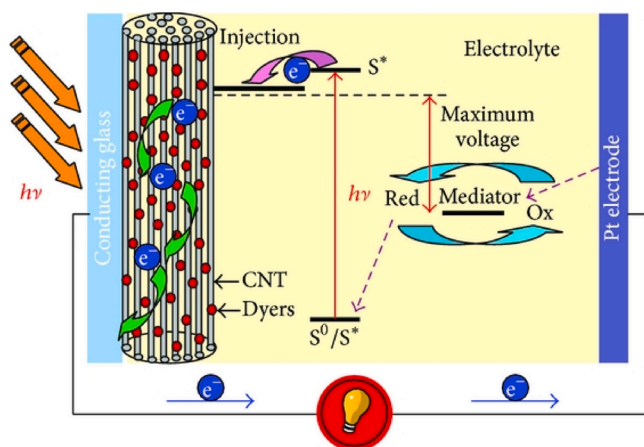**Fig. 5.** Schematic diagram of membrane distillation mechanism on CNT-based membranes [157].

Fig. 6. Energy-related applications of CNTs [162].

Table 4
Performance of various CNT-based energy storage materials along with their application.

Nanomaterial	Application	Performance	Ref.
CNT-bimetal-organic frameworks	Li-ion battery	Reversibility charge capacity of 1250 mA h g^{-1} at 0.5 A g^{-1}	[176]
CNT-Porous TiO$_2$	Solar cell	Reversible charge capacity of 200 mA h g^{-1} at 0.1 A g^{-1}	[177]
	Supercapacitor	specific capacitance of 145 F g^{-1}	
CNT-graphene	Mechanical energy storage device	reversible energy storage density of 130 kJ kg^{-1} energy storage efficiency of 83%	[178]
MWCNT-Pt	Fuel cell	maximum power density of 0.360 mW cm^{-2}	[179]
CNT-CoP-porous carbon	Li-ion battery	reversible charge capacity of 1187.5 mAh·g − 1.at 0.1 A g^{-1}	[180]
nitrogen-doped CNTs- CoTe$_2$	Solar cell	power conversion efficiency	[181]
CNT foam (CNTF)	Li-ion battery	specific capacity of 690 mA h g^{-1} at 0.2 A g^{-1}	[182]
	Supercapacitor	specific capacitance of 144 F g^{-1}	
CNT- micro yarn	Solar cell	average power conversion efficiency of 0.631%	[183]
CNT-carbon fiber	Fuel cell	maximum power density of 2015.6 mW m^{-2}	[184]
CNT-CuCo$_2$S$_4$	Supercapacitor	specific capacitance of 557.5 F g^{-1}	[185]
CNT-embedded PEDOT:PSS	Solar cell	average power conversion efficiency of 16%	[186]
CNT-polypyrrole	Fuel cell	maximum power density of 1876.62 mW m^{-2}	[187]

reaching the center through diffusion [51]. This is countered by the second intrinsic pore structure called the interstitial pores. This refers to the space surrounded by the adjacent in-bundle SWNTs or DWNTs. These interstitial pores inside a CNT bundle are connected to each other, making it possible for small sized adsorbents to access the central portion of the bundle. The third intrinsic pore structure is the interlayer space between the coaxial cylindrical carbon layers (only for DWNTs and MWNTs) [164]. The interlayer distances of MWNTs and DWNTs are typically about 0.34 nm and 0.37 nm respectively, larger than that of graphite (0.335 nm) [165]. Therefore, the interlayer spaces may also be accommodation sites for H$_2$ and Li$^+$, whose diameters are 0.289 nm and 0.07 nm, respectively [164]. Besides the intrinsic pores in CNTs, the aggregated pores may also contribute to the storage performance. These pores are the voids between aligned/ randomly packed MWCNTs or SWCNTs/DWCNTs bundles [164]. These properties of CNTs makes them applicable in energy conversion and storage materials such as solar cells, fuel cells, hydrogen storage, lithium ion batteries and electrochemical supercapacitors, Table 4 [166].

Solar cell devices can potentially alleviate the high energy demand. However, their complex fabrication and cost implication remains a critical challenge. CNTs have high optical absorption, they provide an ideal light-harvesting medium that has a wide range of direct band gap, excellent chemical stability and ballistic charge transfer [167]. A comprehensive review by Xuemei et al. [168], showed the application of CNTs in flexible photovoltaic devices which creates a great avenue in portable or wearable electronics, power-generated textiles, building-integrated photovoltaic systems, electric automobiles, unmanned aerial vehicles and space robots [168]. Satheish et al. [167], studied the effect of metal oxide charge transfer layers on the photovoltaic performance of CNT heterojunction solar cells. It was observed that the absorption of MWCNTs was higher than that of metal oxides, absorbing light over a wide spectral range (from UV to NIR regions). MWCNTs are shown to be an ideal candidate as active absorbing material for solar cells [167]. CNT composites for quantum dot sensitized solar cells have been developed with the power conversion efficiency of 5.70 % [169]. Yu et al. [170], fabricated graphene/SWCNTs as counter electrodes for dye-sensitized solar cells with up to 10.69 % power conversion efficiency [170]. Recently, the power conversion efficiency was improved to 11 % by Mousavi et al. [171], using −OH functionalized MWCNTs decorated with rhodium as a counter electrode [171].

Supercapacitors are the 21st century energy storage system that supersedes the power of batteries due to their high-power output, fast charge-discharge process and long cycling life [49]. They have various applications ranging from electric vehicle, pulse power systems to portable systems and devices [49]. Carbon-based supercapacitors are further characterized by more exceptional electrochemical cycling stabilities, rapid charge/discharge capacity, and high-power density [172].

Branched CNT/carbon nanofibers nanocomposites were synthesized using CVD method and applied as electrodes in supercapacitors. The supercapacitors carried a high value of specific capacitance and charge/discharge rate of 1 A g^{-1}. An exceptional cycling stability of 95.6 % was shown by the supercapacitor after 5000 charge/discharge cycles at a high current density of 20 A g^{-1}, and high energy density of 63.7 Wh kg^{-1} in EMIBF4 organic electrolyte [173]. Zhou et al. [174], fabricated hollow–structure NiCo hydroxide/ CNT composites for high performance supercapacitors. The incorporation of CNTs improved the structural stability of the electrode and displayed higher specific capacitance compared to that of NiCo hydroxide [174].

Remarkably, carbon-based supercapacitors have been recently considered to complement the fuel cells by improving their power density. The hybrid system can provide the steady-state and peak power demand. This makes supercapacitors an important component for fuel cell vehicles, which are considered the future in response to environmental, economic and political considerations [175]. The application of CNTs in the energy industry is a growing market and it will play a role in ensuring sustainable energy production.

3.3. Carbon nanotubes in the food industry

Food security is a major concern worldwide because its availability is highly dependent on other resources including water and energy. According to the FAO 2016 report, 815 million people are malnourished and 20 % of those people are Africans [188]. This is a matter of urgency and the solution is not only on increasing food production, but also on the preservation of the produced food to avoid wastage. Nanotechnology seeks to ensure security by addressing various areas within the food industry (i.e. increasing the shelf life of food, food safety, coloring, flavoring, and nutritional additives, and using the antimicrobial ingredients for food packaging) [189]. Nanomaterials have been shown to penetrate the cell walls and cell membranes of plants which aids plant

growth and development (Fig. 7) [190]. However, the challenge with CNTs is their hydrophobicity, which can be easily altered by functionalization. Triphati et al. [190], applied water soluble carbon nanotubes (wsCNTs) on Cicer arietinum plants. They observed an enhanced growth rate in every part of the plant including the roots, shoots and in branching with 6 mg· mL^{-1} wsCNTs. The growth on the wsCNTs treated plants was attributed to improved water absorption and retention [190].

A study on the enhancement of wheat plants by oxidized MWCNTs (o-MWCNTs) was performed. The o-MWCNTs significantly promoted cell elongation in the root system and increased the dehydrogenase activity, resulting in faster root growth and higher biomass production [191]. Furthermore, Martínez-Ballesta et al. [192], added MWCNTs on broccoli cells and observed an improved nutrient and water uptake and enhanced growth [192]. These observations open new perspective in the application of CNTs in the agricultural sector as growth regulators rather than the use of conventional fertilizers which are expensive to produce.

Monitoring of food products especially processed food is of paramount importance to ensure safe consumption and protect people from infectious diseases primarily caused by foodborne pathogenic bacteria. The 2018 listeriosis outbreak in South Africa, registered 216 deaths due to microbial food contamination. Therefore, the need for the development of portable devices that will detect biomolecules, diagnose diseases is imperative to give out early warnings to the consumers. CNT biosensors are potential candidates for food monitoring due to their reliability, cost-effectiveness, selectivity, sensitivity and rapid analysis. They can detect different analytes in food products as shown in Table 5. Choi et al., [193] developed a SWCNT biosensor to detect *Staphylococcus aureus,* which is one of the leading causes of gastroenteritis from the consumption of contaminated food. The SWCNT-based biosensors were developed using only 0.1 mg/mL of SWCNTs and showed great sensitivity towards *S. aureus* [193]. Sobhan et al. [194], fabricated a SWCNT biosensor to detect *Y. enterocolitica* in Kimchi (traditional Korean fermented food). This gram-negative bacterium can grow in refrigeration temperature proving that storing food in cool places alone is not enough to ensure safe consumption [194].

Furthermore, pesticides have been long used in the agriculture sector to prevent pests from affecting crop production. Monocrotophos (Ops) is a commonly used pesticide due to its toxicity towards the pests. However, its residues are found in crops which has a negative impact on food safety and human health. Consequently, a MWCNT-based biosensor was developed for fast and accurate detection of Ops. This sensor was evaluated on real vegetable samples and showed great sensitivity and low detection limits [195]. Diazinon is another type of Ops which is

widespread used in agriculture. It is as toxic and has adverse effects if consumed. A CNT-based biosensor was successfully used to detect it in fruit samples at low detection limits [196]. CNTs play a significant role in food safety and security by ensuring that the food products are of required quality for human consumption.

4. The global carbon nanotube market

CNTs have been shown to be advanced materials with the potential of addressing the WEF-nexus. This means that these materials need to be produced in large quantities to meet the demand. The CNT market is estimated to grow from $4.55 billion in 2018 to $9.84 billion by 2023 with a compound growth annual rate (CGAR) of 16.7 % [204] as more knowledge about the material is gained and new industries being forged. On account of diverse applications and low cost, MWCNTs dominate the market with a market share of 95 % [204]. The major manufactures of CNTs are the USA and China. China, being the largest global consumer of CNTs due to its increased industrial production. However, these manufactures are still using the conventional synthesis approach. This could be due to insufficient studies around the quality and quantity of CNTs that can be produced using green methods.

The global market of CNTs is generally segmented by MWCNTs, SWCNTs, and others (DWCNTs, FWCNTs). Currently, MWCNTs comprise the biggest share in terms of sales volumes, and production capacities [204]. Demand for MWCNT is mainly from composites, energy storage, and electronics industries [30,51,178]. There are several large companies with MWCNT production capacities of hundreds of tonnes per year who work to meet the growing demand (Table 6). The global market of CNTs is also segmented by production processes and applications. The CVD production process and application in electronics are expected to have the highest CGAR in the projected period (2018–2023) [178].

SWCNTs are vastly superior to MWCNTs in terms of their specific properties. However, they have not been used in industry until the last few years due to the lack of efficient mass production technology and high cost [179]. However, recent mass production technology is making SWCNTs more affordable for use in range of industries. The largest producer of SWCNTs is OCSiAl (European company), producing 75 tonnes per annum. The global CNT growth rate is low in South America, Middle East and Africa (Fig. 8) and this is also accounted for by the small number of CNT production companies in these regions (Table 6). Sabi-Nano (South African Company) is the only CNT manufacturing company in South Africa, revealing a slow growth rate and low CNT application
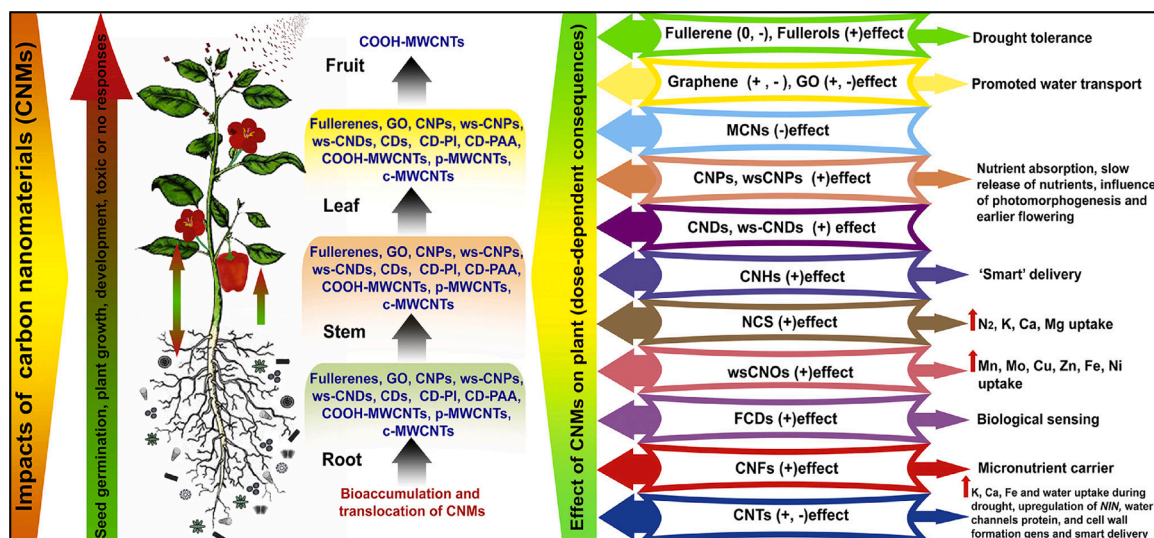

Fig. 7. Impacts of CNTs on plant growth and development [111].

Table 5
Application of CNT-based sensors in food products.

Nanomaterial	Analyte	Linear range	Detection limit	Ref.
SWCNTs	*S. aureus*	–	10^4 CFU/mL	[193]
SWCNTs	*Y. enterocolitica*	10^6 to 10^4 CFU/mL	10^4 CFU/mL	[194]
CNTs	Diazinon	1×10^{-4}–0.06 μM	4.5×10^{-4} μM	[196]
Cu-Co PBA/MWCNTs	nitrite	10–400 μM and 400–2100 μM	0.5 μM	[197]
c-MWCNTs /MONPs/ TCNQ/CHIT/GCE	xanthine	1.9 μM – 230 μM	0.20 μM	[198]
c-MWCNTs–SnO$_2$–GR–CS	ʟ-lysine	0.99 μM – 160 μM	0.15 μM	[199]
poly(L-aspartic acid)/MWCNT	Xanthine	0.001–0.004 μM and 0.005–50.0 μM	3.5×10^{-4} μM	[200]
MWCNTs/BMIMBF$_4$/NOR	Nitric oxide	0.23–4.76 μM	0.07 μM	[201]
β-cyclodextrin-CNT- TRGO	Ascorbic acid	20–30,000 μM	–	[202]
Ni/Cu/CNTs	Glucose	20 mM – 4500 μM	2.0 μM	[203]

Table 6
Global carbon nanotube production companies and their production processes and capacity.

Company	Country	Production Process	Annual production capacity (Metric tonnes)	Ref
Advance Nanopower Inc.	Taiwan	CVD	–	[206]
Arkema	France	CCVD	400	[207]
Arknano	China	CVD	1.2	[208]
Avansa Technology & Services	India	CVD	–	[209]
Beyond Pacific	USA	CVD	100	[210]
Bottom-up Technology Cooperation	India	CVD	–	[211]
Carbon Solutions Inc.	USA	CVD	–	[212]
CHASM Advanced Materials	Norman	CCVD	–	[213]
Cheaptubes	USA	CVD	–	[214]
CNano Technology Limited	USA	CCVD	500	[215]
HeJi Inc.	China	CVD	–	[216]
Hyperion Catalysis	USA	CVD	50	[217]
Jeio	Korea	CVD	100	[218]
Kleancarbon Inc.	Canada	CVD	1.0	[219]
Meijo Nano Carbon Co. Ltd	Japan	Enhanced direct injection pyrolytic synthesis (eDIPs)	–	[220]
NanoAmor	USA	CVD	–	[221]
Nano-C	USA	CVD	–	[222]
NanoChemazone	Canada	CVD	–	[223]
Nanocs	USA	CVD/ Arch discharge	–	[224]
Nanocyl	Belgium	CCVD	400	[225]
NanoIntegris	Canada	CCVD	–	[226]
Nanografi	Germany	CVD	–	[227]
NanoLab	USA	CVD	–	[228]
NaturalNano	New York	CVD	–	[229]
NanoShel	India	CVD	–	[230]
OCSiAl	Luxembourg	–	75	[231]
Raymor Industries	Canada	Radio frequency induction plasma /HiPco	–	[232]
Reinste Nano Ventures	India	CVD/arch discharge	–	[233]
SabiNano	South Africa	CVD	1.0	[234]
Showa Denko KK	Japan	CCVD	500	[235]
SkySpring Nanomaterials	USA	CVD	–	[236]
Toray Industries Inc.	Japan	CCVD	1.5	[237]
Unidym	USA	HiPco	1.5	[238]
US Research Nanomaterials	USA	CVD	–	[239]

- data not available.

industries in the country and Africa in general. The production and consumption of CNTs in the continents with low and medium growth will increase as new CNT industries emerge which will cause the demand to increase.

5. Challenges and perspective

The green synthesis of CNTs remains a challenge especially for industrial production due to insufficient literature around the quality and quantity of the CNTs produced. Green precursors predominately show synthesis of high quality CNTs. However, these are lab scale studies where a few grams are produced per day [74]. Albeit the greener approach for synthesis of CNTs addresses environmental concerns, the quality of the produced CNTs can be compromised at mass production. Therefore, extensive optimization is needed for large scale production of CNTs *via* greener routes. Several companies have adopted conventional CCVD method for CNT synthesis. This method has shown consistent quality and quantity of CNT production over years of application [27,50,61,205]. Interestingly, green synthesis methods can be incorporated into the existing CCVD processes. For example, plant derived catalysts can be used instead of metal catalysts. Furthermore, the catalysts can be synthesized *via* microwave irradiation, thus, reducing the reaction temperature, time and carbon footprint [179]. Optimization studies are being carried out at SabiNano (Pty) Ltd (South Africa), where synthesis methods reported by Mhlanga et al. [47], are modified to greener methods. A breakthrough in green synthesis methods and availability of research data would encourage most production companies to consider greener approaches for cost-effective production processes. This cost-effective production of CNTs will significantly reduce the price of CNTs which remains high. Thus, affording the development of advanced commercial products with the potential of solving several global challenges such as the WEF-nexus.

The application of CNTs to improve the performance of many products has led to an occupational and public exposure to these nanomaterials. Hence, it becomes a major concern to analyze the toxicity of CNTs and find the best suitable ways to counter those challenges. Although it is not in the scope of the study, toxicological effects of CNTs need to be thoroughly understood for their safe application. More especially in the WEF-nexus, where there is direct exposure and consumption of the water and food products treated by the incorporation of CNTs. Notably, the use of natural catalysts offers a sustainable solution, as the catalysts are organic and produce CNTs that are free from metal impurities. Consequently, studies need to be undertaken to understand whether CNTs synthesized *via* greener approaches are biocompatible. This would not only reduce their environmental impact but also ensure their safety to the consumers.

6. Conclusion

The sustainability of water, energy and food sectors can only be reached through the employment of innovative approaches. The use of environmentally friendly and renewable methods remains at the core of
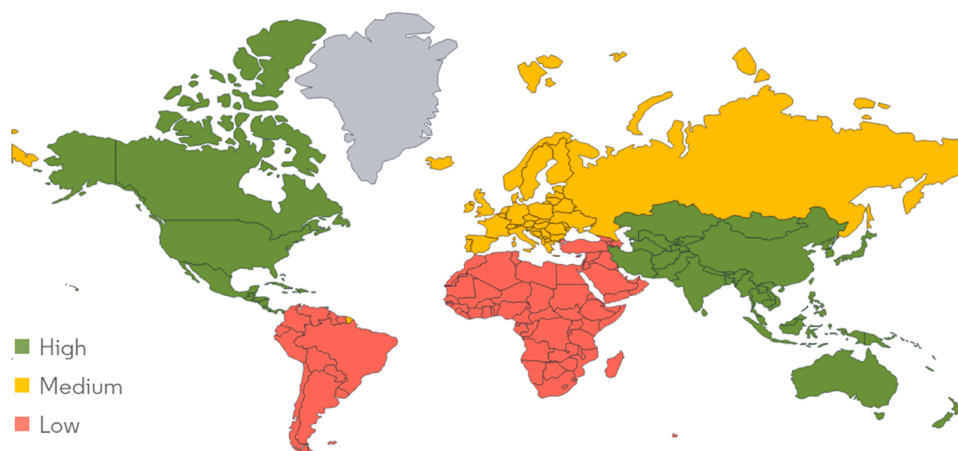
Fig. 8. CNT worldwide market growth rate, 2019-2024 [178].

sustainable development. This review revealed that greener processes have the potential of being competent and even better than conventional methods in the production of CNTs. However, more studies need to be undertaken to ensure the quality of CNTs is not compromised as well as the production capacity is viable for large scale application. CNTs synthesized using green processes make it possible to solve problems associated with the WEF-nexus without detrimental effects to the environment and life. As such, the choice of carbon precursors and the design of the CNT production processes must be well-crafted to sustainably address the challenges presented by the convoluted relationship between water, energy and food. Furthermore, the global demand of CNTs is increasing as new industries that utilize these materials emerge. Thus, the large-scale production of CNTs is inevitable and must be carried out in a way that will not pose a negative effect to the environment. Finally, the ethical and social implications of using CNTs in the WEF-nexus must be considered of great importance because public perception is the ultimate barrier to technology acceptance before it can be applied at full scale.

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgments

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