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Review

A review of the electrical and mechanical properties of carbon nanofiller-reinforced polymer composites

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ABSTRACT

Within decades of development, carbon nanomaterials such as carbon black, fullerene, carbon nanotube, carbon nanofiber, graphene and their combined nanofillers have been tremendously applied in polymer material industries, generating a series of fascinating multifunctional composites in the fields from portable electronic devices, sports, entertainments to automobile, aerospace and military. Among the various material properties of the composites, electrical conductivity and mechanical performance are the two most important parameters for evaluating the effectiveness of nanofillers in the polymer matrices. In this review, we focus on the electrical and mechanical properties of diverse dimensional carbon nanofillers (e.g., zero-, one-, two-, three-dimensional nanofillers or their combinations)-reinforced polymer composites to seek the most efficient and effective approach to obtain high-performance polymeric nanocomposites.

Abbreviations

3D	Three-dimensional
APTES	Aminopropyltriethoxysilane
CB	Carbon black
CNF	Carbon nanofiber
CNT	Carbon nanotube
CTAVB	Cetyltrimethylammounium 4-vinylbenzoate

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CVD	Chemical vapor deposition
DBSA	Dodecylbenzenesulfonic acid
DMF	Dimethylformamide
EBA	Ethylene-butyl acrylate
EDA	Ethylenediamine
EMI	Electromagnetic interference shielding
ESO	Epoxidized soybean oil
GNP	Graphite nanoplatelet
GO	Graphene oxide
HDPE	High-density polyethylene
LLDPE	Linear low-density polyethylene
MWCNT	Multi-walled carbon nanotube
NBR	Acrylonitrile butadiene
NP	Nanoparticle
ODA	Octadecylamine
PA-6	Polyamide-6
PANI	Polyaniline
PC	Polycarbonate
PDLA	Poly(D-lactide)
PDMS	Poly(dimethylsiloxane)
PE	Polyethylene
PEDOT	Poly(3,4-ethylenedioxythiophene)
PEG	Polyethylene glycol
PEI	Polyetherimide
PES	Poly(ether sulfone)
PGMA	Poly(glycidyl methacrylate)
PI	Polyimide
PLA	Polylactic acid
PLLA	Poly(L-lactide)
PMMA	Polymethylmethacrylate
POM	Polyoxymethylene
PP	Polypropylene
PPD	*p*-Phenylenediamine
PS	Polystyrene
PSS	Polystyrene sulfate
PPE	Polyphenylene ethynylene
PVDF	Polyvinylidenefluoride
PVA	Poly(vinylacetate)
PVP	Polyvinylpyrrolidone
SAN	Styrene-acrylonitrile
SDS	Sodium dodecyl sulfate
SDBS	Sodium dodecylbenzene sulfonate
RGO	Reduced graphene oxide
SWCNT	Single-walled carbon nanotube
THF	Tetrahydrofuran
TPU	Thermoplastic polyurethane
UHMWPE	Ultra high molecular weight polyethylene
UV/O_3	Ultraviolet/ozone

Introduction

The discovery of various carbonaceous nanofillers, such as carbon black (CB), fullerene, carbon nanotubes (CNTs), carbon nanofibers (CNFs) and graphene largely extends the functionalities of polymers from traditional features: lightweight, cost-effective, easily formed, to new ones, including electrically and thermally conductive, electromagnetic shielded, wave absorbed and shape memorized, etc. [1–3]. Therefore, a series of multifunctional polymeric nanocomposites are reported. Recently, with the ever-increasing utilities of multifunctional polymer composites applicable in the electronics, sensors, energy, automobile and aerospace industries, the electrical and mechanical properties are among the two most important parameters to determine the performance of the polymeric nanocomposites.

Achieving polymer nanocomposites with high electrical and mechanical properties requires meticulous design of three segments: construction of strong and conductive nanofiller networks, appropriate polymer matrix, and good interfaces between the two components, as illustrated in Fig. 1. In specific terms, building efficient interconnected networks is closely related to the selection of carbon nanofiller type(s), which directly controls the geometric shape of the network structure. In addition, nanofiller content determines the amount of "bricks" for the network building and there is always a minimum content (percolation threshold content) required to ensure the build-up of the structure, from which the polymer instantly transforms from insulated to conductive [4]. Dispersion and distribution are two essential issues for the establishment of the conductive networks [5]. Good dispersion and homogeneous distribution of carbon nanofillers are definitely beneficial for constructing interconnected networks. However, certain extent of agglomeration and uniquely designed nonhomogeneous distribution can also realize interesting segregated structures with excellent electrical and mechanical properties [6], depending on the type of the polymer matrix. For example, good processability of thermoset polymers enables relatively easy effort to achieve good dispersion of nanofillers in the polymer matrix via solution mixing, ultrasonication, spin coating, etc., while for high viscosity thermoplastic polymers, those aforementioned methods are not effective for improving the dispersion of nanofibers and alternative approaches, such as melt mixing, milling, latex technology, double percolation method, were adopted, resulting in very different dispersion and distribution characteristics. The interface between nanofiller and polymer matrix is also a critical factor in influencing the electrical and mechanical properties of the bulk nanocomposite [7]. Surface modifications can be divided into covalent and non-covalent techniques, which are widely used to improve interfacial interactions by means of chemical bonding or surfactant coating approaches. Covalent techniques can generate strong C–C bond between nanofillers and molecular chains of the polymer matrix to provide superb mechanical properties. However, the electrical property maybe otherwise damaged due to the destruction of the harsh treatment of carbon nanofiller surfaces [5]. Non-covalent technique, on the other hand, can preserve the integrity and pristine electrical property of the nanofillers, while the mechanical properties may vary regarding the efficiency and compatibility of surfactants, types of polymer matrix and fabrication method. In general, comprehensive consideration of the types and contents of nanofillers, the polymer matrix, fabrication method affecting dispersion quality and interfacial behavior should be taken into account for the design of polymer nanocomposite with both high electrical and mechanical performance.

Therefore, in this paper, the electrical and mechanical properties of diverse dimensional carbon nanofiller-reinforced polymer nanocomposites are reviewed and summarized in Tables 1, 2, 3, 4 and 5. Here, we arrange the main body of the paper according to the different dimensional structures of the nanofillers (including 3D foam and hybrid structures) aims to focus on the construction of highly conductive composite ($\geq 10^{-5}$ S/m) [6] that has potential applications in electric power, energy batteries and aircraft structure fields, etc. [8–10]. Other issues, such as polymer types, fabrication methods, and surface treatment methods are also discussed within this framework. We hope this review paper can provide readers with a broad and systematic landscape and guidance to the design of polymeric nanocomposites with high electrical conductivities and mechanical performance.

Zero-dimensional carbon nanofiller-reinforced polymer composites

Carbon black (CB)-reinforced polymer composites

Carbon black (CB) is one of the most frequently applied zero-dimensional carbon nanofillers in the conductive polymeric composites because of their abundancy, superior electrical conductivity and chemical stability. However, CBs are often agglomerated into large aggregates due to the strong Van der Waals force, leading to high-percolation threshold content (15–20 wt%) for achieving conductive networks [11, 12]. In the meanwhile, the large clusters of aggregates often result in bad interfacial interaction and defects to the polymer matrix and decrease the mechanical properties of the bulk nanocomposites. Traditional methods to improving dispersion of CBs involved using chemical or physical modifications, for instance, covalently graft chemical groups [13] or non-covalent surfactants [172] on the CB surface. However, these treatments showed no significant decrease in percolation threshold and even caused adverse effects on the electrical properties of the resulting nanocomposite materials.

Recently, a so called double percolation method was used to solve the high percolation threshold problem for CB-reinforced nanocomposites. The critical technique is to selectively localize CB nanoparticles in a polymer blend made of two immiscible polymer matrices. In specific, CB is firstly dispersed and percolated in one polymer phase of the blend, and this filler-rich phase is then dispersed in

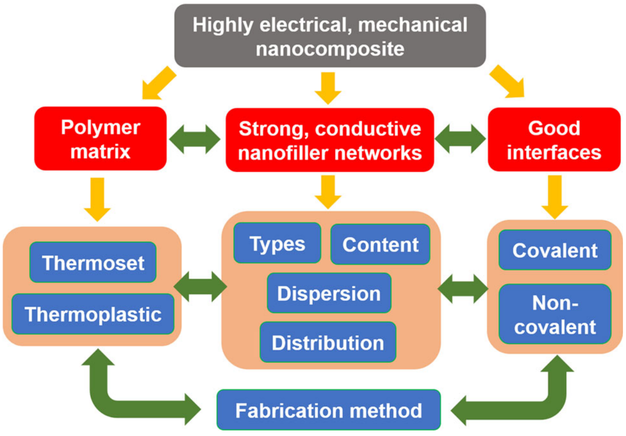

Figure 1 General considerations for the fabrication of high electrical and mechanical performed nanocomposite.

Table 1 Summary of the electrical and mechanical properties of CB-reinforced polymer composites

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
CB	Polyester	Mechanical mixing	10 wt%	20 wt%	0.1	50 wt%	23 (− 49%)	1.75 (+ 80%)	[11]
CB	PP	Mechanical mixing + Injection molding	7 wt%	15 wt%	1	–	–	–	[12]
CB (Diaryl)	PP	Solvent mixing + Hot pressed	10 wt%	20 wt%	25	–	–	–	[13]
CB	PP	Melt mixing	2.75 vol%	6.5 vol%	2	4.0 vol%	22 (− 54%)	0.17 (− 46%)	[14]
CB	PP/PS	Melt mixing + PS remove + Compress	0.37 vol%	9.0 wt%	20	4.0 vol%	26 (− 45%)	0.25 (− 21%)	[14]
CB	TPU	Melt mixing	6.93 vol%	18 vol%	1.5	–	–	–	[15]
CB	TPU/COPA	Melt mixing	3.68 vol%	10 vol%	1	–	–	–	[15]
CB	PLA/TPU	Melt mixing	3phr	10phr	10	3phr	30 (+ 36%)	0.98 (+ 4%)	[16]
CB	PS/PP (SEBS)	Melt mixing	1.5 wt%	3.5 wt%	0.1	1.5 wt%	32 (+ 18%)	–	[17]
CB	POM/SAN/TPU	Melt mixing	3.8 wt%	7.5 wt%	0.1				[18]
CB	POM/SAN/PA6	Melt mixing	4.28 wt%	9.0 wt%	0.5				[18]
CB	PMMA/Epoxy	Mechanical mixing	–	15 vol% (10 vol% PMMA)	0.05	15 vol% (10 vol% PMMA)	35 (− 39%)	1.76 (− 14%)	[19]
CB	PLLA	Melt mixing + Compress	2.7 wt%	5.0 wt%	1	5.0 wt%	67 (+ 3%)	3.9 (+ 11%)	[20]
CB	PLLA/PDLA	Melt mixing + Compress	2.23 wt% (1 wt% PDLA)	5.0 wt%	1	5.0 wt% (10 wt% PDLA)	75 (+ 15%)	3.95 (+ 12%)	[20]
Fullerene soot	PA-6	Solution mixing	–	0.1 wt%	10^{-5}	0.05 wt%	103 (+ 10%)	8.35 (+ 11%)	[21]

Table 2 Summary of electrical and mechanical properties of CNTs-reinforced polymeric nanocomposites

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
MWCNT	Epoxy	Mechanical stirring	0.0025 wt%	1 wt%	2	–	–	–	[22]
MWCNT	Epoxy	Mechanical stirring	0.011 wt%	1 wt%	0.4	–	–	–	[23]
MWCNT	Epoxy	Mechanical stirring	0.5 wt%	3 wt%	1	4 wt%	33 (+ 6%)	2.6 (+ 100%)	[24]
MWCNT	Epoxy	Mechanical stirring	0.0025 wt%	0.5 wt%	0.4	–	–	–	[25]
SWCNT	Epoxy	Mechanical stirring + Sonicated	0.05 wt%	0.5 wt%	0.03	–	–	–	[25]
SWCNT (Ball milled)	Epoxy	Ball milled + Mechanical stirring	0.23 wt%	0.5 wt%	0.001	–	–	–	[25]
MWCNT	Epoxy	Sonicated + Mechanical stirring	0.03 wt%	0.15 wt%	0.5	–	–	–	[26]
MWCNT	Epoxy	Sonicated + Two-roll milling	0.04 wt%	0.5 wt%	0.5	–	–	–	[27]
SWCNT (Acid)	Epoxy	Sonicated + High shear mixing	0.04 wt%	4 wt%	10	–	–	–	[28]
MWCNT (UV/O_3)	Epoxy	Sonicated	0.1 wt%	1 wt%	0.2	–	–	–	[29]
MWCNT	Epoxy	Magnetic stirring + Hot pressing	0.7 wt%	4 wt%	5	4 wt%	10 (+ 150%)	0.46 (+ 320%)	[30]
MWCNT	Epoxy	Sonicated + Stirring	0.5 wt%	1.5 wt%	0.5	1.5 wt%	80 (+ 17%)	3.8 (+ 35%)	[31]
MWCNT (Silane)	Epoxy	Sonicated	0.05 wt%	0.5 wt%	0.06	0.25 wt%	125^F (+ 18%)	3.5^F (+ 20%)	[32]
MWCNT (Plasma)	Epoxy	Shear mixing + Sonicated	0.1 wt%	1 wt%	0.02	1.0 wt%	68 (+ 51%)	1.3 (+ 116%)	[33]
MWCNT	Epoxy	Three-roll milling	0.25 wt%	0.75 wt%	0.006	–	–	–	[34]
MWCNT (NH2)	Epoxy	Sonicated + Hot pressing	0.1 wt%	0.5 wt%	0.001	0.25 wt%	120^F (+ 12%)	3.3^F (+ 14%)	[35]
MWCNT	Epoxy	Sonicated + Hot pressing	0.1 wt%	0.5 wt%	0.01	0.5 wt%	112^F (+ 5%)	3.2^F (+ 10%)	[35]
MWCNT	Epoxy	Sonicated + Spin coating	0.8 vol%	1 vol%	0.0007	1 vol%	50 (− 9%)	2.5 (− 7%)	[36]
MWCNT (UV/O_3)	Epoxy	Sonicated + Three-roll milling + DC electric field alignment	0.0048 wt%	0.5 wt%	0.5 (Parallel)	0.3 wt%	–	4.2 (+ 45%)	[37]
MWCNT	Epoxy	CVD + Rolling + Hot press infiltration	–	53 wt%	3000	56 wt%	–	15.5	[38]
MWCNT	Epoxy	Mechanical stirring	0.2 wt%	1 wt%	10^{-5}	1 wt%	72 (+ 6%)	3.7 (+ 17%)	[39]
MWCNT	Epoxy	Mechanical mixing + Ice templating	–	1.5 wt%	15.8 (Parallel)	1.5 wt%	150^C (+ 12%)	1.9^C (+ 25%)	[40]
MWCNT	Epoxy	Mechanical mixing + Ultrasonication	0.5 wt%	2 wt%	0.01	0.1 wt%	72 (+ 20%)	2.7 (+ 8%)	[41]
MWCNT	Epoxy	Mechanical mixing + Hot pressing	–	1 wt%	10^{-5}	0.4 wt%	42 (− 2%)	2 (− 2%)	[42]
MWCNT (Acid)	Epoxy	Ultrasonication	0.4 wt%	1 wt%	0.003	1 wt%	125 (+ 160%)	–	[43]
MWCNT	Epoxy	Mechanical mixing + CO_2 assisted dispersion	0.1phr	0.2phr	0.8	0.3phr	49 (+ 63%)	0.37 (− 2%)	[44]
MWCNT	Epoxy	Shear mixing + Ultrasonication	0.2 wt%	0.8 wt%	0.03	0.6 wt%	79 (+ 14%)	3.08 (+ 12%)	[45]

Table 2 continued

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
MWCNT (Pyrogallol)	Epoxy	Mechanical mixing	0.2 wt%	1.5 wt%	0.6	1.5 wt%	76 (+ 24%)	–	[46]
MWCNT (Polyaniline)	Epoxy	Mechanical mixing + Casting	0.6 wt%	1.5 wt%	0.001	1 wt%	57 (+ 61%)	0.73 (+ 43%)	[47]
MWCNT (Gelatin)	Epoxy	Mechanical mixing + Sonication	0.05 wt%	0.5 wt%	10^{-4}	0.5 wt%	98 (+ 16%)	2.91 (+ 18%)	[48]
MWCNT (Grinding)	TPU	Mechanical STIRRING	1 wt%	15 wt%	2000	15 wt%	25 (− 8%)	0.057 (+ 330%)	[49]
MWCNT (Acid)	TPU	Sonicated + Stirred + Roll milled	0.009 vol%	4 vol%	8	–	–	–	[50]
MWCNT (Acid)	TPU	Sonicated + Stirred + Roll milled	0.05 vol%	2 vol%	3	–	–	–	[51]
MWCNT	TPU	Sonicated + Stirred	0.1 wt%	1.5 wt%	0.08	1.5 wt%	6.93 (+ 5%)	0.015 (+ 50%)	[52]
MWCNT (Acid)	TPU	Sonicated + Stirred	0.1 wt%	1.5 wt%	0.1	1.5 wt%	8.75 (+ 32%)	0.024 (+ 140%)	[52]
MWCNT (Acid)	TPU	Sonicated + Stirred	–	1.5 wt%	0.0016	1.5 wt%	63 (+ 40%)	0.095 (+ 280%)	[53]
MWCNT ($Fe(acac)_3$)	TPU	Sonicated + Stirred	0.02 wt%	3 wt%	0.01	1.5 wt%	14 (+ 100%)	–	[54]
MWCNT (Methanol)	TPU (Acetone)	Sonicated	–	35 wt%	2300	35 wt%	41 (+ 20%)	1.27 (+ 950%)	[55]
MWCNT (Amide)	WBPU	Sonicated + Stirred	0.8 wt%	1 wt%	0.005	0.5 wt%	12 (+ 20%)	0.007 (+ 10%)	[56]
MWCNT	TPU	Melting blending + Annealing	5 vol% (4 h anneal)	15 vol%	5	10 vol% (10 min anneal)	15.2 (− 35%)	0.01 (+ 14%)	[57]
MWCNT (SDS)	TPU	Sonicated + Spray dried	5 vol%	8 vol%	0.01	8 vol%	26.7 (− 1%)	–	[58]
MWCNT (Silane)	TPU	Sonicated + Stirred	0.5 wt%	1 wt%	10^{-5}	0.5 wt%	15.5 (+ 25%)	–	[59]
MWCNT	PA-6	Sonicated + Melt mixing	2.5 wt%	4 wt%	0.03	–	–	–	[60]
MWCNT (Na-AHA)	PA-6	Sonicated + Melt mixing	0.25 wt%	3 wt%	0.01	–	–	–	[60]
MWCNT	PA-6	Extruded	7 wt%	16 wt%	10	12 wt%	–	4.1 (+ 61%)	[61]
MWCNT (Amide)	PA-6	Sonicated + Melt mixing	1phr	5phr	0.3	5phr	112 (+ 27%)	3.76 (+ 25%)	[62]
MWCNT (P3HT-g-PCL)	PC	Sonicated	0.5 wt%	5 wt%	64	0.5 wt%	61 (+ 50%)	2.1 (+ 50%)	[63]
MWCNT	PC	Extruded	1.5 wt%	15 wt%	10	–	–	–	[64]
SWCNT (PPE)	PC	Shaken + Sonicated	0.1 wt%	7 wt%	500				[65]
SWCNT	PC	Extruded	1.8 wt%	10 wt%	0.1	5 wt%	28 (− 41.6%)	1.22 (− 9.6%)	[66]
SWCNT	PC	Sonicated + Hot pressed	0.5 wt%	7 wt%	10	5 wt%	50 (+ 11.1%)	1.3 (+ 8.3%)	[66]
MWCNT	PC	Sonicated + Stirring + Hot pressed	0.02 wt%	0.1 wt%	0.45	–	–	–	[67]

Table 2 continued

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
MWCNT	PC	Extruded + Hot pressed	0.28 wt%	3 wt%	20	1 wt%	72 (+ 7%)	1.5 (+ 7%)	[68]
MWCNT (PC)	PC	Extruded	0.7 wt%	2.5 wt%	0.001	2 wt%	61 (+ 26%)	1.45 (+ 24%)	[69]
MWCNT	PC	Extruded	2 wt%	10 wt%	1	2 wt%	79 (+ 19%)	1.56 (+ 13%)	[70]
MWCNT	PC	Extruded	0.2 wt%	3 wt%	100	1 wt%	60 (– 3%)	1.6 (+ 14%)	[71]
MWCNT (Acid)	PE (UHMW)	Mechanical stirring + Hot pressed	0.07 wt%	0.7 wt%	0.01	–	–	–	[65]
MWCNT (Acid)	PE (UHMW)	Mechanical stirring	0.14 wt%	0.7 wt%	0.1	–	–	–	[65]
MWCNT	PE (LLD)	Ball milling + Hot pressed	2 wt%	10 wt%	3	3 wt%	18 (+ 140%)	0.6 (+ 33%)	[72]
MWCNT	PE (UHMW)	Dry mixed + Hot pressed	0.28 wt%	1 wt%	10	3.5 wt%	30 (– 33%)	0.9 (+ 180%)	[73]
MWCNT (DMF)	PE (UHMW)	Sonicated + Dry mixed + Hot pressed	0.19 wt%	1 wt%	100	3.5 wt%	36 (– 33%)	1.1 (+ 190%)	[73]
MWCNT (SDS)	PE (UHMW)	Sonicated + Dry mixed + Hot pressed	0.045 wt%	1 wt%	50	3.5 wt%	38 (– 30%)	1.2 (+ 166%)	[73]
SWCNT	PE (UHMW)	Dry mixed + Hot pressed	1.2 wt%	3 wt%	0.005	3 wt%	30 (– 33%)	0.6 (+ 66%)	[73]
SWCNT (DMF)	PE (UHMW)	Sonicated + Dry mixed + Hot pressed	0.09 wt%	2 wt%	0.05	2.5 wt%	35 (– 36%)	0.78 (+ 85%)	[73]
SWCNT	PE (UHMW)	Sonicated + Dry mixed + Hot pressed	0.25 wt%	3 wt%	0.5	2.5 wt%	33 (– 38%)	0.72 (+ 80%)	[73]
MWCNT	PE (UHMW)	Sonicated + Stirred + Hot pressed	0.072 vol%	0.8 vol%	5	–	–	–	[74]
MWCNT	PE (UHMW)	Dry mixing + Hot pressed	0.1 wt%	10 wt%	88	–	–	–	[75]
MWCNT	PE (UHMW)	Solution mixing + Sintering	0.04 wt%	10 wt%	0.5	10 wt%	22 (+ 37%)	0.25 (+ 20%)	[76]
MWCNT	PE (UHMW)	Solution mixing + Hot pressed	0.1 wt%	1 wt%	0.3	2 wt%	32	0.7	[77]
MWCNT	PE (UHMW/ HD)	Solution mixing + Mechanical stirred + Hot pressed	0.1 wt%	1 wt%	0.09	3 wt%	30	0.95	[77]
MWCNT	PE (HD)	Extruded + Compression molded	2 wt%	8 wt%	10	8 wt%	18 (– 34%)	2.3 (+ 163%)	[78]
MWCNT	PP	Sonicated + Extruded	0.07 wt%	0.4 wt%	0.1	–	–	–	[79]
MWCNT	PP	Extruded + Hot pressed	0.44 wt%	9 wt%	2	–	–	–	[80]
MWCNT (Acid)	PP	Extruded + Hot pressed	1.5 wt%	5 wt%	0.2	–	–	–	[81]
MWCNT	PP	Sonicated + Freeze dried + Hot pressed	0.3 wt%	1 wt%	1.5	–	–	–	[82]
MWCNT	PP	Melt mixing + Extruded	0.45 wt%	15 wt%	0.99	15 wt%	47 (+ 38%)	0.37 (+ 54%)	[83]
MWCNT	PP	Melt mixing + Extruded	1.07 wt%	15 wt%	0.23	15 wt%	38 (+ 11%)	0.39 (+ 62%)	[83]
MWCNT	PP	Ultrasonicated + Hot pressed	0.013 vol%	5 wt%	117	1 wt%	25	2.0	[84]
MWCNT	PP	Injection molded	3.8 wt%	12 wt%	20	8 vol%	–	1.4 (+ 100%)	[85]

Table 2 continued

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
MWCNT	PP	Grinding + Injection molded	0.9 wt%	5 wt%	0.01	5 wt%	36	1.8	[86]
SWCNT (CTAB)	PS	Solution mixing	0.045 wt%	1 wt%	0.1	1 wt%	48^F (− 36%)	2.8^F (+ 5%)	[87]
SWCNT (Acid)	PS	Ultrasonicated	0.8 wt%	5 wt%	0.001	1 wt%	12.7 (+ 12%)	0.01 (− 11%)	[88]
SWCNT (PPE)	PS	Shaken + Sonicated	0.05 wt%	7 wt%	7	–	–	–	[89]
MWCNT (DMF, THF)	PS	Sonicated + Stirred + Hot pressed	0.018 vol%	8 vol%	1	–	–	–	[90]
SWCNT (PS)	PS	Sonicated	0.75 wt%	3 wt%	0.1	–	–	–	[91]
MWCNT (SDS)	PS (Latex)	Solution mixing	1.1 wt%	3.0 wt%	100	–	–	–	[92]
SWCNT (SDS)	PS (Latex)	Solution mixing	0.6 wt%	2.0 wt%	50	–	–	–	[92]
SWCNT (SDS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	0.28 wt%	1.5 wt%	1	–	–	–	[93]
MWCNT (SDBS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	0.9 wt%	6 wt%	0.1	–	–	–	[94]
SWCNT (SDS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	0.25 wt%	1.5 wt%	1	–	–	–	[95]
MWCNT (SDS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	1.5 wt%	5.5 wt%	1	–	–	–	[96]
SWCNT (SDS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	0.3 wt%	1.5 wt%	1	–	–	–	[97]
MWCNT (PGMA)	PMMA	CVD + Mechanical stirring	–	8 wt%	0.1	2 wt%	53 (+ 9%)	1.3 (+ 43%)	[98]
MWCNT (Acid)	PMMA	Solution mixing	0.25 wt%	5 wt%	2000	5 wt%	30 (+ 200%)	1.3 (+ 188%)	[99]
MWCNT (DMF, MeOH)	PMMA	Solution mixing	0.01 wt%	3 wt%	123	–	–	–	[100]
SWCNT	PMMA	Mechanical stirring	0.17 wt%	10 wt%	1700	1 wt%	16 (− 27%)	0.18 (− 64%)	[101]
SWCNT (SOCl2)	PMMA	Mechanical stirring	0.17 wt%	13.5 wt%	10000	1 wt%	33 (+ 50%)	0.65 (+ 30%)	[101]
MWCNT	PMMA	Sonicated + Mechanical stirring	0.4 wt%	10 wt%	100	–	–	–	[102]
SWCNT ($SOCl_2$)	PMMA	Shaken + Ultrasonicated	0.1 wt%	0.5 wt%	47	–	–	–	[103]
MWCNT	PMMA (Latex)	Solution mixing	0.8 wt%	1.2 wt%	10	–	–	–	[92]
SWCNT	PMMA (Latex)	Solution mixing	0.6 wt%	1.6 wt%	100	–	–	–	[92]
SWCNT	PMMA (Latex)	Sonicated + Freeze dried + Hot pressed	0.8 wt%	1.6 wt%	0.1	–	–	–	[93]
Ag-MWCNT	PP	Melt mixing + Molding	0.38 vol%	10 wt%	0.5	5 wt%	39 (+ 8%)	0.9 (+ 28%)	[104]
Ag-MWCNT	PS	Ultrasonic + Hot pressed	0.35 vol%	10 wt%	65	10 wt%	44 (+ 69%)	2.0 (+ 66%)	[104]
Ag-MWCNT	Epoxy	Sonicated + Hot pressed	0.1 wt%	0.5 wt%	81	0.1 wt%	118^F (+ 10%)	3.17^F (+ 9%)	[35]
Ag/MWCNT	PU	Sonicated + Stirred + Roll milled	–	10 vol%/ 2 vol%	25600	–	–	–	[50]

Table 2 continued

Filler (treatment)	Matrix	Fabrication method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
MWCNT	Ag-PANI	Mechanical stirred	–	8 wt%/ 6 wt%	424	–	–	–	[105]
Ag-MWCNT	PVDF	Sonicated + Stirred	–	8.6 wt%/ 1.8 wt%	200000	–	–	–	[106]
Ag-MWCNT	PVDF	Sonicated + Stirred + Hot rolling	–	8.6 wt%/ 1.8 wt%	571000	–	–	–	[106]
Ag-MWCNT	PANI	Sonicated	–	–	40	–	–	–	[107]
Ag-MWCNT	PANI	Stirred + Sonicated	–	–	1574				[108]
Au-SWCNT	PEDOT-PSS/ PVAc	Sonicated	–	15 vol%/ 60 vol%	600000	–	–	–	[109]
TiO_2-MWCNT	Epoxy	Sonicated	0.2 wt%	1 wt%	10^{-5}	0.5 wt%	37 (+ 117%)	2.1 (+ 50%)	[110]
Cu-MWCNT	PLA/ESO	Mechanical stirred + Sonicated	1 wt%	2 wt%	0.5	2 wt%	0.97 (+ 54%)	0.8 (+ 33%)	[111]
ZnO-MWCNT	Epoxy	Sonicated	–	1.7 wt%	100	1.7 wt%	61 (+ 20%)	3.6 (+ 51%)	[112]
CNF (2500 °C heat)	Epoxy	Mechanical mixing	0.32 wt%	1.3 wt%	1.37	1.3 wt%	–	3.1 (+ 6%)	[113]
CNF	PP	Extrusion + Compressed	0.75 vol%	2.5 vol%	10	0.9 vol%	– (+ 30%)	–	[114]
CNF	Epoxy	Mechanical stirred + Sonicated	–	0.75 wt%	0.27	0.75 wt%	2.4 (+ 41%)	60 (+ 57%)	[115]
CNF	PMMA	Solution mixing	5 wt%	10 wt%	0.5	5 wt%	45 (+ 18%)	0.85 (+ 13%)	[116]
CNF (Soy protein)	Epoxy	Mechanical mixing + Sonication	0.5 wt%	2.0 wt%	10^{-4}	–	–	–	[117]
CNF (Soy protein)	PC	Mechanical mixing + Sonication	0.5 wt%	2.0 wt%	0.001	–	–	–	[117]
CNF	PMMA	Mechanical mixing	2 wt%	10 wt%	1	0.5 wt%	37 (− 27%)	1.3 (+ 4%)	[118]
CNF	PDMS	Solution mixing	1 vol%	8 vol%	1	–	–	–	[119]
CNF (NH_2)	PDMS	Solution mixing	–	40 wt%	31	30 wt%	3.7 (+ 500%)	0.06 (+ 110%)	[120]
CNF	PE	Solution mixing + Melt mixing	3 wt%	10 wt%	0.001	1 wt%	0.004 (+ 100%)	17 (+ 124%)	[121]

F: Flexural properties; C: Compression properties; Others are tensile properties

Table 3 Summary of the electrical and mechanical properties of graphene-reinforced polymer composites

Filler (treatment)	Matrix	Method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
GNP (UV/O$_3$)	Epoxy	Shear mixing + Ultrasonicated	0.5 wt%	4 wt%	200	2 wt%	80^F (− 27%)	3.4^F (+ 13%)	[122]
Graphene (PVP)	Epoxy	Solution mixing	0.08 vol%	0.46 vol%	1e−5	0.13 vol%	50 (+ 11%)	1.5 (+ 16%)	[123]
Graphene	Epoxy	Sonicated + Spin coating	0.1 vol%	0.6 vol%	0.006	1 vol%	20 (− 63%)	1.8 (− 33%)	[124]
GNP	Epoxy	Three-roll milling	0.3 wt%	2 wt%	0.002	0.5 wt%	–	3.18 (+ 15.6%)	[125]
GNP	Epoxy	Sonicated + High speed shear mixing	0.3 wt%	1 wt%	10^{-6}	–	–	–	[125]
Expanded GNP	Epoxy	Sonicated + Stirred	3 wt%	8 wt%	0.08	–	–	–	[126]
Graphene	Epoxy	Sonicated + Stirred	8 wt%	15 wt%	0.8	–	–	–	[127]
Graphite	Epoxy	Three-roll milling + Mechanical stirring	0.52 vol%	3 wt%	0.01	4 wt%	83 (+ 27%)	5.2 (+ 160%)	[128]
Graphene	PC (chloroform)	Solution mixing	0.38 wt%	4 wt%	22.6	0.55 vol%	–	1.2 (+ 20%)	[129]
Graphene (Triton)	PC (PVP)	Emulsion mixing	0.14 wt%	4 wt%	51.2	2.2 vol%	–	1.2 (+ 20%)	[129]
R-GO	PMMA (Latex)	Stirring + Crushed + Hot pressed	0.16 vol%	2.7 vol%	64	4 wt%	–	4.7 (+ 30%)	[130]
Graphene	PP (Latex)	Sonicated + Freeze dried + Hot pressed	1.2 wt%	3.5 wt%	0.9	–	–	–	[82]
Graphite	PP (Latex)	Sonicated + Freeze dried + Hot pressed	7 wt%	10 wt%	0.1	–	–	–	[82]
R-GO	PP (Latex)	Sonicated + Hot pressed	0.033 vol%	0.2 vol%	0.002	–	–	–	[131]
GO (Isocyanate)	PS	Solution mixing	0.1 vol%	2.5 vol%	1	–	–	–	[132]
R-GO (PSS)	PS (Latex)	Sonicated + Freeze dried + Hot pressed	0.6 wt%	1.6 wt%	15	–	–	–	[133]
R-GO	PS (Latex)	Water based blending + Hot pressed	0.08 vol%	4 vol%	20.5	–	–	–	[134]
R-GO	PS (Latex)	Mixing + Filtering + Hot pressed	0.15 vol%	4.8 vol%	1083.3	4.8 vol%	–	2.0 (+ 100%)	[135]
GNP	PP	Melt mixing + Injection molding	3 vol%	9.3 vol%	1.7	9.3 vol%	34.3 (− 1%)	0.6 (+ 28%)	[136]
GO (Alkylamine)	PP	Sonicated + Melt blending	0.63 wt%	1 wt%	0.03	1.0 wt%	49 (+ 29%)	1.6 (+ 47%)	[137]
R-GO	PP	Sonicated + Sheet casting	0.1 wt%	2 wt%	0.17	2.0 wt%	28 (+ 10%)	1.0 (+ 40%)	[138]
GO (ODA)	PI	Mechanical stirred + Thermal imidization	0.5 wt%	3 wt%	0.038	3 wt%	105 (+ 40%)	3.0 (+ 95%)	[139]

Table 3 continued

Filler (treatment)	Matrix	Method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
GO (APTES)	PI	Mechanical stirred + Thermal imidization	0.3 wt%	1.5 wt%	0.003	1.5 wt%	143 (+ 79%)	3.29 (+ 132%)	[140]
Graphite (APTES)	PA-6	Mechanical stirred + Hot compression	0.032 vol%	2.45 vol%	60	1.45 vol%	55 (− 6%)	0.97 (+ 102%)	[141]
Graphene (Dioxolane)	PC	Mechanical stirred + Hot compression	0.55 vol%	10 wt%	0.001	1 wt%	60 (+ 11%)	1.45 (+ 26%)	[142]
Expanded graphite	PE	Mechanical stirred	8.4 wt%	20 wt%	0.013	5.4 wt%	24 (− 4%)	0.58 (+ 11%)	[143]

F: Flexural properties; Others are tensile properties

another polymer matrix, constructing a co-continuous double percolated structure, as shown in Fig. 2. It should be noted that the weight ratio between the two polymer phases is an important value as it changes the morphology of the blend to different features. Polymers, such as polystyrene (PS) [14, 17], thermoplastic polyurethane (TPU) [15, 18] and polyamide-6 (PA6) [18, 173], have been selected as the individual phase due to their good interfacial interactions with CB and this affinity can be determined by the surface tension of every single constituent in the blend [14, 17, 18, 173]. Gong et al. [14] introduced PS in the CB/polypropylene (PP) composite and successfully reduced the percolation threshold from 2.75 to 0.37 vol%. The results showed that the CB were selectively dispersed at the interfaces between PP and PS, which led to decrease in the percolation and increase in the conductivity and mechanical properties. It is noted that the polymer blend was not always restricted to the two phases. Some researchers introduced a third component in polymer blends to perform either as surfactant to tailor the location of the CB in the matrix or as compatibilizer to increase the mechanical properties of the bulk polymer composites [17, 18]. Apart from double percolation, constructing segregated structures in a polymer matrix is another way to fabricating conductive composites with low percolation threshold. Phua et al. [19] added polymethylmethacrylate (PMMA) powder in the epoxy matrix to separate CB nanoparticles so as to promote conductive networks for achieving good electrical and mechanical properties. Liu et al. [20] proposed a similar but cleverer approach to adding poly(D-lactide) (PDLA) in the poly(L-lactide) (PLLA) matrix for spontaneously producing stereo-complex crystallites through co-crystallization in the two components in order to construct segregated conductive structures (Fig. 3). Results showed that the crystallites can not only reduce the percolation threshold value of the composite but also perform as toughening agents to strengthen the mechanical properties.

Fullerene-reinforced polymer composites

Fullerene is another type of zero-dimensional carbon nanofiller with an interesting soccer ball shaped close-cage molecular structure. Among its various allotropic family members, the most well-known fullerene material is C_{60}, which is consisted of 12

Table 4 Summary of the electrical and mechanical properties of 3D nano-foam-reinforced polymer composites

Filler (treatment)	Matrix	Method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
R-GO (Reduced)	Epoxy	Solution mixing with NH_3 + Lyophilized	0.91 vol%	6.5 vol%	1	–	–	–	[144]
R-GO (Reduced)	Epoxy	Vacuum-assisted impregnation	0.21 vol%	0.95 vol%	1	0.95 vol%	0.75^C (+ 172%)	–	[144]
R-Graphite	PDMS	One-dimensional freezing + Thermal reduction	–	0.4 wt%	3.2	–	–	–	[136]
MWCNT	PLLA	Mechanical mixing + PIF processing + CO_2 foaming	0.16 vol%	1.47 vol%	3.4	10 wt%	54^C	–	[145]
MWCNT	PU	High shear mixing + Mechanically stirred with CO_2	0.9 wt%	5 wt%	0.01	–	–	–	[146]
Graphene	PMMA	Mechanical mixing + CO_2 saturation	0.6 vol%	1.8 vol%	3.11	1 wt%	30 (+ 12%)	1.01 (+ 20%)	[147]
Graphene	PEI	Mechanical mixing + H_2O induced phase separation	0.18 vol%	1.38 vol%	0.002	5 wt%	5.5 (– 33%)	0.29 (+ 61%)	[148]
MWCNT	PU	Mechanical mixing + H_2O induced forming	1.2 wt%	2.0 wt%	0.0005	2.0 wt%	4.8^C (+ 31%)	0.11^C (+ 31%)	[149]
MWCNT	PU	Ball milling + H_2O_2 treat + mechanical mixing	0.05php	0.3php	25	–	–	–	[150]
R-Graphene	PVDF	Mechanical mixing + AIBN forming	0.5 wt%	10 wt%	10.16	–	–	–	[151]
CB	Epoxy	Mechanical mixing + Microsphere foaming	5 wt%	7 wt%	0.01	7 wt%	10.8^C (+ 316%)	0.24^C (+ 480%)	[152]
Graphene (CVD)	PDMS	PMMA coating + Ni etching + PMMA dissolving	–	0.5 wt%	1000	0.5 wt%	1.7 (+ 30.7%)	0.0018 (+ 38.4%)	[69]
Graphene (CVD)	PDMS	Coating + Ni etching	–	0.8 wt%	200	–	–	–	[153]
Graphene (CVD)	Epoxy	Hot pressing + Ni etching	0.05 wt%	0.1 wt%	97	0.2 wt%	130^F (+ 53%)	3.8^F (+ 38%)	[154]
MWCNT (Polysorbate)	Phenolic	Mechanical mixing + Foaming + Carbonize	1 wt%	5 wt%	1.3	2 wt%	0.47 (+ 264%)	1.76e– 3 (+ 125%)	[155]
Graphene (3D foam)	Epoxy	Mold-casting	–	0.13 wt%	150	0.13 wt%	210^F (+ 10%)	–	[156]
MWCNT	Epoxy	Mechanical mixing + Three-roll milling + Microsphere foaming	0.3 wt%	7 wt%	0.1	4 wt%	7.5^C (+ 50%)	0.18^C (+ 125%)	[157]
MWCNT (Sponge)	Epoxy	Vacuum-assisted impregnation	–	0.66 wt%	148	0.66 wt%	79 (+ 64%)	1.7 (+ 2%)	[158]

F: Flexural properties; C: Compression properties; Others are tensile properties

Table 5 Summary of the electrical and mechanical properties of hybrid carbon nanofiller-reinforced polymer composites

Filler	Matrix	Method	Electrical property			Mechanical property			Refs.
			Percolation threshold	Filler content	Electrical conductivity (S/m)	Filler content	Strength (MPa)	Modulus (GPa)	
GNP/CB	EBA	Extrusion	0.3 vol% (8:2)	14 vol% (8:2)	5	11 vol% (8:2)	8 (–)	0.55 (+ 336%)	[159]
MWCNT/CB	PE	Mechanical mixing + Hot pressing	0.18 wt% (3:1)	4 wt% (3:1)	3.3	–	–	–	[160]
GNP/CB	NBR	Solution mixing + Two-roll milling	–	15ph/10phr	10^{-4}	15ph/10phr	20 (+ 676%)	0.005 (+ 233%)	[161]
Graphite/MWCNT	PE (HD)	Mechanical stirring + Melt mixing + Hot pressing	–	18 wt% (5:1)	10	18 wt% (5:1)	30	1.45	[162]
GNP/MWCNT	Epoxy	Mechanical stirring + Sonication	0.62 wt% (2:8)	4 wt% (2:8)	0.01	0.1 wt% (2:8)	–^{F} (+ 12%)	–^{F} (+ 18%)	[163]
R-GO/MWCNT	PI	Mechanical stirring + Thermal imidization	–	1.1 wt% (10:1)	0.003	1.1 wt% (10:1)	162 (+ 118%)	4.79 (+ 94%)	[164]
R-GO/Oxidized MWCNT	PES	Sonication + Mechanical stirring	0.3 wt% (1:1)	12 wt% (1:1)	0.01	5 wt% (1:1)	78 (+ 11%)	3.6 (+ 100%)	[165]
GNP/MWCNT	Epoxy	Three-roll milling + Hot pressing	–	20 wt% (1:4)	250	20 wt% (1:4)	53 (– 34%)	2.14 (+ 33%)	[166]
GNP/MWCNT	Epoxy	Sonicated + Spin coating	–	0.5 vol% (1:4)	0.018	0.5 vol% (1:4)	64 (+ 21%)	2.9 (+ 17%)	[167]
R-GO/MWCNT (Cu^{2+})	SBS	Sonication	0.25 wt% (1:3)	6 wt% (1:3)	0.2	0.5 wt% (1:3)	–	0.2 (+ 653%)	[168]
GO/MWCNT	PVA	Ultrasonication + Solution casting	– (0.025:1)	1.2 vol% (0.025:1)	0.03	1.8 vol% (1:2)	230 (+ 187%)	13.6 (+ 299%)	[169]
GNP/MWCNT	PVA	Sonicated + Hot pressing	–	1.5 wt% (1:5)	42	1.5 wt% (1:5)	82 (+ 270%)	–	[170]
R-GO/MWCNT	PI	Solution mixing	0.3 wt% (1:1)	1 wt% (1:1)	10^{-4}	1.0 wt% (1:1)	160 (+ 100%)	–	[166]
GNP/MWCNT	PC	Extrusion	–	20 wt% (3:1)	90	–	–	–	[171]

F: Flexural property; Others are tensile property

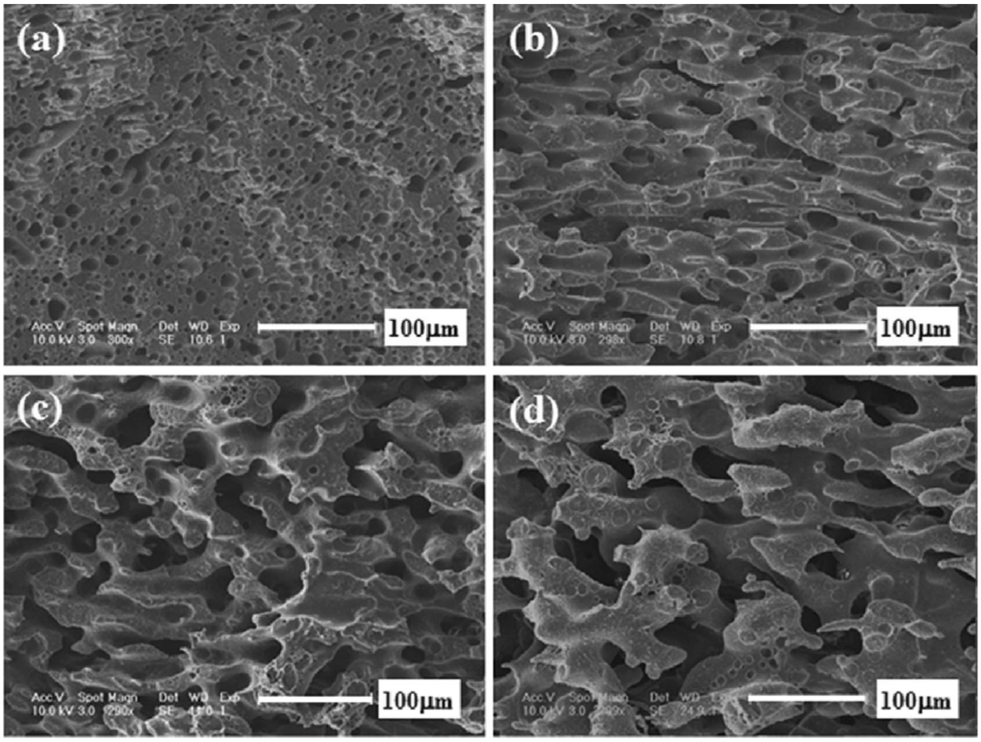

Figure 2 SEM images of co-continuous morphology of PS/PP polymer blend under different PS/PP weight ratio. Reproduced with permission from [17]. Copyright: 2017 Elsevier.

pentagons and 20 hexagons from sp^2 hybridized carbon atoms. However, literature showed that the number of the published papers on the electrical and mechanical properties of fullerene-based polymer composites is significantly less than the CB-based ones. This is probably due to its relatively low electrical conductivity (10^{-5} S/cm) [174] and high cost in comparison to other carbon nanofillers. In fact, some results even showed that fullerene embedded polymer materials can be applied in the energy storage area due to its good dielectric properties and high resistivity [175]. Regarding mechanical properties, surface treatments via acid oxidation and silanization, and fabrication methods such as in situ polymerization were found to be effective approaches for strengthening the fullerene-based polymer composites [21, 176, 177].

One-dimensional carbon nanofiller-reinforced polymer composites

Carbon nanotubes (CNTs)-reinforced polymer composites

Carbon nanotubes (CNTs) are typical one-dimensional conductive fillers due to their remarkably high length-to-diameter ratios (aspect ratio ~ 1000) [178], impressive electrical conductivities (10^7–10^8 S/m) [179] and mechanical properties (modulus 1 TPa, strength 100 GPa) [180]. As such, numerous studies have been conducted on various CNT/polymer composites to explore their electrical and mechanical potentials. Single-walled CNTs which were first discovered by Iijima in 1991 [181] is a capping ended cylindrical structure composed of hexagonal carbon atoms connected by sp^2 bonds. Depending on different fabrication methods, CNTs can be further divided into single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs) as demonstrated in Fig. 4. A SWCNT is shaped from the roll-up of one single layer graphene sheet into a perfectly bonded tube and a MWCNT is formed by a series of

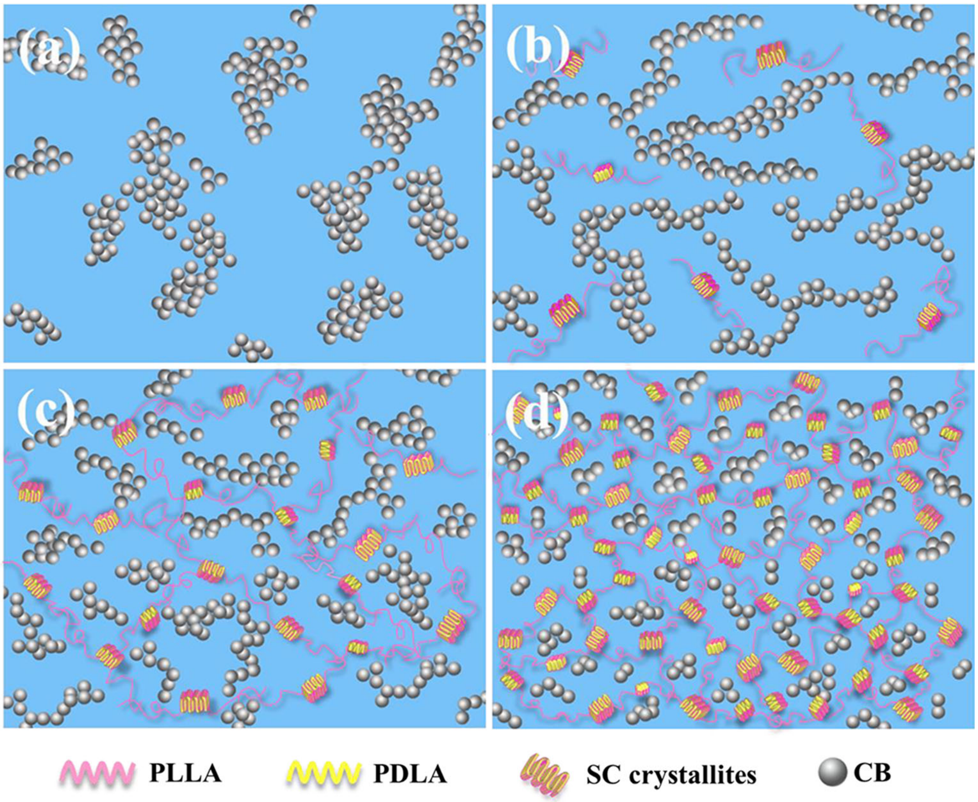

Figure 3 Schematic formation of stereo-complex crystallites to construct conductive pathways in the CB/PLLA nanocomposites. Reproduced with permission from [20]. Copyright: 2018 American Chemical Society.

untouched concentric single-walled tubes linked by Van der Waals force. The distance between these adjacent tubes is 0.34 nm [179].

Since the first reported research of a CNT/epoxy nanocomposite in 1994 [182], tremendous efforts have been made to increase and maximize electrical and mechanical properties of various CNT-reinforced polymeric nanocomposites. From the reported studies, it can be found that the content of CNT filler in the polymer is usually controlled under 10 wt% due to the significant increase of viscosity, which leaded to the poor processability and inferior quality of the final nanocomposite material. Typically, when the CNT filler content exceeds percolation threshold, the electrical conductivity of the CNT-reinforced polymer composites can be increased by 4–10 orders of magnitude due to the superb electrical conductivity and high aspect ratio of CNTs. The random dispersion of CNT fillers in a polymer matrix can form multiple conductive pathways to transform the insulating polymeric material into a conducting composite. However, the mechanical properties of the CNT-reinforced polymer composite are not always satisfied as expected. The modulus and strength of various CNT-based polymer composites vary in a large range from more than 200% to less than 20% of the original polymer matrix (Table 2). Purity, integrity, content, dispersion of the CNT as well as polymer types are all believed to be the essential factors of influencing the electrical and mechanical properties of the composites. To make a systematically analysis, different polymer typed CNT nanocomposites will be reviewed and discussed under thermoset and thermoplastic frameworks as follows.

Thermoset polymer composites

Epoxy resins are one of the mostly used thermoset polymer materials in the world. The unique epoxide functional groups can react with a vast of co-reactants (hardener) to form multiple cross-linked structures, resulting in good mechanical, adhesive, chemical and temperature resistance properties. Taking advantage of these excellent characteristics, researchers extensively investigated epoxy-based CNT nanocomposites worldwide to exploit the potential application in automotive, aerospace, civil construction and electronics industries [183].

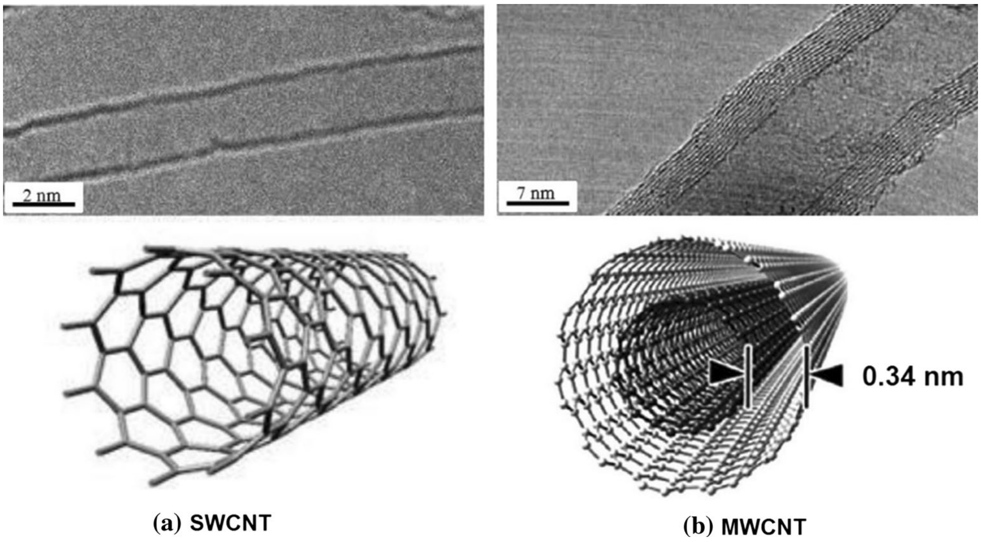

Figure 4 TEM images and molecular structures of different typed CNTs (a) single-walled CNT (b) multi-walled CNT. Reproduced with permission from [180]. Copyright: 2016 Elsevier.

Similar to CB, CNTs also tend to form aggregates that prevent the fully display of the attractive features in the nanocomposites. Therefore, significant efforts have been contributed to improvement of the dispersion of CNTs in polymer matrices, for which the methods were categorized into physical and chemical methods. Physical methods are those that mechanically disperse CNTs in the polymers by intensive stirring [22, 23, 39, 46] ,ultrasonication [25–27, 35, 36, 41, 43, 45, 48], high shear mixing [28, 45], ball milling [25, 29, 35], spin coating [45] hot pressing [30, 35, 42] and combination of some of these methods [25–28, 31, 33, 36, 45, 48]. The reported results showed that the electrical conductivity of the non-treated CNT/epoxy composites can reach a range from 10^{-5} to 10 S/m within 5 wt% depending on the specific parameter settings and the geometric characteristics (aspect ratio) of the nanofillers [39]. On mechanical properties, however, the improvement was marginal and even decreased with addition of CNTs in the epoxy matrix [36, 42], which was largely caused by the entanglement of CNTs and poor interfacial bonding between CNTs and the polymer matrix.

To solve the entanglement issue, chemical methods were applied via introducing chemical moieties covalently or non-covalently onto CNTs to simultaneously improve dispersion and strengthen interfacial bonding with the resin matrix material. Strong acids were mostly used to untangle C–C bonds and generate carboxyl groups [184, 185], amino groups [35, 186], silane groups [32], epoxide groups [187], etc., on the sidewall of CNTs. These chemical methods resulted in remarkable increases in the mechanical performance for the bulk nanocomposites. UV/O_3 and plasma treatments are two other interesting methods to relax inter-tube interactions by introducing hydrophilic groups such as oxygen molecules on the surface of CNTs to facilitate good dispersion in the epoxy [30, 32, 33, 37]. However, excessive chemical modification can damage the π conjugation of the CNTs and results in adverse effect to its superb electrical property. As such, non-covalent modifications using surfactants were proposed as alternative approaches for fabricating high-performance nanocomposites. It is noted that, apart from frequently used surfactants such as sodium dodecyl sulfate (SDS) [73, 96] sodium dodecylbenzene sulfonate (SDBS) [94, 188], polystyrene sulfate (PSS) [189], polyphenylene ethynylene (PPE) [65], polyaniline (PANI) [47, 190], Triton X [129], environmentally friendly bio-surfactants such as gelatin [48], soy protein [117], chitosan [191] have been applied to functionalize CNTs for making epoxy nanocomposites with impressive augments of both electrical and mechanical properties (Fig. 5). Some results suggested that these protein molecules were not only effective surfactants, but also compatible with different types of polymer matrices (e.g., epoxy, PC, PEO), showing adaptive interface properties [117].

Besides the above two major methods, some novel fabrication routes to artificially manipulating orientation, loading and distribution of CNTs were

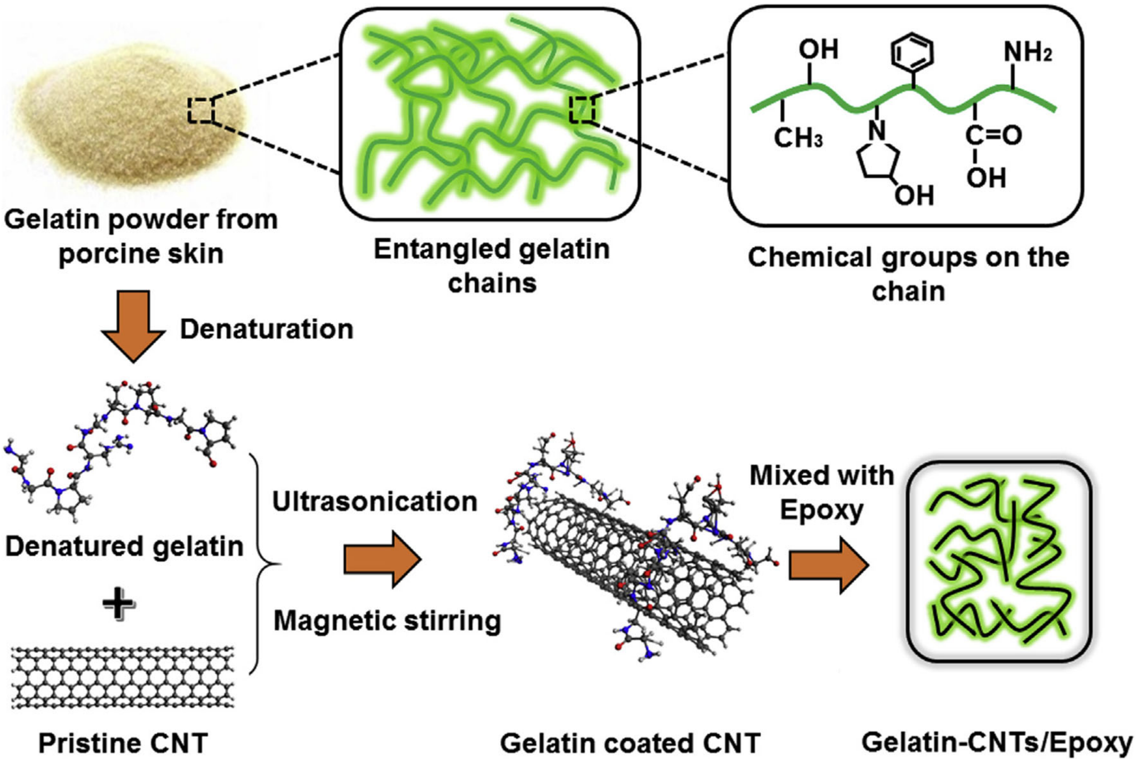

Figure 5 Schematic of the fabrication procedure of gelatin-CNT/epoxy nanocomposite. Reproduced with permission from [48]. Copyright: 2018 Elsevier.

proposed. Mei et al. [40] developed an ice templating method to prepare a CNT/epoxy composite. The researchers freeze-dried the polymer mixtures in a narrow, upper cover-opened container to create a one-directional temperature gradient to realize aligned lamellar CNT structures with conductivity up to 15.8 S/m with only 1.5 wt%. Khan et al. [37] proposed a similar CNT-aligned nanocomposite via applying a DC electric field during the curing process as presented in Fig. 6. It was shown that the percolation of the nanocomposites was only 0.0048 wt% and modulus and fracture properties were excellent. Mecklenburg et al. [38] reported a hot-press infiltration method to obtain an ultra-high loading nanocomposite. The critical technique was to use a semi-permeable membrane to enwrap CNT assemblies to impregnate epoxy resin. A composite with a loading of 56 wt% was achieved with significantly high electrical conductivity (3000 S/m) and modulus (15.5 GPa). Zaidi et al. [44] fabricated a highly conductive CNT/epoxy composite with the help of supercritical carbon dioxide. The dilated CO_2 gases made CNTs redistributed in a segregated interconnected framework with good conductivity and strength.

Thermoplastic polymer composites

Polyurethane (PU)-based matrix Thermoplastic polyurethane (TPU) is a linear segmented block copolymer composed of two special parts called hard segment and soft segment. The hard segment is usually based on aromatic or aliphatic isocyanates; the soft segment is a polyether or polyester typed polyol. By varying the ratio of these two components, TPU can have a considerable number of physical properties, which make it an ideal CNT-based matrix for fabricating nanocomposites with superior multifunctional properties.

To achieve good dispersion of CNTs in the TPU matrix, chemical modification with acids [52], surfactants [51], in situ polymerization [49, 53, 56, 59] are mostly used procedures. In the meanwhile, some post-processing techniques like annealing [57], spray drying [58], roll milling [50] are also found to be beneficial to the uniformity of CNTs. However, it was found that the CNTs could influence the crystalline

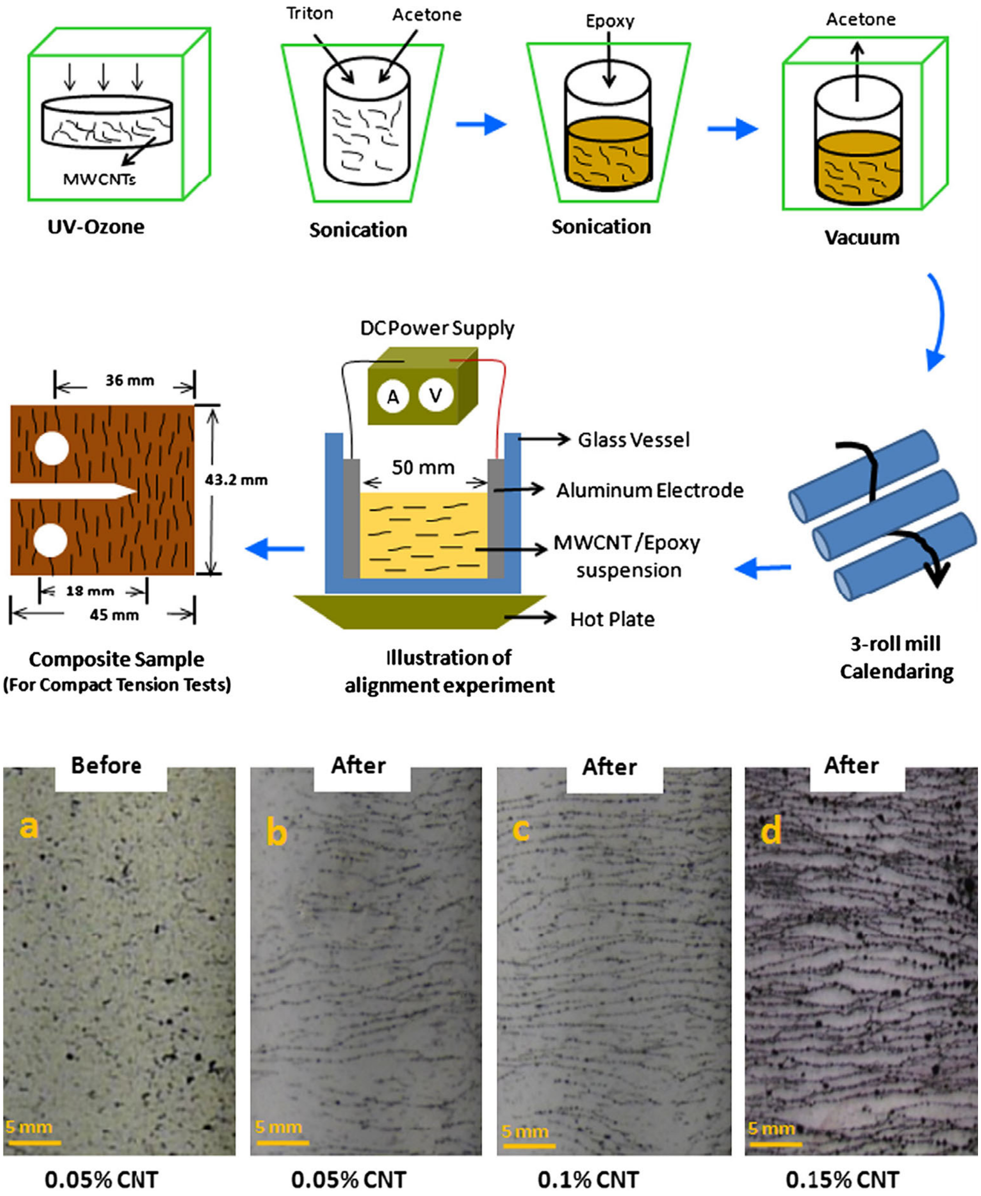

Figure 6 Fabrication method (above) and CNT alignment in acetone solution under different loadings (below). Reproduced with permission from [37]. Copyright: 2013 Elsevier.

behavior of the soft segment of TPU and damage the strength of the resulting composite [49]. A novel approach to facilitating filler agglomeration that can accelerate the polymerization rate during the in situ processing of the composites via microwave irradiation or addition of catalyst was reported. For example, Yakovlev et al. [54] found adding catalyst such as iron acetylacetonate (Fe(acac)$_3$) could greatly increase the formation of urethane bonds to achieve cross-linked PU/CNT with significantly low percolation threshold. In another work, a solvent/nonsolvent one-step filtration method was proposed to fabricate TPU/CNT composite as shown in Fig. 7 [55]. The core idea was to mix a solvent which can well dissolve TPU (acetone) with a CNTs filled nonsolvent (methanol) so that phase separation could occur that drove the polymer onto the surface of the CNTs by non-covalent interaction. Through this approach, the weight ratio between CNTs and TPU were randomly designed and precisely controlled, resulting in a composite with ultrahigh conductivity (2300 S/m) and significantly improved mechanical properties (e.g., 950% increase in modulus).

Polyamide-6 (PA-6)- and polycarbonate (PC)-based matrix Polyamide-6 and polycarbonate are two widely used thermoplastic polymers because of their excellent mechanical, abrasion and chemical resistant

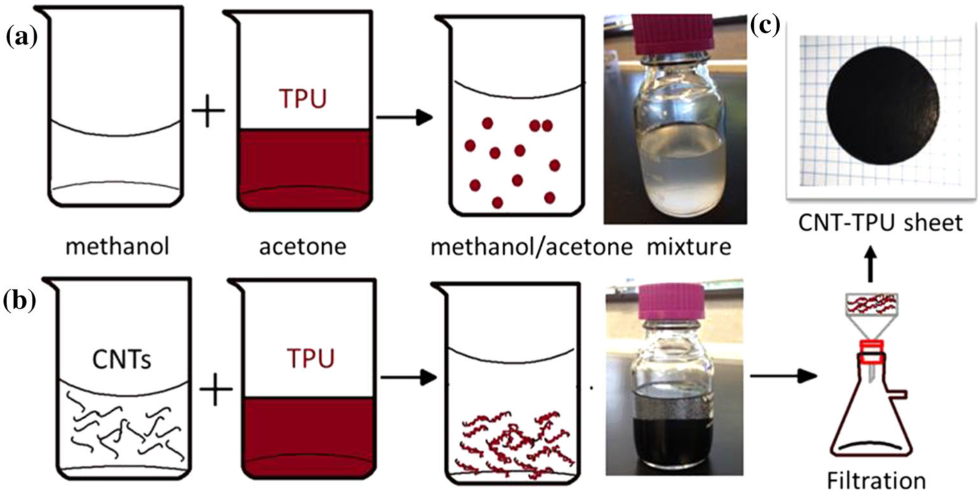

Figure 7 Schematics of the one-step CNT-TPU sheets fabrication method. **a** TPU phase separation in the solvent/nonsolvent mixture. **b** TPU immobilization on the CNTs surface. **c** Photo of a TPU-CNT composite sheet [55]. Copyright Published.

properties, as well as optical, thermal properties and dimensional stabilities. However, the poor dispersion of CNTs caused by the high melt viscosity of the two matrices hinders their applications of the nanocomposites. Extrusion [60, 61, 64, 192] and surface modification [62, 63, 65, 69] of CNTs along with the combination of the two techniques are good choices to obtain CNT/PA-6 and CNT/PC composites with improved performance. Specifically, Babal et al. [70] prepared MWCNT/PC composites using an industrial twin-screw extruder with back flow channel (Fig. 8). The tensile and flexural properties of the composite increased by 15% with only a very low filler loading. Maiti et al. [67] proposed a novel method (Fig. 9) to selectively localize CNTs at the interfaces of PC beads to fabricate conductive segregated CNT/PC composite with a high electrical conductivity (0.45 S/m) at extremely low filler content (0.02 wt%). For surface modification efforts, Ryu and Han [62] found that by non-covalently functionalizing pyrene moieties with longer alkyl chains on the CNTs could dramatically enhance the electrical and mechanical properties of the composites due to the improved dispersion and formation of the α-crystalline form during the processing of the PA-6 composites. Kim et al. [63] designed a novel surfactant (poly(3-hexylthiophene)-g-polycaprolactones, short for P3HT-g-PCLs, Fig. 10) and grafted it to the MWCNTs by non-covalent bonding. The conductivity of the surfactant treated CNT/PC composite dramatically increased to 64 S/m at 5 wt% loading due to the significantly improved dispersion of CNTs. The strong π–π interaction between CNTs and surfactant also benefited for the interfacial bonding between CNTs and the matrix, contributing to a 50% increase for both modulus and tensile strength.

Polyethylene (PE)-based matrix Polyethylene (PE), which consisted of numerous ethylene monomers ($(C_2H_4)_n$), is one of the simplest structured and commonly used plastics. Based on its density and branching, PE can be subsequently classified into ultra high molecular weight polyethylene (UHMWPE), high-density polyethylene (HDPE), linear low-density polyethylene (LLDPE), etc., among which UHMWPE is considered as a superb matrix candidate to be used in the nanocomposite areas due to its outstanding mechanical, chemical resistance, moisture resistance and tribological performances. However, because of its high melt viscosity and molecular weight, traditional molding and extrusion methods such as melt compounding and injection molding would break its long molecular chains and negatively influence its excellent properties. Thus, alternative processing such as dry mixing [73, 75], ball milling [72] combined with hot pressing [65, 67, 72–75, 77], sintering [76] or blown film extrusion [78] were investigated, in particular for the improvement of electrical and mechanical properties of CNT/PE composites. Many researchers attempted dry mixing followed by hot pressing or compression molding to disperse nanofillers directly in the macromolecular PE powders to avoid complex chemical-based mixing methods [73–75]. The advantage of this process is that CNTs can gather at the interfaces of PE powders to form multiple conductive paths, which is beneficial for the creation of high electrical composites at relatively low percolation

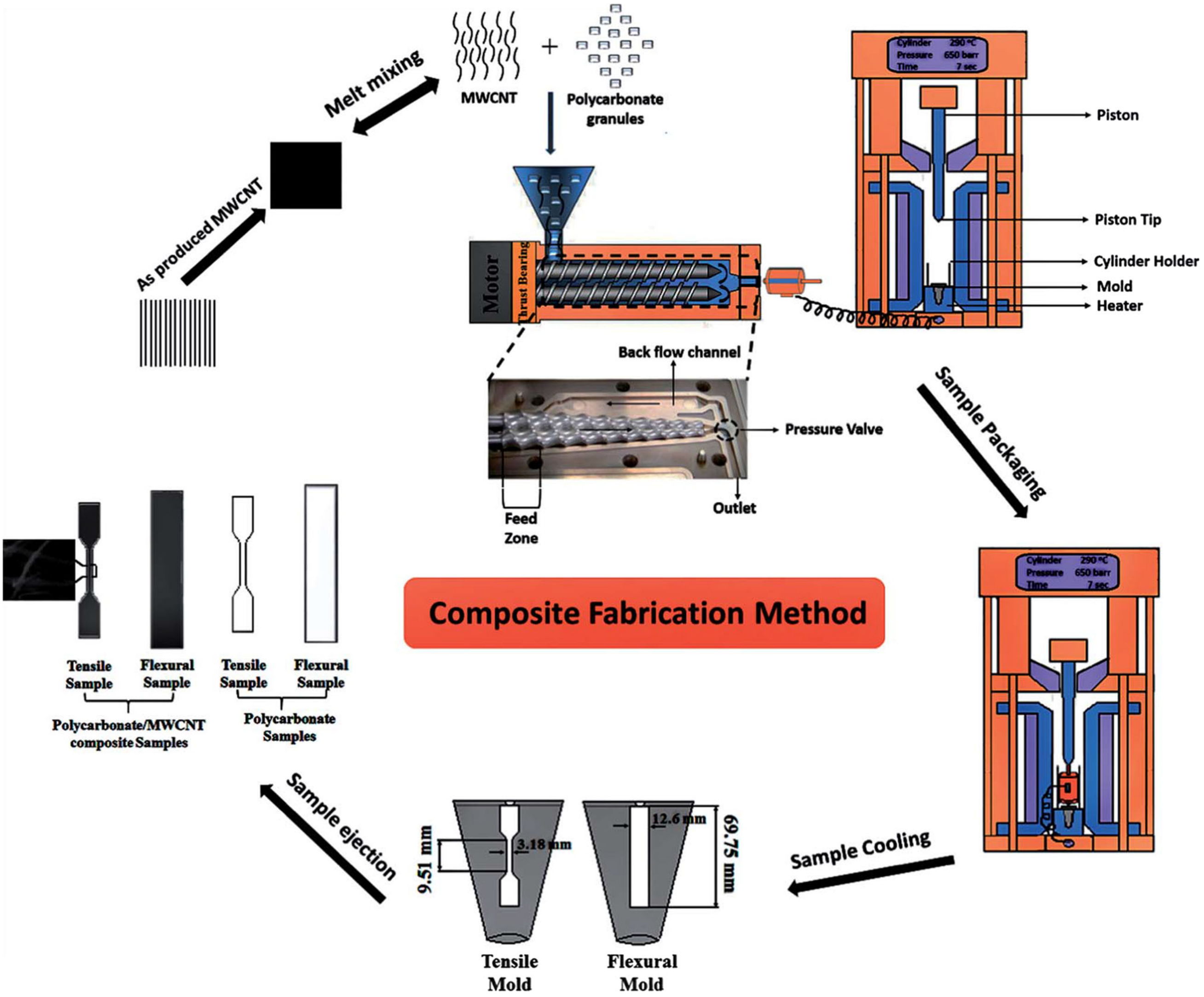

Figure 8 Preparation of MWCNT/PC composites by twin-screw extrusion procedure. Reproduced with permission from [70]. Copyright: 2014 RSC Publishing.

(Fig. 11a). However, parameters such as molecular weight of PE, compaction temperature, residence time and even sample thickness et al. [76, 77] should be carefully selected, which greatly influences the CNT dispersion and the crystallization of the polymer matrices.

Polypropylene (PP)-based matrix Polypropylene is gathered from chains of propylene monomers which has similar structure to PE. Good chemical, thermal, dielectric, optical as well as mechanical properties make it a desirable candidate in the automobile, civil, toy and medical industries. The most commonly used method to fabricate CNT/PP composites is extrusion/injection molding and hot pressing with the electrical conductivity attaining around 0.1–10 S/m in less than 2 wt% [79, 80]. Recently, a new method called latex technology to fabricate highly conductive CNT/PP composites was proposed [82, 84] via mixing CNTs with latex PP matrix (aqueous emulsion) to form a mixture, then freeze-dried (remove aqueous solution) and hot pressed to fabricate the composites (Fig. 11b). Similar to the dry mixing method mentioned above, the latex technology could also manipulate nanofillers to be distributed the interfaces of polymer particles to achieve good electrical and

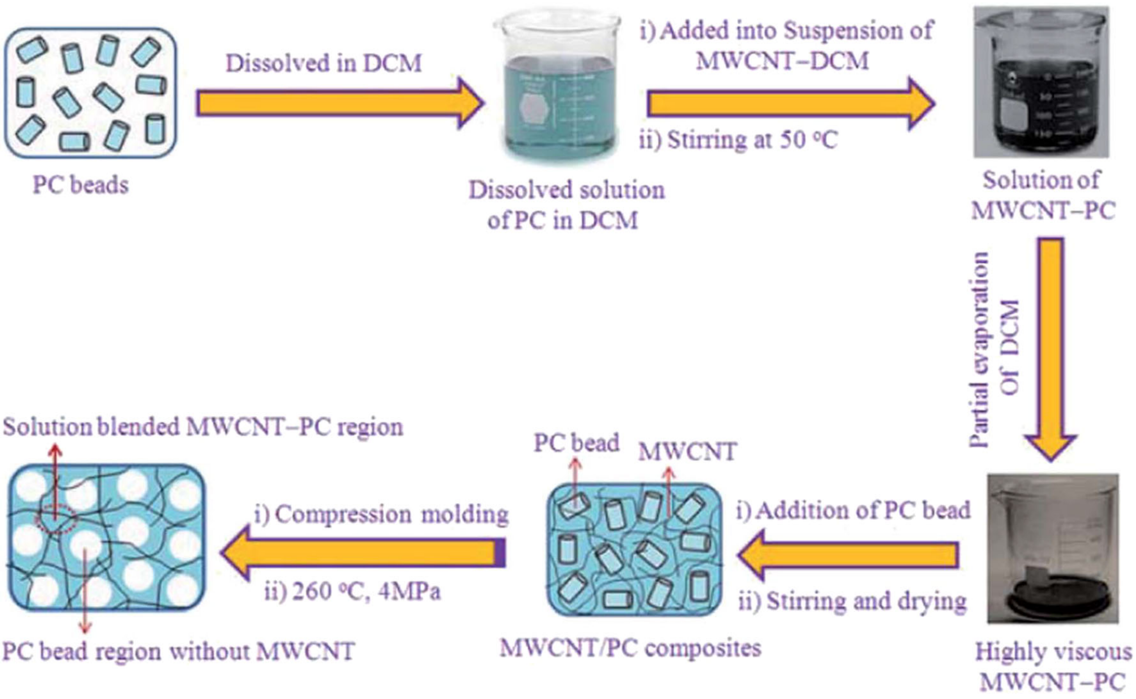

Figure 9 Preparation of MWCNT/PC composite by blending PC beads with MWCNTs. Reproduced with permission from [67]. Copyright: 2014 RSC Publishing.

mechanical properties with an ultralow filler content. However, this method was restricted to the highly viscous polymers to prevent the diffusion of CNTs into the polymer matrix. For semi-crystalline low-viscosity polymers, Wu et al. [84] wisely selected temperature and pressure as the hot-pressing parameters according to the phase diagram of the polymer (melt temperature curve), and then obtained the composite with ultrahigh conductivity (117 S/m at 5 wt%) and electromagnetic interference shielding (EMI) shielding properties. An excellent review paper on fabrication of segregated conductive polymer composites using this method can be referred in [6].

Polystyrene (PS)-based matrix Polystyrene is an aromatic thermoplastic polymer polymerized from monomer styrene. It has widely been used in the consumer products like cups, dishes, bags and commercial packaging due to its inexpensive cost, excellent mechanical properties and chemical stability. Traditional fabrication methods for making CNT/PS composites are mechanically blending or in situ polymerization coupled with hot pressing, injection molding, casting or spin coating [89–91]. However, these methods usually require lengthy blending or sonication, which may damage CNTs and decrease its electrical or mechanical performance in the polymer matrix. In terms of the significantly high viscosity of PS matrix, recently many researchers attempted the latex technology mentioned above to make high quality CNT/PS composites. Solvents such as dimethylformamide (DMF), tetrahydrofuran (THF) [90], acid [88, 193], chloroform [89] and surfactants such as cetyltrimethylammounium 4-vinylbenzoate (CTAVB) [87], sodium dodecyl sulfate (SDS) [92, 93, 95–97], sodium dodecylbenzene sulfonate (SDBS) [94] were used first to increase the dispersion of CNTs, then blended and freeze-dried in the latex PS matrix to form conductive paths at latex interfaces, finally hot pressed to finish specimen fabrication. The maximum electrical conductivity can be achieved to 10^2 S/m in 3% CNT loadings [92]. It is noted that the type and content of surfactants [93, 96], molecular weight of the matrix [92, 97] and molding parameters [94] are essential to the performances of the nanocomposite made via latex technology.

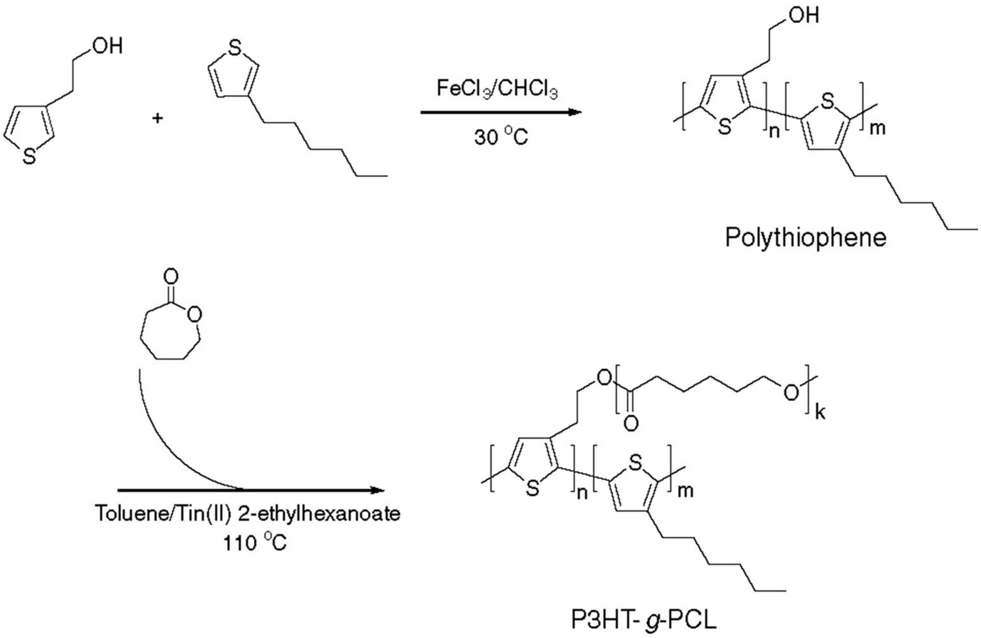

Figure 10 Synthesis procedure of surfactant P3HT-g-PCLs. Reproduced with permission from [63]. Copyright: 2009 Elsevier.

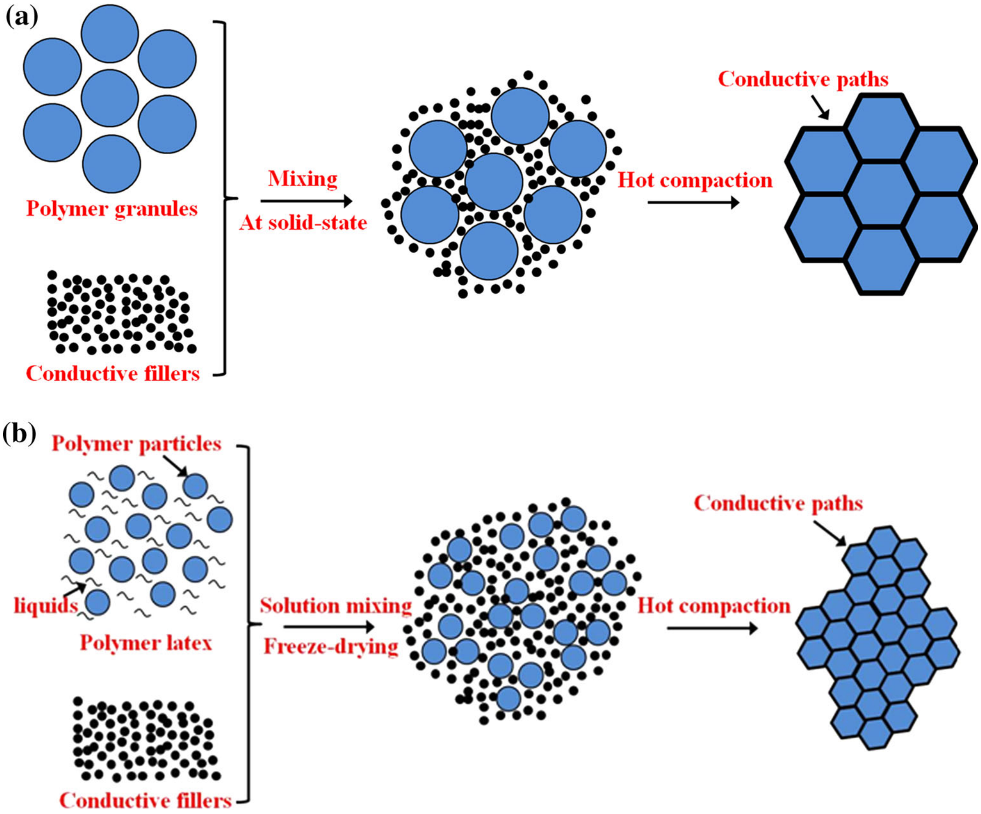

Figure 11 **a** Dry mixing and **b** latex technology methods to fabricate segregated conductive nanocomposites. Reproduced with permission from [6]. Copyright: 2014 Elsevier.

Poly(methylmethacrylate) (PMMA)-based matrix Poly(methylmethacrylate) (PMMA) is another commonly used thermoplastic polymer. Its excellent transparency, mechanical properties and lightweight make it a suitable alternative to polycarbonate and glass. Like other thermoplastic-based nanocomposites, good dispersion of nanofillers in the matrix is the most critical issue to be solved. Apart

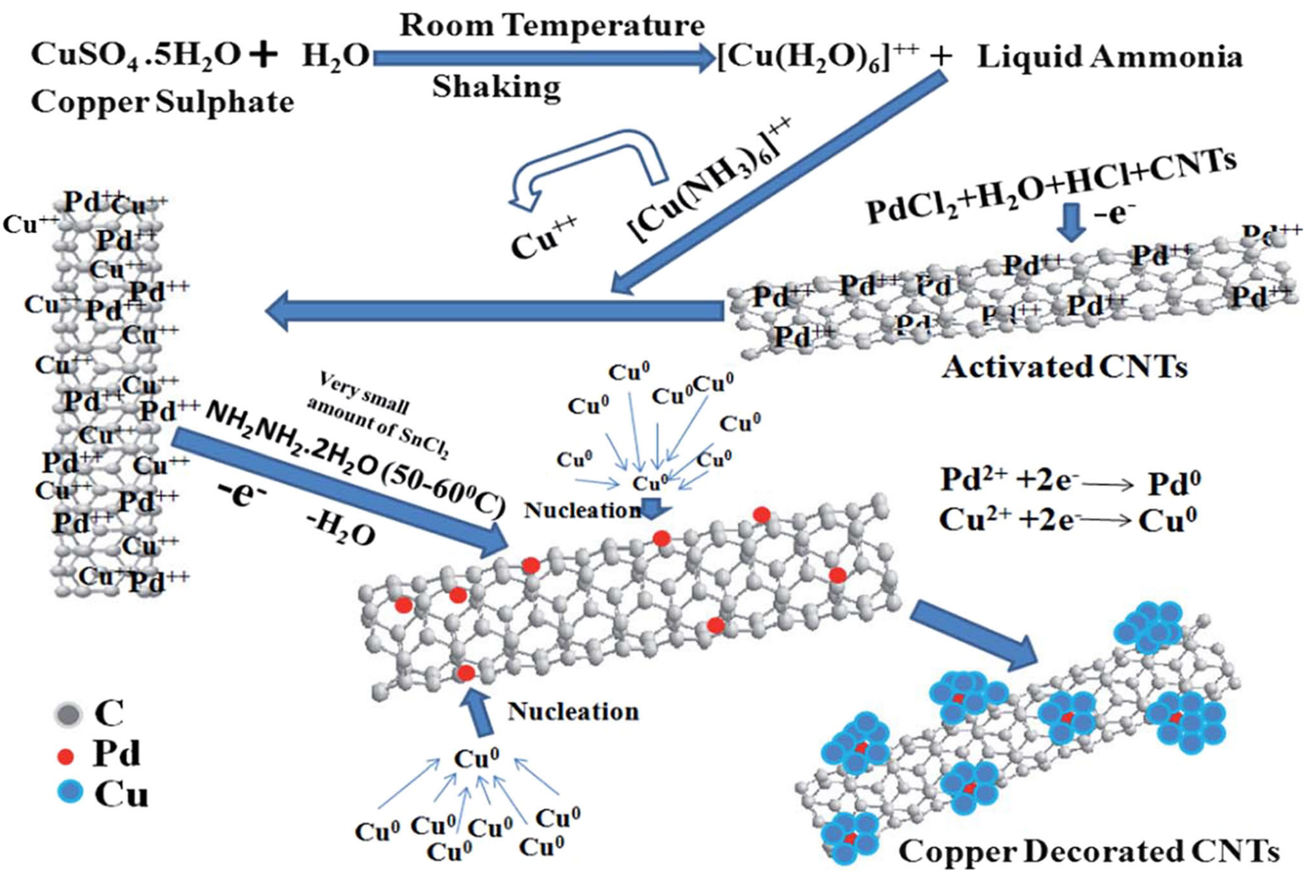

Figure 12 Synthesis procedure for the fabrication of Cu-CNTs by Pd linkage. Reproduced with permission from [198]. Copyright: 2015 Royal Society of Chemistry.

from the popular latex technology [92, 93], some researchers [99, 101, 103] found that $SOCl_2$ and COOH– functionalized CNTs can significantly increase both electrical and mechanical performance of the composites. Koysuren et al. [98] non-covalently coated poly(glycidyl methacrylate) (PGMA) on the surface of CNTs via initiated chemical vapor deposition (CVD) method, which greatly improved the interaction between the nanofiller and the matrix.

Metal-CNTs-reinforced polymer composites

Metal-CNTs are a new type of functionalized nanotubes decorated with noble metal nanoparticles (NPs) such as Au [194, 195], Ag [35, 50, 104–108], Cu [111, 196], Pt [195, 197], Rh [195] and their combinations [198]. Due to the unique conductivity, magnetic and active chemical behavior of the metal NPs, this new type of nanofiller can further elevate the properties of nanotubes to be used in broader areas, especially for electronic, medical, biological and energy sectors [199]. Generally, there are two ways to attaching metal NPs onto CNTs: (1) direct attachment of metal NPs on the CNTs; (2) indirect attachment of metal NPs around CNTs through specific linkages. Direct attachment is to introduce moieties such as carboxyl and hydroxyl groups on CNTs by means of acid, laser, sputtering, electron-beam or thermal evaporation, etc., to provide harbor points for the deposition of NPs [35, 104, 105, 107, 108, 111, 194–196, 200]. This method is always followed by a reduction process through reducing agents such as DMF [35, 104], dodecylbenzenesulfonic acid (DBSA) [105], hydrazine [107, 200], Ethylene glycol [194], polyethylene glycol (PEG) [195] to transfer metal ions to NPs. Although it is a simple and effective method, the damaged CNT sidewalls will inevitably decrease their essential properties and degrade the performances of the bulk polymeric composite in addition to the expenditure of vast amounts of hazardous

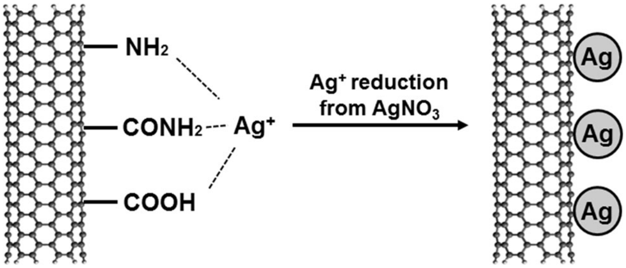

Figure 13 Ag nanoparticles attach on the functionalized CNTs.

chemicals. On the other hand, indirect attachment is to bridge CNTs and NPs by molecules or chemical groups under chemical or physical interactions. For example, Li et al. [197] proposed a comprehensive approach to fabricate metal-, semiconductor- and insulator particle-CNTs by adoption of 1-aminopyrene. Kumar et al. [198] doped Pd on the surface of CNTs as nucleation sites to anchor Cu NPs (Fig. 12). Chun et al. [106] used a synthesized "Ag NPs with phenyl rings" molecular to associate CNTs and Ag flakes and obtained a conductive polymer composite with significantly high electrical conductivity (10^5 S/m). Compared to the direct attachment approach, the indirect one could largely preserve the nanofiller to fulfill its functionality and achieve superb properties although the material design procedure was sophisticated and highly technical. Even though many noble metal particles have been reported to be grafted to CNTs, very few of them have been implemented in the application of conductive polymer composite except for Ag NPs. Amino [35, 104] and carboxyl [107, 108] groups functionalized CNTs have been reported to successfully capture Ag NPs by electrostatic interactions (Fig. 13). These modified nanofillers were easily mixed with PP, PS, epoxy, PU, PANI, PVDF, etc., to fabricate conductive polymer composites with conductivity higher than 10^2 S/m. However, some researchers pointed out that the pH of the CNT solution influenced the formation and the density of Ag-NPs on the CNTs [35]. Some other studied showed that the metal particles could not only increase the electrical performance of the composites but also helped to improve the dispersion and interfacial adhesion of the CNTs, resulting in better mechanical properties compared to pristine CNT/polymer composites [35, 104, 110–112].

Carbon nanofibers (CNFs)-reinforced polymer composites

Besides CNTs, another frequently used one-dimensional carbon nanofiller is carbon nanofiber (CNF), as shown in Fig. 14. Different from the tube-like structure of CNTs, the typical CNFs are consisted of a series of graphite basal planes stacking one by one with an angle of about 25° along the longitudinal direction of the fiber [113]. Due to this unique feature, the diameter of the CNFs (50–200 nm) is usually larger than CNTs ($\sim$ 10 nm), while more importantly, active carbon atoms are exposed on the outside surfaces of the nanofibers that effectively increase their chemical reactivity [114]. Although the mechanical, electrical and thermal properties of CNFs are slightly lower than that of CNTs, the low production cost along with their good material properties still makes CNFs an excellent carbon nanofiller for application in the polymeric industries.

To obtain uniform dispersion of CNFs in a polymer matrix is always the essential issue for achieving good performed nanocomposites. Many of the fabrication methods applied in the CNT-based composites can also be used for making CNF composite because of the similarity between the two nanofillers, which can be roughly categorized into solution mixing and melt mixing. Solution mixing includes mechanical stirring [201], ultrasonication [115, 202], solvent casting [116, 203], etc. Sometimes, chemical modifications such as silanization [204], surfactants [117], and in situ [116, 119, 203], as well as physical treatments such as plasma [205] and high temperature heat treatment [113], were involved to preprocess CNFs to enable better dispersion quality in the solutions. However, the effects of these treatments on the final electrical and mechanical properties of the composites were not always positive. For example, Nie et al. [204] found silanization of CNFs via APTES could effectively strengthen the tensile and shear strength of the composite while its electrical conductivity deceased more than 16 times compared with the untreated ones. Guadagno et al. [113] showed high temperature heating could significantly lower percolation threshold and increased electrical conductivity through enhancement of stiffness and smoothness of CNF surfaces. However, this treatment of CNF surface weakened interfacial bonding and lowered its bulk mechanical properties. When the viscosity of the polymer matrix was high, melt

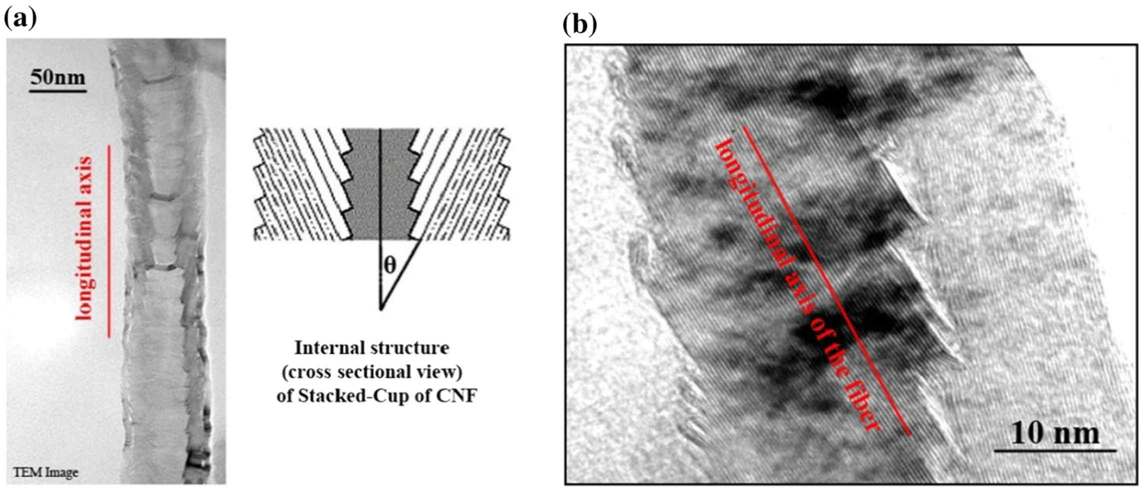

Figure 14 TEM images of carbon nanofiber structure in different magnification levels [113]. Copyright Published.

mixing approach such as twin-screw extrusion [114, 206] was applied to disperse CNFs in the matrices. Nevertheless, the high shear forces generated from extrusion greatly shortened CNFs and reduced the electrical performance. To solve the problem, chaotic mixing [118], surface pre-treatments combined with low shear mixing [207], condensation cure method [120] and solution cum melt mixing [121] were proposed and resulted in improved properties.

Two-dimensional carbon nanofiller-reinforced polymer composites

The most representative two-dimensional carbon filler is graphene, a one-atom-thick hexagonal lattice connected carbon nanomaterial. Although it has long been known that graphene is the basic layer of graphite, the discovery of the individual graphene did not come until in 2004, which led to the two scientists' Nobel Prize [208]. The quick and wide acceptance of graphene that has great potential for many important and significant application is due to its superb properties, such as extremely high mechanical properties (e.g., 1.1 TPa modulus), electrical conductivity (10^8 S/m), thermal conductivity (5000 W/mK) and optical properties (98% transmittance) [209], which make it a fascinating nanofiller for making multifunctional polymer-based nanocomposites. Various fabrication methods, such as scotch tape, chemical exfoliation, reduction of graphene oxide and CVD have been designed to produce graphene with high quality and large scale, generating several modified graphene derivatives such as graphene oxide (GO), graphite nanoplatelet (GNP) and expanded graphite [179, 210]. All of these materials can be basically categorized into two types: graphene and graphene oxide materials. Accordingly, in this section electrical and mechanical performances of graphene-based and graphene oxide-based polymer composites will be briefly reviewed.

Graphene-based polymer composites

Graphite nanoplatelet (nanosheet) or expanded graphite is a 2D stacked nanofiller yielded either from the exfoliation of natural graphite or derived from graphite intercalation compounds (GIC), an intermediate modified graphite with different atomic or molecular chemicals (e.g., acids and alkali metals) between the graphene layers. Followed by excessive heating and ultrasonication, hundred to thousand length of expanded graphite nanoplatelet with a thickness ranging from a few to a hundred nanometers can be obtained [211].

Excellent material properties of GNPs enable its wide application in the polymeric-reinforced nanocomposites. However, high surface-to-mass ratio, molecular scale forces and dispersion of GNPs in the polymer matrix are big issues to prohibit its full potential and influence the overall properties of the composites [124]. Some researchers used very long sonication time (2–3 h) to break GNP agglomerates in the polymer matrices [126, 127]. Their results showed very high percolation thresholds and maximum electrical conductivity filler content. Although the electrical performance of the GNP/polymer composites was improved (Table 3), the aggressive method would inevitably waste a large amount of GNP fillers by damaging their structural integrity. Alternative methods, such as three-roll milling, high shear mixing and injection molding, have been tried with improvements on the properties of nanocomposites [125, 128, 136]. Latex technology is an effective way to tackling the agglomeration of GNPs by forcing them on the interfaces of the polymer emulsions. Ghislandi et al. [82] adopted this method to fabricate a highly conductive graphene/PP composite (1 S/m) with relatively a low percolation threshold (1.2%). They also noticed that hot pressing may improve the electrical property by re-orienting nanofillers in the matrix. Li et al. [122] found that by treating GNPs with UV and ozone before mixing with polymer matrices could eliminate the highly resistant interfacial voids and gaps, which led to notably increase in conductivity and modulus of the nanocomposite. In other reported studies, surfactants, such as PVP [123], Triton [129], APTES [141], and dioxolane [142], were used to cover graphene fillers in order to improve the dispersion and distribution states, resulting in very good conductivity and mechanical properties.

Graphene oxide (GO)-based polymer composites

The poor dispersion of graphene or GNPs is largely due to their insolubility in most of the commonly used solvents. Since graphene oxide (GO) derived from oxidation of pristine graphene nanofillers contains various hydrophilic groups such as epoxide, hydroxyl and carboxyl, it can be easily dissolved in water and various water-soluble polymers. As such, many researchers used GO as a modified graphene nanofiller to fabricate polymer nanocomposites. Ryu and Shanmugharaj [137] grafted long chain alkylamines on GO through amidation and nucleophilic reactions to covalently strengthen the interfacial bonding of the PP-based composite. However, the disadvantage of GO is that, unlike graphite or graphene, GO is interior in electrical conductivity, which must be reduced to RGO to restore electrical conductivity. The most frequently used reduction agents are hydrazine and DMF [130–133, 138], which have strong reduction effect and low cost. However, these chemicals are highly toxic and harmful to human beings and environments. Due to these reasons, some researchers studied green reduction agents like vitamin C [134, 212], saccharide [213], hydrogen iodide [135] and thermal treatment methods (imidization) [139, 140] to replace hydrazine. For example, Long et al. [134] adopted vitamin C to reduce GO and fabricate RGO/PS nanocomposites with impressive conductive (20.5 S/m). They noticed that hot-pressing step could also convert GO to RGO, which can be a supplementary way to reducing graphene fillers. However, some research results showed that these RGO re-agglomerated into large bundles again by π–π interactions and decreased the properties of the composites. To solve the problem, non-covalent treatment [133] and in situ polymerization [139, 140] were utilized to improve dispersion of RGO after chemical reduction. Pham et al. [130] designed a modified latex method to obtain a highly conductive RGO/PMMA nanocomposite by electrostatic interactions (Fig. 15). Wu et al. [135] applied the same fabrication method using a green reduction agent (hydrogen iodide) and successfully produced a 3D foam-like graphene structural polymer composite with a surprisingly high electrical conductivity (1083.3 S/m).

Three-dimensional carbon nano-foam-reinforced polymer composites

Fabricating three-dimensional (3D) nano-foam structures is a novel route to achieving highly conductive nanofiller-reinforced composites. The widely used protocol was to disperse nanofillers, such as carbon nanotube [145, 146, 149, 150, 155, 157, 158], graphene [69, 136, 144, 147, 148, 151, 156], carbon black [152], carbon nanofiber [214] or their combinations [215] in the solution and mix with proper polymer matrices uniformly. Then, foaming agents, such as NH_3 [144], CO_2 [145, 147, 215], H_2O [148, 149], AIBN [214], pentane [155] and expandable microspheres [152, 157] were added into the mixture to allow gas bubbles to be initiated, grown and broken to form "sponge"-like 3D form structure, as shown in Fig. 16. It can be seen that during the foaming process the nanofillers are forcefully squeezed into multiple interconnected narrow pathways by the bulge of gas bubbles, thus avoiding the problem of agglomeration

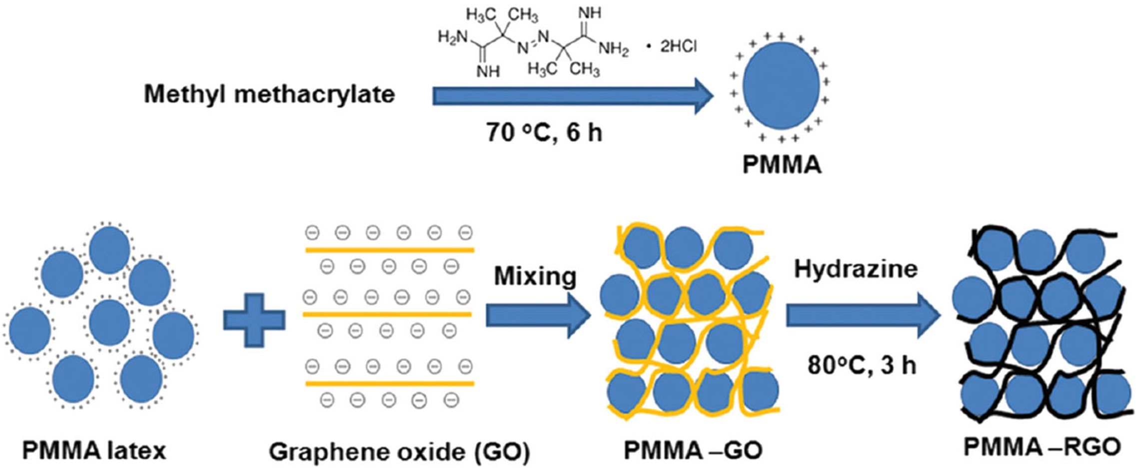

Figure 15 Fabrication method of RGO/PMMA nanocomposite by electrostatic interactions. Reproduced with permission from [130]. Copyright: 2012 American Chemical Society.

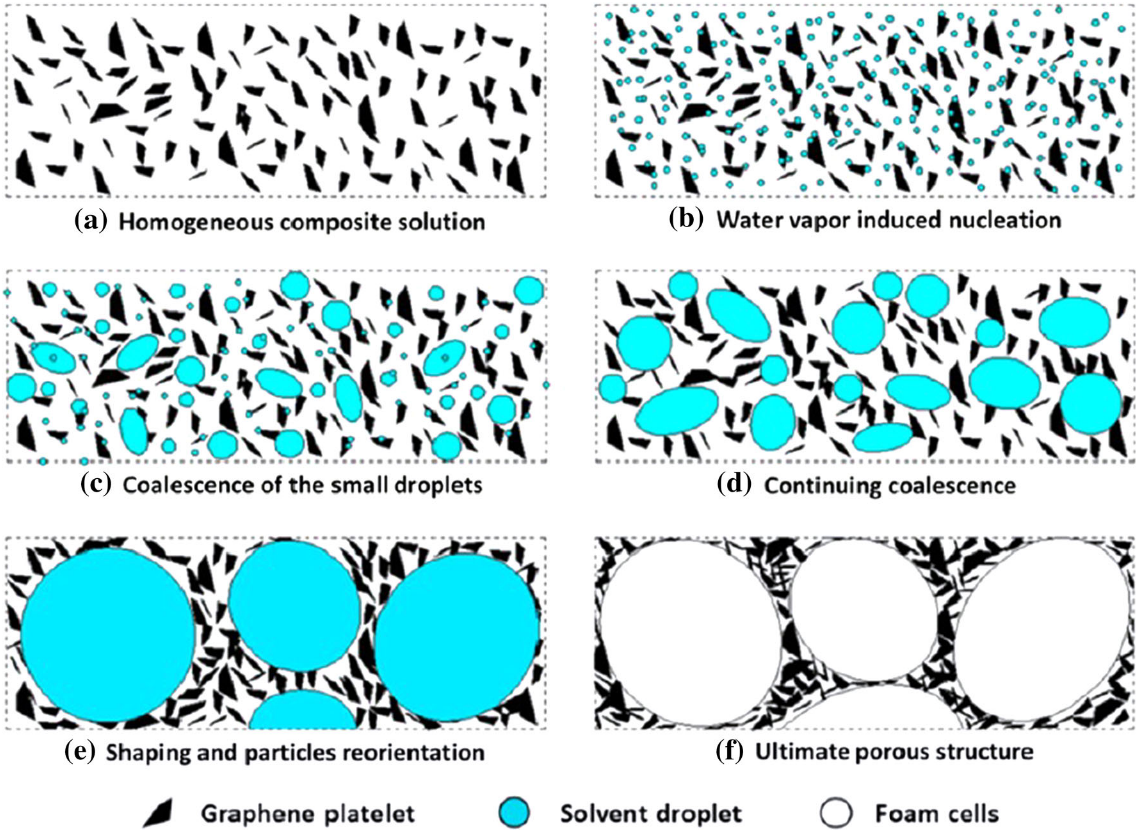

Figure 16 Typical foaming process to fabricate 3D nano-foam structure. Reproduced with permission from [148]. Copyright: 2013 American Chemical Society.

happening from direct mixing of nanofillers in the polymer matrices. The highly efficient conductive pathways could significantly decrease the loading of nanomaterials and increased the conductivity and mechanical properties of the composite. However, these properties were highly influenced by the types of filler materials, polymer matrices, foaming agent contents and temperature as well as fabrication methods [136, 156, 157]. For instance, Jun et al. [136] found that larger sized graphene sheets could greatly improve the electrical conductivity of the composite than the smaller sized ones due to the fewer defects and contact resistance. On the other hand, some researchers proposed a template-induced CVD approach to produce 3D-foam (Fig. 17) [69, 153, 154] or 3D-sponge [158, 216] composites with remarkably high conductivity. First, nanomaterials, such as graphene, was coated on the Ni template by CVD method followed by impregnation or coating of polymeric matrices (epoxy, PMMA, PDMS). The use of polymers was to support and strengthen the nanostructure from collapsing. Then, the original template material was removed by solvents such as acetone or HCl. By controlling the flow rate of the input gas (CH_4) during the CVD, composites with different nanofiller concentrations were obtained. Jia et al. [154] showed that the precise construction of conductive networks on the template could reach up to 1000 S/m with only 0.5 wt% loading in addition to its significant improvement in flexural and fracture properties. In the meanwhile, Cui et al. [158] synthesized a novel CNT sponge via CVD process using ferrocene and dichlorobenzene as precursors. The epoxy-based composite with the proposed CNT sponge structure also demonstrated remarkably high electrical conductivity (145 S/m) and tensile strength (64%) under a low concentration (0.66 wt%). Due to the high electrical conductivity and tremendously increased surface areas, a unique application of 3D carbon nano-foam composite is EMI-shielding [145, 147, 148, 151, 158, 214]. The multiple foams reflected and scatter electromagnetic microwaves between the cell walls and nanofillers to present absorption or reflection type of shielding mechanism, which showed potential applications in electronic and aerospace areas.

Hybrid carbon nanofiller-reinforced polymer composites

Hybridization of diverse dimensional carbon nanofillers, such as CB, CNT and graphene, is recently a popular approach for attaining polymeric nanocomposites with good electrical and mechanical properties. A synergistic effect can be achieved when nanofillers with different geometrical shapes are mixed together, resulting in unique 3D interconnected structures, improved dispersion and behavior change of the crystallinity of the polymer matrices [162, 163, 168]. The zero-dimensional geometry and low surface energy enable CB to be used as a cost-effective filler for reducing the amount of expensive carbon nanotubes and graphene while construct

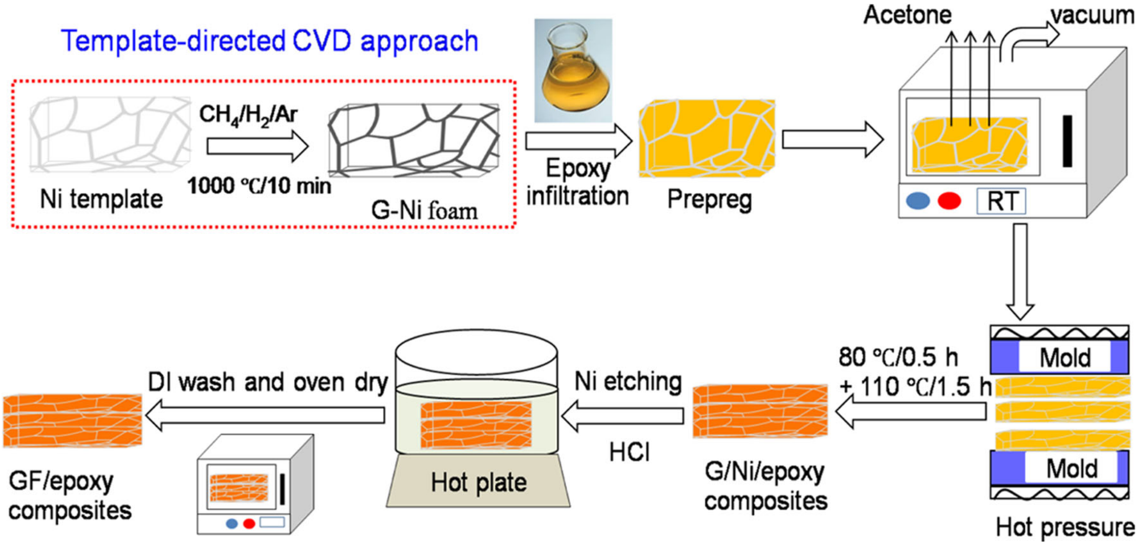

Figure 17 Template-directed CVD approach to fabricate 3D nano-foam structure. Reproduced with permission from [154]. Copyright: 2013 American Chemical Society.

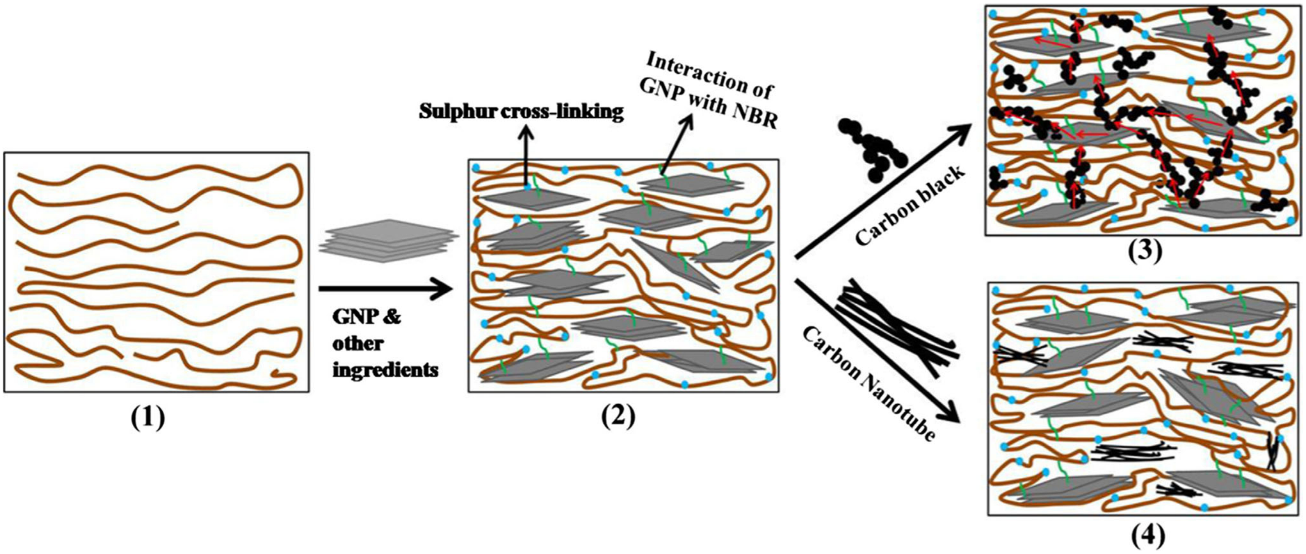

Figure 18 Schematic illustration of the GNP-CB and GNP-CNT hybrid system. Reproduced with permission from [161]. Copyright: 2017 Elsevier.

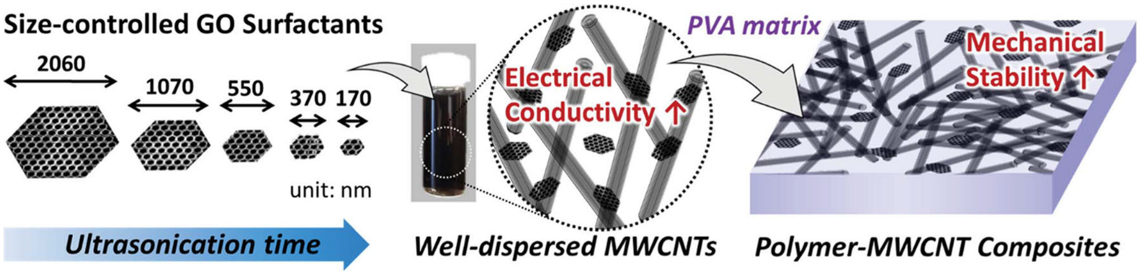

Figure 19 Representation of the size-controlled GO surfactant treated MWCNT/PVA nanocomposite [169]. Copyright Published.

segregated [160] or interlaminar linked hybrid conducted networks [159, 161, 217]. For instance, Mondal and Khastgir [161] found that by partially replacing GNPs with CB one order of magnitude electrical conductivity and 40% increase in tensile strength were achieved compared to that of the pure GNP-reinforced composite. They believed that the reason was that CB could perform as delaminating agent for aiding in the exfoliation of GNPs and meantime strengthened the graphene interlayers (Fig. 18).

Excellent material properties and similar molecular structures make CNTs and GNPs the mostly studied combinations of carbon nanofiller in polymer composites. Research showed that only by simply physically mixing the two nanofillers (untreated), synergistic effect could occur through the non-covalent π–π interaction, which helped disintegrate agglomerates and establish efficient 3D network of conduction paths [162, 163, 166–168, 170, 171]. The specific synergistic mechanism was that, in the hybrid systems, carbon nanotubes can both embed graphene nanofillers and bridge the independent graphene layers into an interconnected entire structure. However, this mechanism was not always true for considering the dispersion quality of the two materials. In other words, no synergistic effect was shown if CNTs could not disentangle into an extent to guarantee its embedding in the interlayers of graphene, which is the case demonstrated in Fig. 18. This finding pointed out the necessity of the pretreatment of the individual nanofillers before mixing them together [165, 168]. In addition, the size of the nanofiller can also significantly change the interaction behavior of the two hybrid components. An interesting study discovered that by ultrasonication GOs into a small size (170 nm), GOs could perform as an

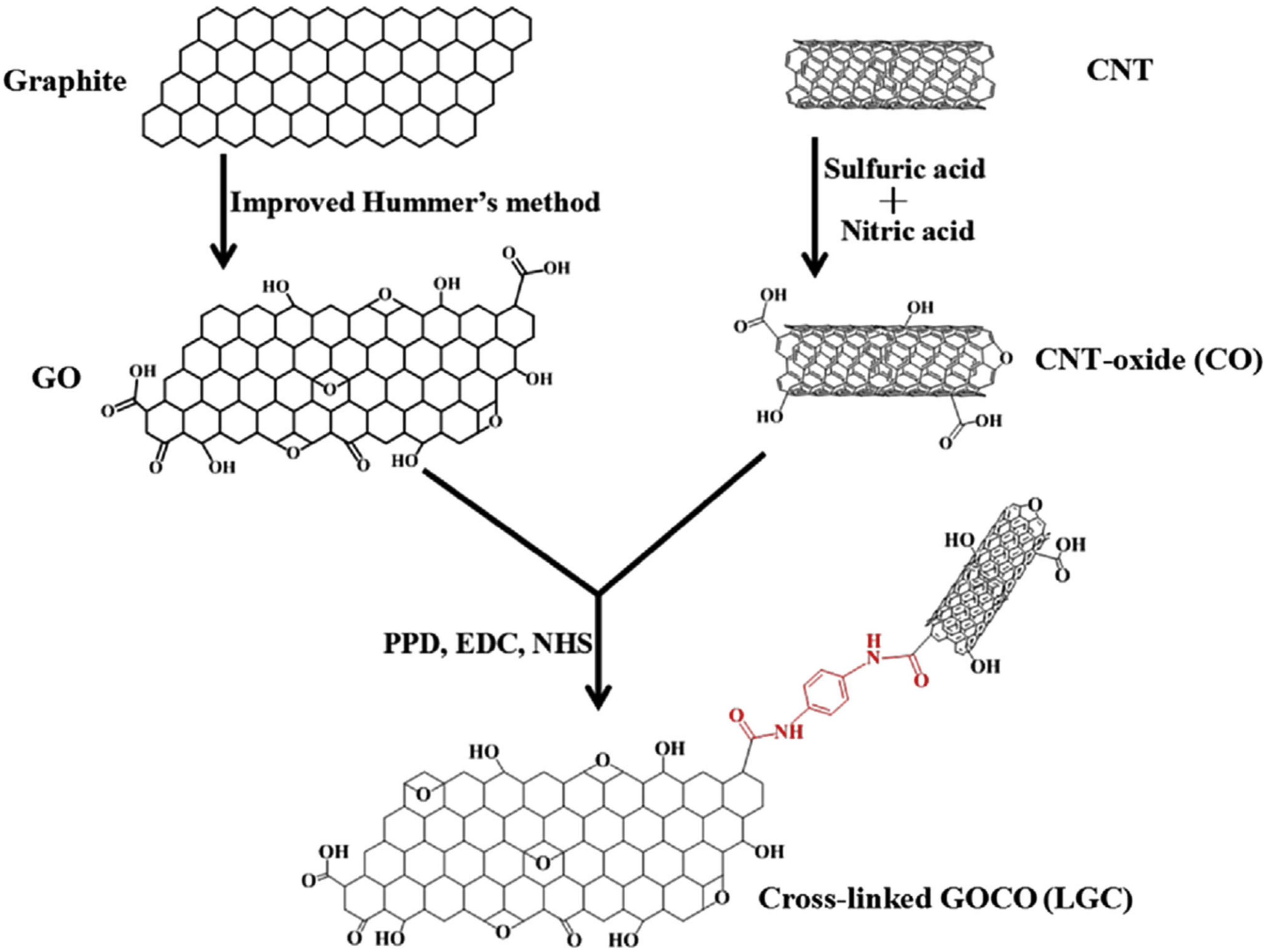

Figure 20 Synthesis process of cross-linked GO-CNT structure by amide bonding. Reproduced with permission from [164]. Copyright: 2017 Elsevier.

effective surfactant to coat CNTs (0.025:1) for fabricating a highly conductive nanocomposite with fascinatingly increased mechanical properties (Fig. 19) [169]. Covalently bonding CNTs and GNPs is another way to obtain well performed multifunctional hybrid nanocomposites [164, 218–220]. The prevalent approach was oxidization of CNTs and GNPs first and then covalently connected by a small intermediate molecular, which was usually the monomer of the polymer matrix to trigger the subsequent in situ polymerization process (Fig. 20). Reported studies showed that octadecylamine (ODA) [164, 220], *p*-phenylenediamine (PPD) [218], ethylenediamine (EDA) and aspartic aid [219], etc., could efficiently link the two carbon nanofillers by amide and hydrogen bonds to produce well-dispersed and strong interfacial interacted polymer nanocomposites.

Outlook and conclusions

This paper briefly reviewed the recent advances of the various carbon nanofiller-reinforced polymer composites with a focus on the electrical and mechanical properties. From the above review, it is shown that the nanofiller types, contents, fabrication methods and polymer matrices all greatly influence the performance of the nanocomposites, which are summarized in Tables 1, 2, 3, 4 and 5. Analyzing the maximum values in these tables, we can make a general comparison and comments on the polymeric composites with diverse dimensional carbon

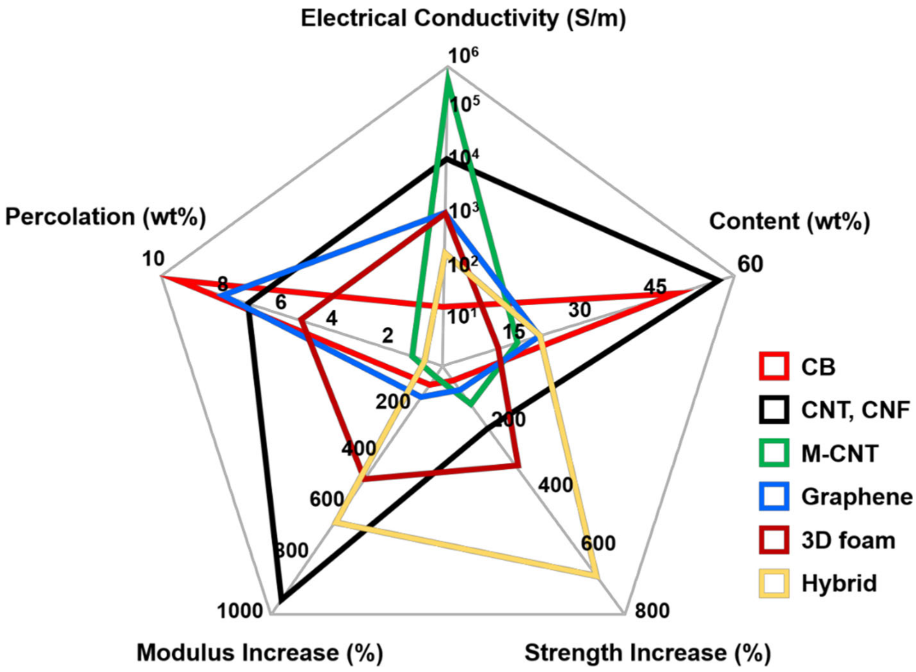

Figure 21 Summary of the electrical, mechanical, percolation content and maximum content of various dimensional carbon nanofiller-reinforced polymer composites.

nanofiller in terms of five key parameters as plotted in Fig. 21.

1. For CB-reinforced composites, the maximum electrical conductivity and mechanical properties increase only attain ~ 10^1 S/m and less than 100%, respectively, even under a big amount of filler content and high percolation threshold. This is largely due to the zero-dimensional shape of the filler, which is hard to construct highly efficient conductive and strong networks.
2. In comparison, incorporation of the one-dimensional CNTs and CNFs can lead to highly conductive polymer composites with over 10^4 S/m and significantly increased modulus (950%) due to the superb essential conductivity and stiffness of CNTs. However, the agglomeration issues of the nanofillers should be carefully tackled to prevent the relatively high percolation values in certain cases. It is noted that by applying unique fabrication method [38, 120] the maximum usage of CNT and CNF could go over 40% despite their tremendously increased viscosity.
3. Metal decorated CNTs further can increase the electrical conductivity, resulting in the best electrical conductivity (10^6 S/m) among all the nanocomposites. In addition, the metal particles on the tube surfaces can also help to reduce agglomeration, leading to significantly decreased percolation threshold and loading level. However, the mechanical properties, especially the modulus increase is not significant as compared to that of pristine CNTs-reinforced composites, which may be due to the relatively low filler loading.
4. Graphene-based polymer composites, including not only graphene, but also graphite and reduced graphene oxide cases demonstrate good electrical conductivity at around 10^3 S/m while the percolation content and mechanical properties are not satisfied as expected. The detachment of graphene layers from graphite and efficient approaches to prevent aggregates are key issues to realize the potential of graphene nanocomposites.
5. 3D carbon nano-foam-reinforced nanocomposites present a rather balanced and impressive improvements in both electrical and mechanical properties along with decreased percolation and loading level. The in situ designed conductive networks enable high conductivity regardless of dispersion situation. At the same time, these networks can also become strong skeletons via manipulating the type, amount and manufacturing of blowing agents and/or from 3D template structures through CVD method.
6. Hybrid carbon nanofiller-reinforced nanocomposites show the best material properties with

10^2 S/m conductivity and over 600% increase for both modulus and strength compared to other nanocomposites. The percolation threshold is also the smallest, i.e., less than 1 wt%. The advantage of this hybrid material is that it can combine the advantages of each individual component while their drawbacks are largely avoided, thus, providing researchers with broad freedom to design various configured structures. It is shows that the hybridization of various nanofillers for achieving high-performance multifunctional composites is a very promising route to broadening the application of polymeric nanocomposites in more diverse fields.

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Compliance with ethical standards

Conflicts of interest There are no conflicts of interest to declare.

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