

Upcycling Waste Plastics into Carbon Nanomaterials: A Review

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ABSTRACT: Polymer production and utilization are currently widespread and have greatly improved people's standards of living. However, due to their stable and nonbiodegradable nature, postconsumer polymers pose challenging issues to the environment and ecosystems. Efforts are being made not only to contain the generation of polymer wastes and associated littering but, also, to find ways of utilizing them sustainably. Aside from mechanical recycling, which turns postconsumer polymers into new polymer products, and thermal recycling, which releases the thermal energy contained within waste plastics through combustion, chemical recycling converts waste polymers into feedstock for chemicals/materials/fuels production. This manuscript reviews prior work on a special application of the particular chemical recycling route that converts polymers into carbon-based nanomaterials. These materials feature extraordinary physical and chemical properties with tremendous applications potential. However, their production processes are both resource- and energy-intensive. Yet, by taking advantage of the high carbon content of waste polymers, as well as of their high energy content, a cost-effective, environmentally-friendly, and self-sustaining production of carbon nanomaterials can be achieved. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2014**, *131*, 39931.

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INTRODUCTION

Plastic Wastes Overview

Over the past 50 years the proliferation of nonbiodegradable man-made materials, such as plastics, has increased dramatically. Major applications of plastics include but are not limited to packaging, building and construction, transportation/automotive, and electrical and electronic components. As illustrated in Figure 1, the world production of plastics in 2011 reached a record high, 280 million tons, roughly two-thirds of which were contributed jointly by China (~23%), European Union (EU-27, 21%), USA (16%), and Japan (5%).¹ Correspondingly, more than 25, 31, 28, and 9 million tons of plastic wastes were generated in Europe,¹ USA,² China,³ and Japan,⁴ respectively.

Plastic Waste Management—Conventional Recycling Solutions

As most polymers are mass-produced and low-priced, they are readily discarded after use. Waste management is needed to minimize the plastic wastes and to reduce their environmental impacts, and typically, waste reduction, recycling (mechanical, chemical, and thermal) as well as landfilling are considered as common solutions.^{5,6} There are issues related with landfilling, however, such as those of (a) continuously decreasing available landfill space and (b) ubiquitous littering causing land and water pollution. Consider for instance the accumulation of

plastics refuse in the North Atlantic Subtropical Gyre,⁷ as well as the Great Pacific Garbage Patch,⁸ where the amount of non-biodegradable wastes (such as plastics and rubber) has increased for more than 100 times in the past 40 years.⁹ Further increases in recycling and reuse are therefore not only desirable but rather essential in addressing the aforementioned issues.

Recycling is to collect waste materials, process or re-manufacture them (either biologically, mechanically, chemically, or thermally) into new products, and to market those products (making the wastes recycled)¹⁰. Biological recycling takes place at the presence of microorganisms, such as bacteria, fungi, and enzymes, to degrade the plastics,^{11–13} whereas mechanical-, chemical- and thermal-recycling are human activity based. Table I summarizes the related terms and definitions used by different countries/regions on these human-activity-involved recycling. Mechanical recycling processes plastic wastes (mainly thermoplastics, e.g. PE, PP, PET) into secondary raw material (termed recycle) for common plastics.¹⁴ No significant changes in the chemical structures occur during mechanical recycling. It starts with materials collection, followed by identification, sorting, grinding, washing, drying, separating, agglomerating, extruding/compounding, and eventually pelletizing. Chemical (or feedstock) recycling partially converts some plastics into their monomers, or into hydrocarbon fractions, which can then be used either as feedstock for polymerization, or in other chemical processes. Thermal recycling

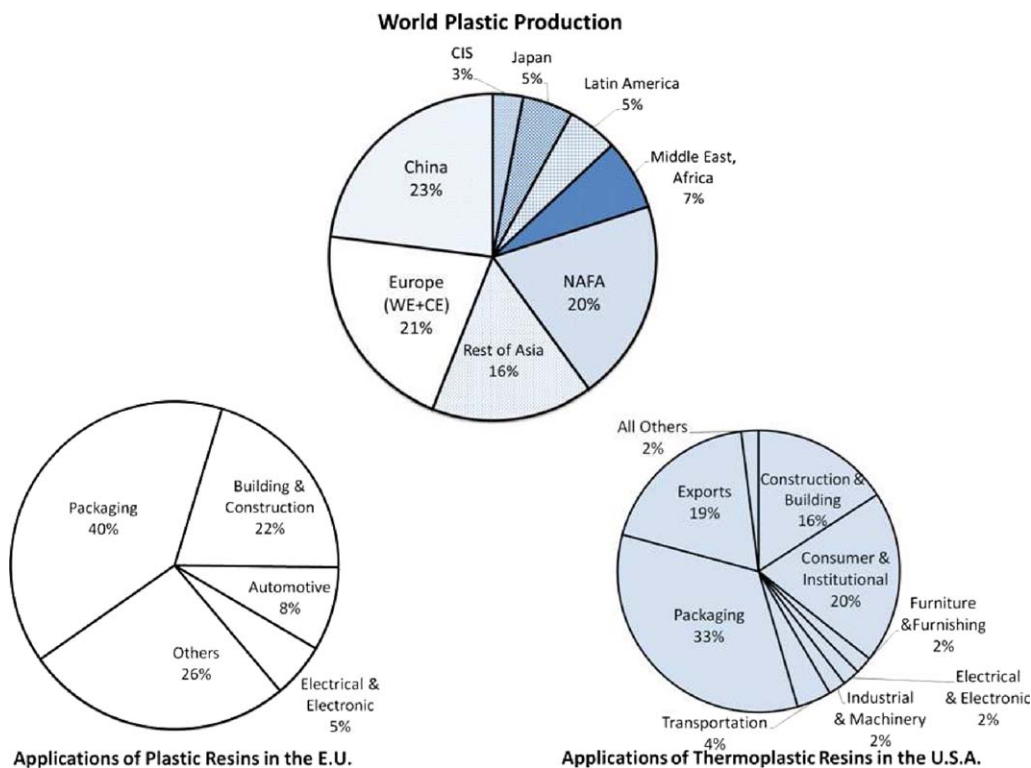


Figure 1. World production of plastics in 2012, with their typical applications in E.U. and the U.S.A. Data cited from Refs. 1 and 2. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Summary of Plastics Recycling Methods.

International Standard Organization (ISO) 15270	Japan	EU	American Society for Testing and Materials (ASTM) International D7209-06	Methods, cited from Ref. 15
Mechanical recycling	Material recycling	Mechanical recycling	Primary recycling	Recycling isolated/segregated plastics to make (primary) plastic raw materials/products
			Secondary recycling	Recycling mixed plastics to make (primary) plastic raw materials/products
Chemical recycling	Chemical recycling	Monomerization/ Feedstock recycling	Tertiary recycling	Monomerization
				Blast furnace reducing agent
				Coke oven chemical feedstock recycling
Energy recovery	Thermal recycling	Thermal recycling	Quaternary recycling	Gasification / Liquefaction to Chemical Feedstock and/or Fuel
				Cement kiln
				Waste power generation/ incineration/Refuse Derived Fuel/Refuse Paper & Plastic Fuel

takes advantage of the high energy content of the plastic wastes (as they are hydrocarbons in nature, which will be discussed later), and recovers the energy through thermal processes (sometimes also called incineration, or waste-to-energy conversion).

Recycling enables substitution of raw materials with post-consumer materials, which would otherwise be obtained from increasingly scarce natural resources, such as metals or petroleum, in cases of both feedstock materials and input fuels. Therefore, recycling is one of the favorite methods in the waste management hierarchy. However, the rate of global recycling is still not as high as it would be economically and environmentally desirable, and it varies in different geographical areas. Japan has the highest recycling rate (78%, 2011),⁴ followed by EU,¹ (59%, 2011) and China (48%, 2011),³ while U.S. has the lowest recycling rate (EPA 8%, 2011, does not include the parts being recycled through combustion).² Thermal recycling (energy recovery) and mechanical recycling are the predominant methods nowadays,¹⁵ while chemical (feedstock) recycling is significantly utilized only in limited countries (areas), such as in Japan.

There are many factors that jointly lead to such low recycling rates, including: (1) the lack of access to recycling facilities,¹⁶ (2) the complexity of waste identification, separation and cleaning,^{17,18} and (3) the limited municipality and consumer compliance and the limited markets for the recycled plastics.¹⁹ Public awareness, government legislation, and technology improvements in the recent past have partly addressed this problem.¹⁹ However, as recycled plastics often have compromised properties, they cannot be used in their original applications.²⁰ Recycling therefore may not be treated as a sustainable solution by itself, especially taking into account that such efforts are affected largely by the overall economics: the recycling market is cyclic and depends on the strength (and viability) of the economy.²⁰ Identifying high-capacity markets for recycled products is problematic, thus creation of a new market for recycled plastics, particularly leading to high-value products, would be desired to spur interest in recycling and reuse of such polymers. It will also then be beneficial for the recycling markets, the associated infrastructure and, eventually, the public's mindset for recycling.

Plastic Waste Management—Advanced Upcycling Solutions

It is well known that the products of chemical recycling are majorly hydrocarbons, which can be used either as fuels for power generation, or as feedstocks (such as monomers) to produce new materials. It has been suggested that, in the traditional chemical industry process, raw materials account for 60% to 90% of production costs,²¹ thus inexpensive feedstocks yielded from chemical recycling enable a new pathway to lower the cost of manufacturing value-added products. The processes involved herein will then be termed upcycling process, as the quality/value of the final products is upgraded, and there are studies and reviews on related topics, such as upcycling plastics into chemicals for monomer feedstock,^{22–27} fuels,^{22,28–33} etc. Due to the fact that carbon is the major constituent of plastics (see Table II), the waste plastics can therefore provide a carbon source for carbon-based value-added products.

Such products include but are not limited to light hydrocarbons,^{34–37} carbon black/activated carbon,³⁸ carbon fibers,^{39,40}

Table II. Carbon Contents in Major Commercial Polymers (Plastics)

Polymer	Molecular formula	Carbon content (in wt %)
Polyethylene (PE)	(C ₂ H ₄) _n	85.6
Polypropylene (PP)	(C ₃ H ₆) _n	85.6
Polystyrene (PS)	(C ₈ H ₈) _n	92.2
Polyethylene terephthalate (PET/PETE)	(C ₁₀ H ₈ O ₄) _n	62.6
Polyacrylonitrile (PAN)	(C ₃ H ₃ N) _n	67.9

fullerenes,^{41–43} carbon nanotubes,^{44–48} and graphene.⁴⁹ Nowadays, there are various applications of graphite and carbon black at annual production rate exceeding 1 million tons. Meanwhile, there are certain carbon allotropes being considered as advanced materials, as they possess unique properties, in nanoscale, leading to unprecedented functions with numerous applications. Examples of these carbon allotropes include, but are not limited to: nanodiamond, fullerene, carbon onion, carbon nanotubes, and graphene, see Figure 2. Therefore converting waste plastics into such value-added products is an excellent example of upcycling.

This study will focus on the efforts, made in recent years, to explore ways of upcycling plastic wastes into carbon nanotubes (CNTs), a category of fullerenic materials. The background of CNTs will be introduced first, including their types, properties and applications. Typical methods of producing CNTs will then be reviewed, followed by existing processes that utilize waste plastics as carbon feedstocks for their production. At the end of this review, challenges faced by upcycling process and suggested solutions will be discussed.

Carbon Nanotubes: Synthesis, Properties, and Applications

CNTs consist of coaxial tubular graphene sheets, with lengths in the order of micrometers (1×10^{-6} m) and diameters in the order of nanometers (1×10^{-9} m).^{50,51} Carbon nanotubes (CNTs) have been studied for more than 20 years since being reported by Iijima.^{52–54} The appealing properties of CNT fall essentially in four categories: (i) Electrical: semiconducting or metallic behavior, 100 times more conductive than copper.^{55,56} (ii) Mechanical: very high tensile strength (100 times higher than steel).^{57–62} (iii) Thermal: high thermal stability and thermal conductivity.⁵⁷ (iv) Chemical: chemically inert while affinitive with chemical/biological functional groups.^{63–65} (v) Structure: ideal

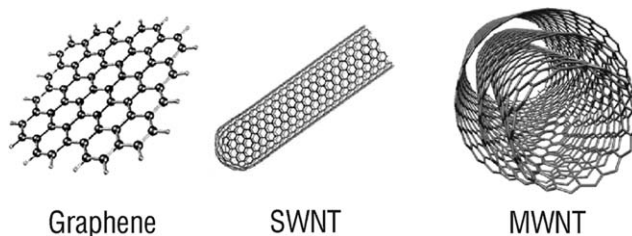


Figure 2. Examples of carbon-based nanomaterials. Reprinted from Ref. 52, with permission from American Chemical Society.

Table III. Potential Applications of Carbon Nanotubes

	Large-volume applications	Limited-volume applications (mostly based on engineered nanotube structures)
Present	Battery electrode additives (MWNT); composites (sporting goods; MWNT); composites (electrostatic discharge applications; MWNT)	Scanning probe tips (MWNT); specialized medical appliances (catheters) (MWNT)
Near term (less than 10 yr)	Battery and supercapacitor electrodes: multifunctional composites (3D, damping); fuel-cell electrodes (catalyst support); transparent conducting films; field emission displays/lighting; CNT-based inks for printing	Single-tip electron guns; multitype array X-ray sources; probe array test systems; CNT brush contacts; CNT sensor devices; electromechanical memory device; thermal-management systems
Long term (beyond 10 yr)	Power transmission cables; structural composites (aerospace and automobile, etc.); CNT in photovoltaic devices	Nanoelectronics (FET, interconnects); flexible electronics; CNT-based biosensors; CNT filtration/separation membranes; drug-delivery systems

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one-dimensional (1D) system with anisotropic properties, with extremely high aspect ratios.^{66–68}

Potential applications of nanotubes nowadays have become overwhelmingly widespread,^{47,69–72} examples of which include but are not limited to composites,^{73–78} electrodes,^{79–84} electronics,^{85–92} photovoltaic devices,^{93,94} biological (Table III).^{95,96} Entering into the growth phase of product life cycle,^{47,72} increasing usage of CNTs in commercial products is part of the beginning of the nanotechnological revolution. The global CNT production capacity in 2011 was estimated to be larger than 4500 tons,⁴⁷ and the production capacity is projected to be over 14,000 tons by 2016 at a compound annual growth rate (CAGR) of 30.6%.⁹⁷ Their revenues totaled \$192 million in 2011 and are projected to grow over the next five years at CAGR of 22.4%, reaching \$527 million by 2016.⁹⁸ Expanding markets for CNTs' large volume applications place ever-increasing demands on lowering their production costs to the level acceptable by the end-user applications.⁷² It is estimated (in Figure 3) that the mass application of CNTs will be facilitated only when the price of CNTs approaches that of conductive carbon black.

CNTs are formed under appropriate conditions at the presence of carbonaceous feedstock, heat and, typically, catalysts. There are four major different methods for the production of CNTs: arc discharge, laser ablation, chemical vapor deposition (CVD), and combustion (flame) synthesis. After the discovery of multi-wall and then single-wall carbon nanotubes by carbon vaporization in electric arcs,^{99,100} several varieties of this technique such as metal doping, different configurations of the electrodes or different types of carbon sources have been described.^{101,102} Synthesis of improved-quality single-wall carbon nanotubes have been reported using rather expensive laser irradiation of metal-doped carbon targets.^{103–105} Both methods can provide high quality CNTs; however, their yields are limited, mostly due to the available dimensions of the reactors and high energy consumption. The generation of well-aligned and uniform mats of multiwall as well as single-wall nanotubes, by chemical vapor

deposition (CVD), i.e., growing materials from pyrolyzing hydrocarbons to the gas phase and depositing them onto solid surfaces, has been demonstrated.^{106–108} Additional technologies, such as plasma and microwaves, have been utilized to facilitate the CVD growth. The need for continuous external heating, which consumes large amounts of electric power, is a serious drawback for CVD's use on an industrial scale (e.g., in a fluidized bed reactor^{109–117}) for all but the highest end-value applications.^{118–120} Another drawback of CVD is that most experiments reported require the injection of H₂, a highly flammable gas, to promote the reduction conditions needed to synthesize nanotubes.

Carbon nanotubes have also been generated by combustion synthesis. Compared with CVD pyrolysis, combustion synthesis has the advantage of being an exothermic process, i.e., not external heating (i.e., consumption of electricity) is needed. Carbon

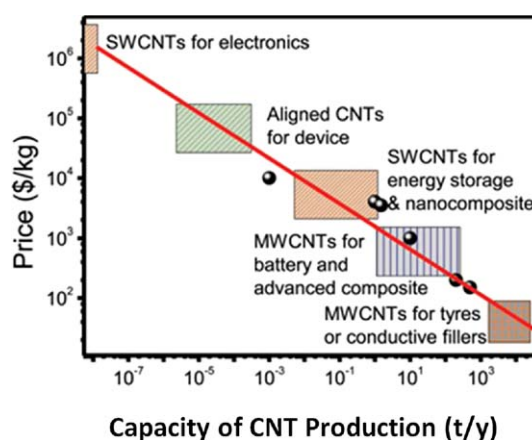


Figure 3. Suggested relations between the CNT production capacity (ton/year) and predicted unit price (\$/kg). Reprinted from Ref. 73, with permission from Wiley. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

donors such as carbon monoxide or small hydrocarbons are generated *in situ* by means of a combustion process while a catalyst (either fixed or floating) is introduced, in a fashion similar to the above-described CVD techniques. The use of suitable combustion equipment and precise settings of operating conditions such as fuel/oxygen ratio, dilution with inert gas, fresh gas velocity, and pressure allows for accurate control of the CNT-forming process. Sufficient understanding of the correlations of the parameter space characterizing the nanotubes is expected to lead to a large-scale and high-yield production process of carbon nanotubes with well-defined and homogeneous properties.^{121–123} Formation of multiwall carbon nanotubes by immersion of metallic substrates in methane- and ethylene-fueled, co-flow diffusion flames,^{124–136} and of single-wall nanotubes by addition of metallocenes to the fuel-stream of acetylene- or ethylene diffusion flames has been reported.¹²⁴ Multiwall carbon nanotubes were also observed in opposed-flow methane-oxygen diffusion flame with and without immersion of metal catalysts.^{137–139} CNTs of different morphologies can be selectively produced with premixed flames of fuels having dissimilar chemical structures.¹⁴⁰ Small amounts of single-wall carbon nanotubes (SWCNT) were identified in the condensed matter of partially premixed sooting acetylene and ethylene flames in which metallocenes ($\text{Fe}(\text{C}_5\text{H}_5)_2$ and $\text{Ni}(\text{C}_5\text{H}_5)_2$) were vaporized and added to the flame-feed,¹⁴¹ whereas multiwall carbon nanotubes (MWCNT) were observed in corresponding benzene flame. Commercially, large-scale production of SWCNTs of good quality has been achieved by using natural gas or methane as fuels.¹⁴² In-line filtration allowing for continuous operation of SWCNT and product recovery was implemented using commercial metallic cartridges.^{143,144}

Current technologies for large-scale production of CNTs (either CVD or combustion synthesis) require intensive consumption of premium feedstocks, such as methane,^{145,146} ethylene,^{147,148} carbon monoxide (CO),^{149–151} and hydrogen (H_2).¹⁵⁰ Therefore using waste plastics as inexpensive feedstocks can contribute to the cost reduction of CNT production, which may accelerate CNTs large-scale use in consumer/industrial products. This will in turn further stimulate the utilization of post-consumer waste plastics.

WASTE PLASTICS AS LOW COST FEEDSTOCKS FOR CNT GROWTH

The concept of synthesizing CNTs using solid polymers was first reported about 16 years ago.^{152–155} Since then there have been numerous efforts on further advancing such low cost CNT synthesis route. Work has been done with regards to types of plastics, conversion processes, growth conditions, catalysts, and quantities and qualities of the resulting CNTs. Various processes have also been explored for such conversions, and there is one (and the only one up-to-date) review article covering this topic.¹⁵⁶ Nevertheless, that work examined processes based on the reactor types alone (autoclave, crucible, fix bed (tube furnace, muffle furnace), moving bed, fluidized bed, etc.). Instead, the authors hereby focus on both the common and on the differentiating features of the existing waste-plastics-to-CNT conversion processes. Thereby, such conversions may be classified into two categories: (1) one-pot conversion where synthesis of

CNTs occurs upon the *in situ* formation of carbon feedstocks from the solid plastic waste; and (2) stepwise conversion where synthesis of CNTs occurs after the formation of carbon feedstocks from the solid plastic waste. Typical processes are summarized in Table IV, which includes their representative CNT products, related conversion conditions, as well as corresponding references.

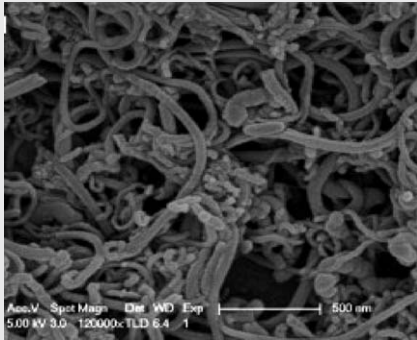
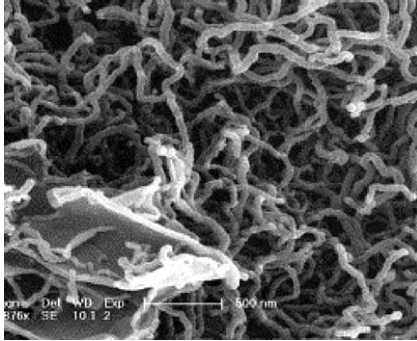
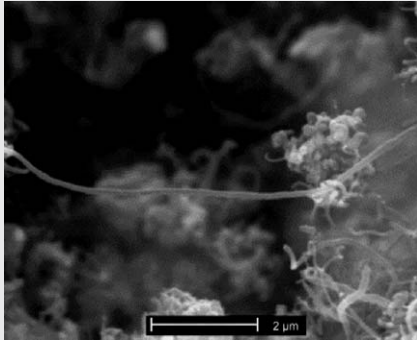
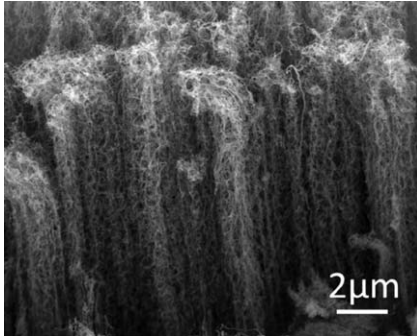
One-Pot Conversion of Plastics into CNTs

One-pot synthesis of carbon nanotubes typically starts with solid polymers which are mixed with catalysts. A heat resource is then applied to catalytically decompose (pyrolyze) the plastics. The decomposed products, either in liquid or gaseous phases, serve as carbon sources for the growth of CNTs on the catalysts. Polymers, such as polyethylene (PE),^{157–161} polypropylene (PP),^{160,162–168} polystyrene (PS),¹⁶³ polyvinyl alcohol (PVA),¹⁶⁹ polyvinyl chloride (PVC),¹⁷⁰ polytetrafluoroethylene (PTFE),¹⁷¹ polycarbosilane (PCS),¹⁷² phenol formaldehyde (PF),¹⁷³ and polyethylene terephthalate (PET),¹⁶⁴ etc. have been studied using this method (Table IV). Various catalysts have been examined, including transition metals in either elemental form (nickel,^{157,174,175} iron,^{163,172} etc) or in chemical compound form (nickel oxides,^{168,176–178} ferrocene,^{158,167} ferrous chloride,¹⁷¹ cobalt acetate,¹⁵⁹ etc.) among others. Heat is supplied by either electric furnaces (fixed beds,^{163,178,179} autoclaves,¹⁷⁵ and fluidized beds,^{160,164}), or by combustion of fuels.^{162,166,168,177,180}

This one-pot synthesis features simultaneous plastic degradation and the CNT synthesis, and efforts have been made to reveal how it proceeds. Jiang et al. proposed a possible reaction mechanism, using PP as the sample feedstock¹⁶⁶ with nickel-based catalysts, as illustrated in Table IV. With the presence of catalysts, plastics are degraded (decomposed), by active intermediates of carbenium ions, but not free radicals which play a major role in noncatalytic thermal decomposition of plastics. It is suggested that the resulting “free carbons” from catalytic pyrolysis of plastics will then dissolve into the catalyst, diffuse through and then precipitate at its surface to form CNTs. Such one-pot synthesis can be greatly affected by utilizing a combination of metal catalysts with solid acids (such as organically-modified montmorillonite (OMMT)^{166,168} or zeolite¹⁷⁷), chlorinated compounds (such as CuCl),¹⁸⁰ and/or activated carbon.¹⁸¹ Solid acids are suggested to provide the intermediate proton acidic sites, which will assist breaking down the molecular chains of the plastics^{166,182} favoring the formation of CNTs. On the other hand, chlorine radicals could promote the dehydrogenation and aromatization during the degradation of the plastics,¹⁸³ resulting in more carbon nanofiber and less CNTs.¹⁸⁰ Activated carbon plays multiple roles in the conversion of plastics into CNTs,¹⁸¹ such as (1) to absorb and to be functionalized by the fragment radicals from the plastics decomposition products, (2) to promote the formation of light hydrocarbons and aromatic compounds (especially the diaromatic and polycyclic hydrocarbons) through the fragment radicals as well as the aromatic intermediates, and (3) to further assist catalyzing the dehydrogenation and aromatization of the aromatic intermediate or PAHs.

Different reaction conditions (temperatures, catalyst compositions/concentrations/loads, reaction durations, etc.) usually lead

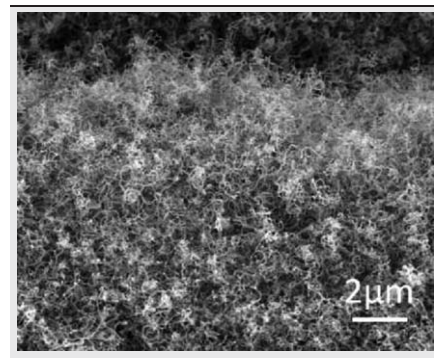
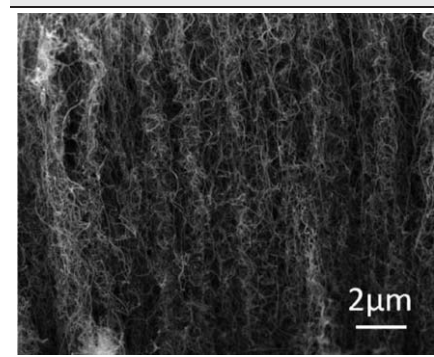
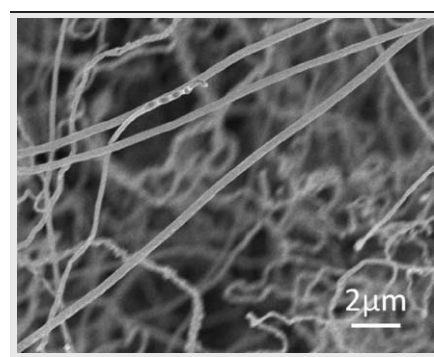
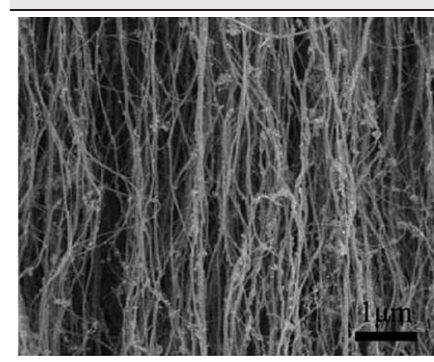
Table IV. Summary of Representative Waste-to-CNT Conversion Processes

Representative CNT products (taken by scanning electron microscopy)	Polymer feedstock; catalyst/temperature/synthesis duration; process description	Research groups, references/permission
	PE + ferrocene + MAPP/700°C/12 hr; plastics mixed with catalysts in an autoclave, heated by electronic furnace, leading to simultaneous plastic decomposition and CNT formation	Kong and Zhan. ¹⁵⁸ Reproduced from Ref. 158, with permission from Elsevier.
	PP + Ni ₂ O ₃ + OMMT + MAPP/830°C/unknown duration; plastics mixed with catalysts in a crucible, heated by flame, leading to simultaneous plastic decomposition and CNT formation	Tang et al. ¹⁵⁹ Reproduced from Ref. 159, with permission from Elsevier.
	PP/PE/PET + quartz-rich sand or alumina particles/450-850°C/unknown duration; plastics mixed with catalysts within a fluidized bed, heated by electronic furnace, leading to simultaneous plastic decomposition and CNT formation	Arena and Mastellone. ¹⁶⁰ Reproduced from Ref. 160, with permission from Elsevier
	PE + stainless steel meshes/800°C/1 min; plastics were first pyrolyzed in quartz tube, the resulting gaseous carbon feedstock were then channeled into a separated reactor where located the catalysts, both heated by electronic furnaces, leading to sequential plastic decomposition and CNT formation	Zhuo et al., unpublished work

to CNTs with various quantities and qualities. In addition to existing reviews on such topics,¹⁵⁶ the authors would like to briefly discuss the one of the features this one-pot synthesis processes: the CNT production yield. It is worth mentioning, that there are at least three different definitions in production

yield: the mass production rate of CNTs (unit: g/h), the mass of CNTs over the mass of the catalyst (unit: g/g), and the mass of CNTs over the mass of the feedstock (unit: g/g or %). The former two definitions are commonly used in traditional CNT production processes, as the corresponding focuses are either on

TABLE IV. Continued

Representative CNT products (taken by scanning electron microscopy)	Polymer feedstock; catalyst/temperature/synthesis duration; process description	Research groups, references/permission
	PP + stainless steel meshes/600-750°C/1 min; plastics were first pyrolyzed in quartz tube, the resulting gaseous carbon feedstock were then channeled into a separated reactor where located the catalysts, both heated by electronic furnaces, leading to sequential plastic decomposition and CNT formation	Zhuo et al., unpublished work
	PS + stainless steel meshes/800°C/1 min; plastics were first pyrolyzed in quartz tube, the resulting gaseous carbon feedstock were then channeled into a separated reactor where located the catalysts, both heated by electronic furnaces, leading to sequential plastic decomposition and CNT formation	Zhuo et al., unpublished work
	PET + stainless steel meshes/1000°C/1 min; plastics were first pyrolyzed in quartz tube, the resulting gaseous carbon feedstock were then channeled into a separated reactor where located the catalysts, both heated by electronic furnaces, leading to sequential plastic decomposition and CNT formation	Zhuo et al. ¹⁹⁴ Reproduced from Ref. 194, with permission from American Chemical Society
	PVC + ferrocene/800°C/0.5 hr; ferrocene was first sublimated in first reactor, and blown into second reactor where plastics were pyrolyzed; the resulting gaseous carbon feedstock/catalyst mixtures were then channeled into third reactor, all heated by electronic furnaces, leading to sequential plastic decomposition and CNT formation	Luo et al. ¹⁶² Reproduced from Ref. 162, with permission from Springer

the end products (CNTs) or on the activities of the catalysts. In waste-to-CNT conversion processes, however, the third definition is predominately used, as the focus of such processes is on conversion efficiency. That is why the data compiled from all different processes is not directly comparable. The yield could

be as low as several percent (e.g. 5% in Ref. 178), and this number can reach 50% or higher,^{156,178} under optimum conditions. Such a high yield conversion is achieved typically in a setting where the mix of plastics and catalysts is enclosed within a container, such as a crucible. Comparing with an open system

such as in a quartz tube reactor, where the pyrolyzed products (usually in gaseous phase) are carried away by the processing gas, this enclosed environment enables a longer reaction time between the plastics pyrolyzates and the catalysts. Therefore this could be a reason for such high conversion rate. High temperatures ($>700^{\circ}\text{C}$) and compound-based catalysts are also found to lead to high CNT yields.^{158,178}

The impurities in the CNTs generated by both one-pot processes and stepwise processes usually consist of residual catalysts and amorphous carbon. Impurities typically account for 10 wt % of the as-produced CNTs. The lowest amount of impurity reported was 0.3 wt %¹⁷⁸ as determined by thermogravimetric analysis (TGA), a common method to investigate the impurity and thermal stability of CNTs. In order to keep the number of impurities low, efforts should be made to optimize process parameters as: the components and dispersion of catalysts, the feeding rates of plastics, the reaction temperature, etc., for the catalysts be kept active and produce CNTs with minimum defects.

Stepwise Conversion of Plastics into CNTs

The stepwise conversion process features sequential reactions, typically starting with the thermal decomposition of plastics. As a second step, the resulting gaseous products (hydrocarbons) are then channeled downstream where they react with catalysts to form CNTs.¹⁸⁴ This is the method applied in the first reported explorations of plastics to CNT conversion,^{153–155} since then additional works have been done to investigate the underlying fundamentals as well as to advance the technology of such conversion processes. The plastics-to-CNTs conversion mechanism is rather complicated, as it combines the steps of high temperature plastic cracking,^{185,186} high temperature oxidation/combustion,^{187,188} thin film formation through chemical vapor deposition and gas-solid chemical conversion through heterogeneous catalysis.^{50,189,190} A comprehensive review has been done by Bazargan and McKay, addressing among other topics this formation mechanism,¹⁵⁶ whereas this concise review highlights significant technology developments.

An inherent feature of such a stepwise process is that it enables individual controls over its subprocesses. That is to say, decomposition (pyrolysis) of plastics can be processed without interference with the synthesis of CNTs. For instance, Yang et al. employed a three-stage reactor, where catalysts (ferrocene) was first sublimated, in the first reactor, before entering the second reactor, where plastics (either PE, PP, or PVC) were decomposed.¹⁹¹ The mixture of the formed catalysts and the gaseous pyrolyzates was then channeled into the third reactor, where CNTs were formed and collected. Their setting enabled independent controls of reaction temperatures ($120\text{--}140^{\circ}\text{C}$ for ferrocene sublimation, 450°C for plastics decomposition, and $800\text{--}850^{\circ}\text{C}$ for CNT growth). With reaction time in the order of 10 min, lengths of CNTs were typically in the order of $100\ \mu\text{m}$. It is not clear, however, whether the iron particles (sublimation products of ferrocene) only catalyzed the growth of CNTs, or they also participated in the thermal decomposition/pyrolysis of plastics.

An improved two-stage stepwise process was investigated by Liu et al.¹⁹² Polypropylene (PP) was catalytically pyrolyzed over

HZSM-5 zeolite in a screw kiln reactor, and the resulting pyrolysis gases were subsequently decomposed over nickel catalysts in a moving-bed reactor. CNTs and hydrogen were produced simultaneously, and it was found that 700°C was the optimum decomposition temperature, in terms of maximum productions of both CNTs and hydrogen with a synthesis temperature of 750°C . Similar work was reported by Wu et al., where a fixed-bed two-stage reaction system was used.¹⁹³ Waste plastics samples were pyrolyzed in N_2 in the first stage at 500°C , and the resulting compounds were further used to synthesize CNTs at 800°C in N_2 . Water was injected into the second reactor, where catalysts (Ni/Ca-Al or Ni/Zn-Al) were also present, to jointly facilitate the CNT formation.

Recently, the authors developed a hybrid reactor, combining sequential feeding, pyrolysis, premixed-combustion, and CNT synthesis process (Zhuo et al.^{194–200}) To achieve conversion of solid hydrocarbon fuels to CNTs, quantities of a solid fuel, in pelletized form, were first thermally pyrolyzed into a stream of gaseous decomposition products (pyrolyzates) inside a pyrolyzer furnace at 600 to 800°C , depicted on the left side of Figure 4. Pyrolysis occurred in inert nitrogen, to prevent the ignition and combustion of the pyrolyzates therein. The pyrolyzates then flowed through a mixing venturi where they were thoroughly mixed with preheated oxygen-containing gases. The oxygen/pyrolyzate charge autoignited at the exit of the venturi, forming a premixed flame. The fuel/oxygen ratio in the venturi was set to be fuel-rich (i.e., oxygen deficient) by adjusting the amount of oxygen introduced to the venturi. This ensured that there were large amounts of carbon-bearing compounds (CO and unburned aliphatic hydrocarbons), hydrogen, and water vapor in the combustion effluent, as needed for the efficient growth of CNTs. The combustion effluent then entered the second furnace where fixed catalyst substrates were also preinserted (such as stainless steel screens or honeycombs). Growth of CNTs occurred on the catalyst substrates at 600 to 1000°C . To prevent soot, which may be generated in the flame, from entering the second furnace and inadvertently depositing on the catalyst substrates and contaminating them, a high-temperature ceramic barrier filter had been inserted at the entrance of the CVD

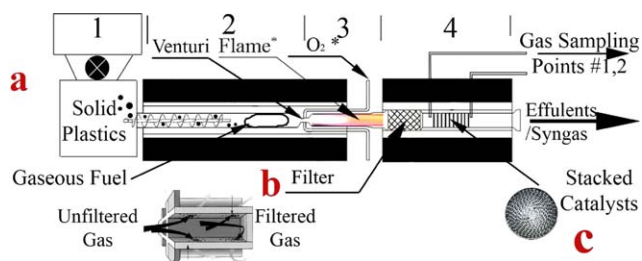


Figure 4. a: Four-stage laminar-flow, electrically-heated, muffle furnace, used for proposed work; zone 1: steady-state, steady flow, continuous feeding stage, zone 2: pyrolysis stage, zone 3: combustion stage*, and zone 4: CVD-synthesis stage. b: Ceramic (SiC) honeycomb filter manufactured by Ividen. c: stacked catalyst in the forms of either woven screens or honeycomb structures. *: A flame is present at the combustion stage only when oxygen-containing gases are added to the venturi. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

synthesis chamber, see Figure 4. This feature allowed operation of the apparatus above and beyond the equivalence ratio ϕ (defined as $(m_{\text{fuel}}/m_{\text{oxygen}})_{\text{actual}}/m_{\text{fuel}}/m_{\text{oxygen}})_{\text{stoichiometric}}$) that corresponds to the soot onset thresholds.^{140,201} The ceramic filter may be periodically thermally regenerated *in situ*, at 1000°C in air, to burn any entrained soot particles.

Another unique feature lie on the design of the venturi section, which enables the introduction of additional materials, such as one or more gases (e.g., oxidizing agents such as oxygen gas, chlorine gas, carbon dioxide, any other gas containing oxygen, and the like), or catalysts (such as ferrocene) to mix with the gaseous decomposition products, which enter the venturi section.

Use of stacked catalyst substrates (screens or honeycombs) will allow not only for high-yield production of CNTs but, also, for selective and detectable depletion of some carbon-bearing gases in the combustion effluents as CNTs form. Sampling the gases before and after the catalyst substrates from sampling ports 1 and 2, as illustrated in Figure 4, can help determine which gases are likely growth agents for CNTs. In summary, this process incorporates the following unique features: (a) It pyrolyzes or gasifies solid wastes to generate gaseous carbon-bearing components. (b) It affords control of the fuel-to-air ratio in the venturi from fuel-lean to fuel-rich and, to the limit, to purely pyrolytic conditions at the absence of oxygen. (c) The pyrolysis and the CNM synthesis occur in two separated stages, which afford individual control of the subprocess conditions. (d) The risks of handling highly reactive gases (such as H₂), and/or highly toxic gases (such as CO), are eliminated as they are generated *in situ* in an inert gas. Finally, (e) this process can generate a hydrogen-enriched syngas.

Moreover, this process can be designed to be energy efficient.¹⁹⁴ For instance, Jinno et al.²⁰² measured the heat of pyrolysis of polyethylene (PE) to be 254 kJ/kg, independently of the heating rate. Comparing this heat of pyrolysis with the heating value (energy content) of this polymer, which is 46,000 kJ/kg,²⁰³ it becomes evident that a small fraction of the heat released during combustion may be fed back and pyrolyze this solid fuel. Similarly, the energy fluxes necessary to pyrolyze polypropylene (PP) and polystyrene (PS) are 44,000 and 40,200 kJ/kg, respectively.²⁰² Thus, the energy balance of the proposed process is overwhelmingly positive. A portion of the heat released during the combustion of the polymer pyrolyzates may be fed back to the pyrolyzer through a heat exchanger to gasify incoming precursors, whereas the remaining heat may be used elsewhere as process-heat or to generate electricity. Regarding the environmental impact of this process, it should be stated that the use of premixed flames, high temperature filtration, and possibly a post-process afterburner can drastically reduce unburned or partially burned health-hazardous species, as shown before in the author's laboratory.^{204–210} The generation of the greenhouse gas CO₂ could be an issue when waste polymers are used as feedstocks. However, by using such waste hydrocarbon-based post-consumer products, the conventionally used, more valuable premium hydrocarbons are merely substituted, and thus, CO₂ emissions (that would have been released during their extraction and processing) are avoided. In addition, there are also benefits

to cleaning the earth of nonbiodegradable wastes. Generating value-added products, such as CNMs, from recycled waste plastics is thought to enhance the motivation for recycling, lower the cost of the products, and in turn, help overcome the hurdles of their large-scale use in consumer and industrial applications.

Challenges and Possible Solutions

One of the major challenges in plastic-wastes-to-CNT up-cycling comes from the nature of waste feedstocks, where there is a lack of consistent and reproducible supply of carbon feedstock with controlled quality (in terms of compositions and impurities). Feedstocks from recycling streams are typically mixtures of various types of plastics.¹⁹ The resulting pyrolysis products, of different compositions, are known to affect the formed CNTs, while the detail correlations are yet to be revealed. What is more, waste plastics containing fillers and/or other additives could have detrimental effects on the quality of formed CNTs.²¹¹ These two problems get further complicated when there are variances among different batches of waste plastic streams.

Another challenge of such up-cycling processes lies on the process complexity. Comprehensive scientific investigations are still needed in both one-pot and stepwise methods, especially those involved on the driving force behind the proposed waste-to-CNT up-cycling process. In the case of stepwise method, for instance, whereas gaseous feedstocks have been correlated directly to CNT products,^{212–216} the fact that these feedstocks pyrolyze at the high reaction temperatures therein (>400°C) to form intermediate products has been largely ignored.^{217–219} Therefore, the CNT catalysts therein have experienced a blend of carbon-bearing gases, which are very different from the input feedstocks. Hence, it remains unresolved whether the CNT synthesis process consumes the carbon-bearing gases equally or selectively.^{194,195} Simply building a connection between the initial feedstocks and the CNT products is not sufficient. The thermally-driven chemistry that takes place between the input and the output also needs to be accounted for. It is therefore worth examining what carbon-bearing gases get consumed at the catalyst substrates, and are thereby converted to CNTs. Further understanding of the detail reaction schemes will help engineers control the quality of produced CNTs that will satisfy the commercialization objectives.

There are solutions to overcome such challenges, *albeit* empirically, by adapting technology advancements from plastic waste recycling processes, as well as from CNT synthesis processes. For example, intelligent waste identification/separations can be assisted by (tribo)electrostatics, laser-induced plasma spectroscopy,^{220,221} laser-induced breakdown spectroscopy,^{222–224} density media,²²⁵ artificial neural networks,²²⁶ etc. Plastic waste decompositions, on the other hand, can be benefited from the well-established pyrolysis/gasification fundamentals,^{227–233} reaction chemistry (thermodynamics,²³⁴ kinetics,^{235–238} mechanisms,²³⁷ etc.), as well as the related technology developments, such as those of catalysts,^{239–241} additives,^{242,243} process,²⁴⁴ or their combinations. Last but not least, CNTs synthesis, especially their yields with controlled quality, could also gain from recent progresses in process engineering at nanoscale.^{21,111,245–249}

CONCLUSIONS

In summary, upcycling plastics waste into high value CNTs is indeed a sustainable solution with a promising future as it allows for conversion of post-consumer products to value-added products and can help alleviate the burden of solid wastes on the environment. Its realization from concept is highly multidisciplinary. Partnerships among fundamental science, engineering development, etc. are essential for integrating such process with existing manufacturing methods.⁴⁷ Knowledge and experience from industry on existing facilities, such as those for recycling, gasification/pyrolysis, as well as those of fluid catalytic cracking and hydrocracking plants, can be instrumental.

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