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Carbon Nanotubes as Reinforcing Nanomaterials for Rubbers Used in Electronics

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Abstract

The field of electronics involves complex systems where the active and passive electronic devices are integrated on the rubber substrate, e.g., silicone (Q), which provides, through potting, a strong assembly of these devices on the circuit board. Several other rubbers are employed in the field to strengthen, insulate and seal the components of the electronic machines and instruments, and therefore protect them against damage. These rubbers are typically strengthened and toughened using carbon black (CB). However, due to its noticeable drawbacks, recent research in the field of rubber and electronics has suggested the use of carbon nanotubes (CNTs) as alternative reinforcing fillers to produce electronics rubber composites that do not only have enhanced electrical conductivity, thermal stability, electromagnetic interference (EMI) shielding, weatherability and insulation properties, but also offer outstanding stretchability, bendability and tear strength under frequent elastic deformation. These performances are similar for both single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) in both the functional and structural composites. Although SWCNTs can result in relatively better homogeneity than MWCNTs, most rubbers often constitute MWCNTs because they are relatively cheaper. The great potential of rubber-CNTs composites being extensively used in the field of electronics is explored in this chapter.

Keywords: carbon nanotubes, electronics, rubber-carbon nanotubes composites, nanoscale filler, rubber properties

1. Introduction

Various electronic machines and instruments are available worldwide, and some of these are shown in **Table 1**. The purpose of these machines and instruments is to make different aspects of human lives easier. Components such as electronic devices (i.e., integrated circuits), wires and cables are central to their make-up [1, 4]. Some of these components are either made of rubber materials or require the use of various types of rubber for their respective functions. These include rubber sheets, grommets, tubes and seals, keypads, wire and cable rubber hoses and insulators, adhesive sealants, flat washers, boots and bellows, bumpers and tips covers, sleeves, and anti-vibration rubber mounts [4–9]. Their main function is to keep the machines and instruments

Electronic machines and instruments	Sector
Stethoscope, respiration monitors, defibrillator, glucose meter, and pacemaker.	Medical
Entertainment and navigation systems, engine, transmission, and devices for safety and driver assistance.	Automotive (automobiles)
Office gadgets, home appliances, audio and videos systems, router, automated teller machine, and barcode scanners.	Consumer
Automation and motion control robotics, power converting technological instruments, hydraulics, and photo voltaic systems.	Industrial
Data logger, hygrometer, anemometer, drifter buoy, barometer, and tipping bucket rain gauge.	Meteorological and Oceanographic
Aircraft and missile launching systems, boom barrier and radars for military, cockpit controllers, and rocket launchers for space.	Defense and Aerospace
High voltage DC transmission, excitation systems, VAR compensation, static circuit breakers, fans and boiler feed pumps, and supplementary energy systems.	Utility systems

Table 1.

Electronic machines and instruments, and sectors where they are used [1–3].

performing at their optimum best by dissipating heat, as well as insulating and sealing the electronic components; thus protecting them against shock, electromagnetic interference, very high and low temperatures, gas permeation, and exposure to dust and fluid (e.g., water, chemicals, solvents, steam, moisture and oil) intrusion that may lead to damage of the machines and instruments [1, 2, 6, 7, 9–11]. Electronic machines and instruments normally operate at different types of environmental conditions, from moderate to high-stress environment. Therefore, in addition to being thermal, gas and fluid resistant, the electronics rubber materials are required to be extraordinarily durable, stretchable and resilient, and yet be easy to use [5, 7, 9, 12].

The common rubbers that are used to manufacture the electronics rubber materials include natural rubber (NR), Ethylene Propylene Diene Monomer (EPDM), silicone (Q), styrene butadiene rubber (SBR), nitrile butadiene rubber (NBR), Fluoroelastomer (FKM/FPM), isoprene rubber (IR) and neoprene [5–7, 9, 13–17]. Generally, these rubbers have excellent elasticity and deformability, but in addition to the fact that some of them are not crystallizable under high strain, their strength and modulus, especially, could not satisfy the requirements of some electronic machines and instruments, especially those that operate at frequent vibrations and high pressures [5, 9, 13, 18–20]. Therefore, it is deemed necessary to further strengthen these rubbers, typically by adding reinforcing filler into them to yield sufficiently high mechanical properties with low hysteresis loss (heat-build up). Other common properties that are improved by reinforcement include electrical, chemical, swelling and thermal properties [9, 13, 15, 16, 21, 22].

Carbon black (CB) and silica are the most conventional fillers used for the reinforcement of rubber. However, there are many emerging fillers such as carbon nanotubes (CNTs) which offer superior reinforcing effect, properties and performances at relatively lower quantities [2, 7, 23–25]. Although there is paucity in CNTs reinforced rubbers as compared to CB reinforced rubbers, several researchers [5, 8, 9, 12, 13, 15, 16, 21, 26–31] postulated that CNTs may be considered by electronic industries in the near future for the production of CNTs-based rubber composites for strengthening, insulating and sealing the components of the electronic machines and instruments. This is due to the resultant extraordinary properties and associated performances that are typically offered by such composites, which, by

definition, refers to multiphase materials that comprises of the individual or hybrid reinforcing filler and the rubber matrix or matrices [21, 24, 28, 32].

Since the discovery of CNTs, there have been several research studies aiming at understanding their structure and properties, as well as developing novel applications for them [2, 13, 21, 33]. The main attracting nature of CNTs to produce the CNTs-based rubber composites include excellent electrical conductivity, thermal stability and chemical stability, as well as the superior mechanical properties for load-bearing reinforcements in rubber composites and for structural applications [2, 22]. Rubber-CNTs composites, with strengthened stress transfer from rubber to CNTs due to their uniform dispersion in the rubber matrix and the strong rubber-CNTs interactions, are relatively lighter and flexible for easy use in the electronic machines and instruments [2, 5, 8, 9, 13, 15]. Their excellent thermal and chemical stability makes them versatile materials for improving the flame retardancy of the electronic rubber materials and for the protection of the electronic components against damage [1, 2, 15, 28, 34]. The extraordinary durability and resilience of rubber-CNTs composites makes them suitable especially for resisting wear, high pressure and vibrations; and their outstanding mechanical properties allows them to exceptionally resist abrasion, tear, high compression set and flex fatigue life that are normally due to prolonged vibrations, high pressure and compressive loads [1, 2, 28]. These properties make them more suited and applicable for use in electronics, as well as the fact that rubber-CNTs compounds are potentially cost-effective than rubber-CB compounds because of the performance of CNTs that is dominant even at smaller loaded quantities [7, 15, 35, 36]. Hence, this chapter investigated the studies that show that CNTs have great potential as the alternative reinforcing materials for rubbers used in electronics, and these rubbers are referred to as electronics rubbers in the chapter.

2. Structure and properties of carbon nanotubes

Since the discovery of carbon nanotubes (CNTs) in 1991 by Iijima using an electric arc-discharge method and transmission electron microscope (TEM), their unique atomic structure and superior properties have attracted the researchers' attention both in academia and industry [1, 22, 24, 37]. As can be seen in **Figure 1**, CNTs are typically one-dimensional quantum nanomaterials with carbon electrons in (ideally) sp^2 hybridized orbitals, and they can also be viewed as a graphene sheet that is rolled up into a nanoscale tubular form [1, 5, 28, 38]. They are commonly synthesized using visible light vaporization, arc discharge or catalytic chemical vapor deposition method, and have been categorized as the fourth allotrope of carbon, following naturally occurring types such as diamond, graphite and fullerenes [16, 24, 37]. Depending on the employed synthesis method, CNTs can be produced as individual cylinders (single-walled carbon nanotubes, SWCNTs) or as concentric tubes (multi-walled carbon nanotubes, MWCNTs), which both have the exceptionally resilient structures due to the carbon-carbon ($-C-C-$) bond and the system controlling these atomic bonds throughout the axis of the tubes [2, 39].

CNTs, due to their size and helical arrangement of graphite rings in the walls, exhibit a wide range of interesting unique properties for various potential applications [16]. They have exceptionally small diameters (several nm) and length (μ m, mm or cm) [39, 40]. Furthermore, CNTs have incredibly high aspect ratio ($\sim 10^6$) and a large surface area (~ 100 m²/g to 1200 m²/g), which often allows them to form superior interaction with the polymer matrix [22, 25, 41–43]. The strength of the sp^2 -C-C- bonds gives CNTs an extremely high tensile strength (~ 150 to 180 GPa), modulus (~ 640 GPa to 1 TPa) and elasticity, and remarkable electrical conductivity, thermal (more than 1000°C) and chemical stability [1, 3, 5, 6]. These distinctive

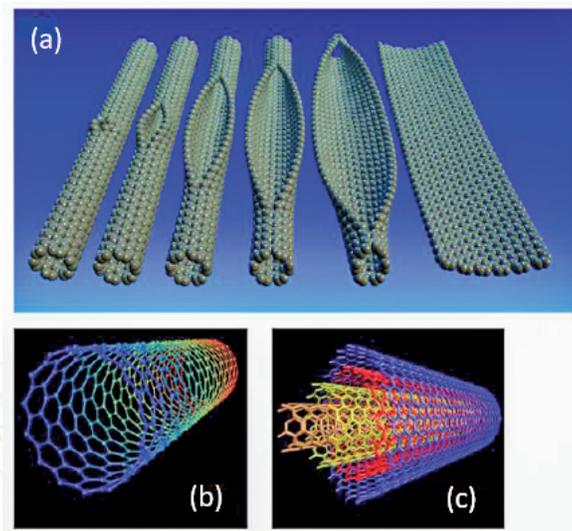


Figure 1. Schematic illustration of CNTs: (a) carbon nano-walls, (b) SWCNTs and (c) MWCNTs (modified with permission from [28]).

properties render CNTs the ultimate nanofiller materials in nanomaterial-based rubber composites for research and industrial applications. The main attractive nature of CNTs for such composites especially in the industrial application is their fracture and deformation behavior [2]. Because of CNTs, rubber-CNTs composites can withstand high loads without showing the sign of fracture, and they can do this by switching reversibly into different morphology patterns (flattened, twisted, and buckled) on strain deformation [2].

3. Carbon nanotubes and carbon black as reinforcing fillers in carbon-based rubber composites

Generally, several fillers have been used for many years as reinforcing materials to prepare the composites mostly with enhanced mechanical properties. The mechanism of the reinforcement is believed to be both chemical and physical in nature, and the surface area and structure of the filler are its primary properties [44, 45]. The greater reinforcement effect has been reported to be given by the material with relatively smaller particle size and larger surface area [45]. Carbon black (CB) is the most widely used and most effective conventional reinforcing filler in the rubber industry because it generally enhances the mechanical properties of various rubbers [8, 18, 24, 27]. However, in addition to its environmental polluting nature, the drawbacks of using CB is that it tends to cause difficult processability due to its relatively larger particle size and high bulk viscosity of most rubber compounds [18, 24]. Additionally, its high loadings (35–45 phr) in the rubber compound formulation is a requisite for its efficiency, and this normally negatively impact the compression set and hysteresis loss (heat-build up) of some vulcanized rubber products [18, 24]. Its high loadings also cause the resultant rubber products to be relatively costly [41]. Therefore, scientific and industrial fields have been focusing on partially or completely replacing CB in rubber formulations with CNTs (single-walled and multi-walled) for the production of carbon-based composites, i.e., CNTs-natural rubber and CNTs-synthetic rubber composites, with relatively excellent properties [8, 39]. The extremely small particle size, high specific surface area and aspect ratio of this new class of fillers makes them superior to CB, and makes it relatively easy for them to uniformly disperse in the rubber matrix as individual particles [8, 28, 41]. The distances between the components of CNTs and the rubber matrix are exceptionally

small and therefore, the interactions at a molecular level between CNTs and the matrix provides remarkable properties compared to conventional fillers [8].

The extraordinary properties of CNTs are the main factor that prompted a great interest in the production of CNTs-based rubber composites for a wide range of applications, including electronics [2]. The commonly used methods to prepare CNTs-based rubber composites with uniformly distributed CNTs and strong rubber-CNTs interactions include solvent/solution blending, melt blending, in-situ polymerization, latex compounding and high-shear (roll mill and internal mixer) mixing [5, 6, 40, 46–48]. The use of solvent to disperse CNTs aid in achieving good defibrillation and necessitate the dispersal of hydrophobic CNTs in the aqueous emulsion; hence surfactants, which typically suppress re-aggregation, are also often employed in melt blending, in-situ polymerization and latex compounding [5]. The high-shearing mixing method is often used for solid rubber and is favored for the industrial production of rubber-CNTs composites, including those used to manufacture electronics rubber materials, because it minimizes both the production time and costs [5, 28].

Since the applications of CNTs-based rubber composites are different, they are normally categorized into two kinds, i.e., functional composites and structural composites [1]. CNTs function differently in these two kinds of composites. In structural composites, they allow for the formation of structural rubber-CNTs material with easy processability, ultralight weight, and high tensile strength, elastic modulus, compression strength and stiffness [1, 36, 49, 50]. For rubber-CNTs functional composites, CNTs function by developing the electrical and thermal conductivity and chemical stability of these composites. Rubber-CNTs functional composites have shown outstanding heat resistance, chemical and swelling resistance, electrical conductivity, electromagnetic absorption and interference shielding, and high energy storing capability [8, 9, 15, 51–53].

4. Parameters affecting the properties of rubber-CNTs composites

CNTs tend to form bundles during growth due to strong van der Waals interactions between individual tubes [3, 39]. Therefore, this allows them to easily form microscale aggregates or agglomerates into a rubber matrix, hence reducing the expected improvements of the properties of the resulting composites. The extent of reinforcement effect of CNTs on rubber for the formation of CNTs-based rubber composites with superior properties is highly dependent on a variety of parameters, which normally influence the overall exploitation of the performance of rubber-CNTs composites in an intended application. These include CNTs fabrication method, ratio of CNTs to the amount of rubber matrix, entanglement state of CNTs in the rubber matrix, if CNTs are functionalized or not, functionalization method, matrix type, rubber viscosity, degree of CNTs wetting with rubber, dispersity and dispersion method, interfacial bonding, CNTs structural defects and composite processing method [13, 38, 54, 55]. These parameters are the main key for ensuring the formation of the effective load/stress transfer, normally monitored by Raman Spectroscopy, from the matrix to individual nanotube, which consequently support the effective processing of the formation of rubber-CNTs composites with optimum properties [1, 13, 21, 54, 56, 57]. Of all these, filler dispersion is the most popular parameter and is normally studied by examining the morphology of the composite, and this is achieved by employing transmission electron microscope (TEM) and scanning electron microscope (SEM). For instance, SEM micrograph can be seen in **Figure 2**, where the uniform distribution of CNTs particles throughout the silicone rubber (Q) matrix is shown in pictures (a), (b) and (c).

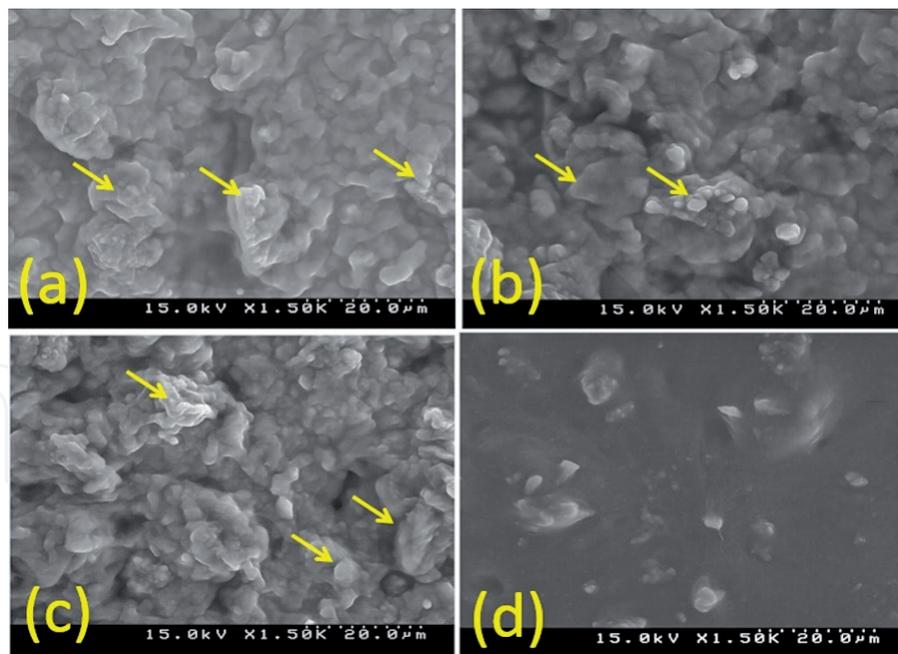


Figure 2.

SEM micrograph of Q-SWCNTs vulcanized composites: (a) Q sample of SWCNTs with 70% carbon, (b) sonicated Q sample of SWCNTs with 70% carbon, (c) Q sample of SWCNTs with 90% carbon, and (d) unfilled Q sample [58].

The parameters that define the quality of rubber-CNTs composites have also been reported to affect one another. For instance, the rubber-CNTs interaction, which generally defines the stress transfer capacity, directly affects the dispersion of CNTs in the matrix [13, 28, 59]. Also, the dispersion of CNTs in the rubber matrix and the interaction of CNTs with the matrix are highly influenced by the functionalization (surface modification) of pristine CNTs which is typically achieved by physical (non-covalent) or chemical (covalent) bonding of organic or inorganic moieties to the tubular structure of CNTs [3, 13, 28, 57, 59]. This surface modification of pristine CNTs typically result to modulation of CNTs physicochemical properties, therefore increasing their ease of dispersion and interaction, as well as processability, among different types of rubbers [38, 57]. However, even though it might possibly lead to weaker rubber-CNTs interaction, non-covalent functionalization method is more preferred than the covalent method for the production of composites as the latter tend to cause structural defects to the tubes by disturbing the π system of the graphene sheets and therefore resulting in shortened CNTs length and hence, inferior properties of the CNTs [3, 27, 60]. Optimization of the ratio of CNTs to the amount of the rubber matrix is also necessary for overcoming the inability to fully explore the properties and performance of CNTs in rubber-CNTs composites for any intended application.

5. Effect of CNTs (SWCNTs and MWCNTs) on various properties of rubber-carbon nanotubes composites for electronics

The main targets under consideration for the application of rubber-CNTs composites in electronics is to manufacture rubber materials that are extremely resistant to different temperature conditions, durable, abrasion resistant, chemical and swelling resistant, thermal resistant, high stretchable and thermo-conductive, and offer proper insulation and sealing [2, 5, 12, 13, 61]. CNTs permits the rubber-CNTs composites to maintain their mechanical strength at temperatures as high as 1200°C, therefore electronics rubber materials that comprise of CNTs would

be very useful for electronics operating at high temperatures [1, 5]. Additionally, researchers [28, 62, 63] believe that rubber-CNTs composites will soon be employed as layers and coatings design to dissipate heat, and as materials that enhances flame retardancy because most rubbers are less thermally stable than CNTs. The very high chemical stability of CNTs makes the rubber-CNTs composites resistant to various solvents, oils, hydrocarbon fuels and acids or alkalis, and therefore far superior to metals as regards to corrosion resistance, and would be excellent for use in covering the electrochemical sensors, power devices and other components of various electronic machines and instruments [1, 5, 6, 28, 64]. Similarly, the high swelling stability of the rubber-CNTs composites makes them resistant to water, steam and moisture and therefore has a great potential for the manufacturing of the rubber materials that would protect the electronics materials like temperature/humidity sensors and conductive electrodes/wires [1, 6, 64]. Given the obvious possibility that water, coldness and heat, are present in the environment in which sealing and protecting rubber materials are used, rubber-CNTs composite materials would thus be best materials for electronics as they can resist hydrolysis, which may lead to degradation of rubber. This has been additionally proven by other researchers [5], where they subjected the rubber-CNTs material, with 1 wt.% CNTs content, into an environment of 280°C temperature and 6.3 MPa pressure for 3 h, and observed no change in hardness and tensile properties of the material, meaning that CNTs improve hydro-thermal resistance even at high pressure. Rubber-CNTs composites have outstanding electrical and thermal conductivity, of which the latter far exceed 400 W/Mk thermal conductivity of copper and 2200 W/Mk of diamond [5]. According to Ata [5] and Dai et al. [13], the main advantage of using rubber-CNTs composites to make stretchable electroconductive and thermo-conductive materials for electronics is that the electrical and thermal conductivity of the composite remains unaffected when the material stretches during service because CNTs form unidimensional particles in the rubber matrix, as opposed to conventional fillers. In comparison to rubber-CNTs composites, a reduction of the conductivity of the composites of conventional fillers, which are typically composites of zero-dimensional (0D) particles, with stretching, is due to the loss of contact between filler particles, as can be seen in **Figure 3** [2, 5, 50]. These conductivity properties have therefore led to a suggestion that rubber-CNTs composites can be used as wiring materials for stretchable and wearable electronic devices and instruments [5, 12, 36, 65, 66]. This has been supported by other authors [61], where they have made stretchable CNTs-based FKM conducting materials that have the potential for application in electronics such as stretchable sensors, stretchable light-emitting diodes (LEDs), and human motion monitoring.

Electronic components, like electronic devices (e.g., electronic circuits), wires and cables, in the electronic machines and instruments are subjected to damage during service. Hence, for rubber materials that typically seal, insulate and therefore protect these components, the improvement of mechanical properties, especially tensile, modulus, durability, flexibility and resilience, is a necessity. Felhös et al. [67] have used varying amounts of CNTs on hydrogenated nitrile butadiene rubber (HNBR), and measured the mechanical properties, including sliding and rolling friction, observing improved properties as the filler content was increased. They also observed that CNTs performance was better than that of silica in drying sliding and rolling. As far as the various required properties for rubber materials used in electronics are concerned, several other studies, on the reinforcement of electronics rubbers by CNTs, are shown in **Table 2**, which also shows the common applications of the stated CNTs-based rubber composites. While most of these studies are more based on enhancement of various properties of electronics rubbers, others [6, 17, 18, 24, 28, 32, 35, 41, 72, 83, 84] also compared the

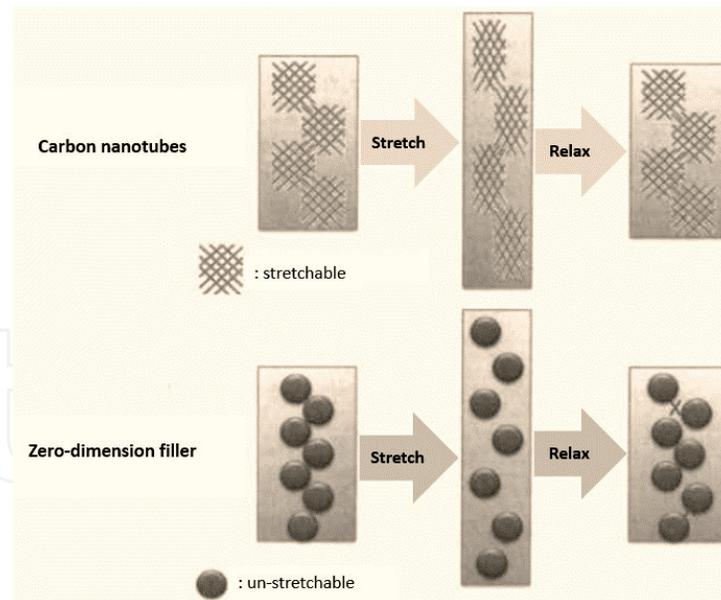


Figure 3.
Network structure in rubber composites of different fillers [5].

reinforcement effect of CNTs to that of CB, and showed that CNTs' performance is dominant even at relatively very low quantities (at or below 1 wt.%), without polluting the working environment.

It is well known that CNTs have a tubular structure of carbon atom sheets with a thickness scaled in less than few nanometers, and depending on the number of carbon atom sheets, they are often simply classified as single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). The performance of SWCNTs and MWCNTs seems to be similar in both the functional and structural composites, even though it is believed that SWCNTs, in comparison with MWCNTs, are relatively more suited for the production of electronics rubber-CNTs composites since they are single-dimensional systems and therefore can easily and uniformly be dispersed in the rubber matrix and are unlikely to cause high stiffness, which might lead to high heat build-up [22, 40, 85, 86]. Nonetheless, on account of their extremely high moduli, both the SWCNTs and MWCNTs are considered fillers that provide much higher reinforcement effects than conventional fillers; hence their use in the enhancement of the matrix of most rubbers, including those that are used in the electronics [39]. It has been suggested by several researchers [6, 18, 24, 28, 32, 35, 41, 84] that the manufacturing and maintenance cost of the electronics rubber materials made of CNTs would be relatively better than those made of CB because CNTs, in addition to the fact that they are effective at very low added concentrations, are also becoming easier to fabricate and therefore cheaper to buy [5, 28, 39, 87]. As far as the cost of rubber-CNTs composites for electronics is concerned, composites made of MWCNTs would be much cheaper in comparison to those made of SWCNTs since the fabrication cost of the former is relatively cheaper than that of the latter. Nonetheless, costs are not the only determining factor on the choice of CNTs, rather the resultant properties of rubber-CNTs composites are also imperative. Therefore, it is important to understand the influence of the different types of CNTs on the properties of different types of rubbers.

5.1 Natural rubber-CNTs and isoprene rubber-CNTs composites

Natural rubber is known for its good performance in both the electronics gaskets and in insulating the electrical wires and cables because of its good elastic modulus,

Rubber-CNTs composites	Effect of CNTs on the rubber properties	Applications in the electronics sector	References
Natural rubber-CNTs	Increased ultimate tensile strength (UTS), modulus (M), tear strength, hardness and dynamic mechanical properties; decreased elongation at break (ϵ_b); and enhanced thermal stability, chemical stability, electrical (direct and alternating current) conductivity and abrasion resistance.	Electronics gaskets, insulating electrical wires and deep-sea cables, as well as electronics that are exposed to high vibrations and pressure such as washing machines, engine seals and belts.	[7, 19, 24, 33, 35, 46, 47, 51, 68]
Ethylene Propylene Diene Monomer rubber-CNTs	Increased UTS, stiffness and compression strength; decreased ϵ_b ; enhanced flexibility, deformability, electrical and thermal conductivity; and improved strain sensitivity and electromagnetic shielding.	Wires, cables, power transformers, distribution automation devices, streetlights, engines and cameras.	[6, 69–71]
Silicone rubber-CNTs	Increased UTS, elastic M and hardness; decreased ϵ_b ; improved thermal stability and conductivity; enhanced electrical conductivity, flexibility, and stretch-ability; and improved strain sensitivity and sensing linearity for pressure.	Cables, potting and encapsulation, particularly as sealing and insulating materials for energy transmission and distribution, electronic utility systems, high frequency communications, wearable electronics, health performance monitoring and automotive electronics.	[5, 15, 16, 50, 72–75]
Styrene butadiene rubber-CNTs	Increased UTS, stiffness, M, tear strength and hardness; decreased ϵ_b ; improved thermal stability and abrasion resistance; and reduced thermogenesis and flammability.	Pressure sensors, capacitive sensors and solar cells.	[24, 41, 76–78]
Nitrile butadiene rubber-CNTs	Increased UTS, fracture strength, M, hardness, toughness strength, abrasion resistance and ϵ_b ; and improved thermal stability and electrical conductivity.	Sealing for all kinds of appliances, machine tapes and power transmission belts.	[6, 18, 79–82]
Fluoroelastomer-CNTs	Increased tensile properties and stretch-ability; reduced swelling; enhanced strain/pressure sensitivity and electrical conductivity.	Electric wire and power cable coverings, Electronic circuits and dynamic pressure monitoring in electronics.	[6, 61]

Table 2.
Studies about the effect of CNTs on various properties of some rubbers used in electronics.

fracture energy and dielectric strength; therefore, it has been used in portable electronics and distributed sensors [88, 89]. But this is typically only possible when it is reinforced. A shift to CNTs from traditional fillers like CB has allowed the production of NR compounds with enhanced properties, especially because it generally fails at higher temperatures (above 100°C) and it has low oil and fuel resistance (swells in

oils and fuels) [90]. Through the high-shear mixing method, Azam et al. [51] have prepared the NR-SWCNTs composites to study the effect of SWCNTs on the tensile, hardness and thermal properties of NR. They observed a reduction in the ultimate tensile strength (UTS) and elongation at break (ϵ_b) and an increase in tensile moduli (M), hardness and thermal property. The unusual decrease in the UTS of NR with increasing SWCNTs content was suspected to have been caused by the agglomeration of SWCNTs particles in the NR matrix and/or the physical contact between adjacent agglomerates. Additionally, the obtained increase in M was reported to be due to an increase in crosslink density, which resulted from increased swelling resistance of the resultant composites and good distribution of SWCNTs into the NR matrix [51]. The uniform distribution of SWCNTs resulted to limited chain movement of NR during deformation, and therefore resulted in high M, which consequently increased hardness of the rubber compounds [51]. The obtained increase in thermal properties was attributed to physical adsorption and good NR-SWCNTs chemical interaction [51]. Gumedde et al. [47] used an internal mixer to prepare the NR-SWCNTs composites, but also passed the compounds through the two-roll mixing mill, which could be a reason why they observed an increase in UTS with an increase in SWCNTs content up to 0.1 wt.%. Unlike authors [47, 51], Anoop et al. [46] prepared the NR-SWCNTs composites through the latex compounding method, also employing the surfactant to improve the dispersion of SWCNTs in the NR matrix, and observed similar results to the authors [47, 51], as far as the tensile properties are concerned, and an increase in tear strength and electrical conductivity, while thermal properties remained unchanged after adding SWCNTs. These findings were attributed to good interfacial bonding between NR and SWCNTs [46]. Other researchers [19, 24, 33, 68] used MWCNTs instead of SWCNTs to prepare the NR-CNTs composites, and these studies have shown that MWCNTs are as effective as the SWCNTs for the enhancement of NR properties. For instance, Jose et al. [33] used a two-roll mixing mill to prepare NR-MWCNTs composites, and found that MWCNTs were uniformly dispersed into the NR matrix (see **Figure 4**), which resulted to enhanced thermal, mechanical and electrical conductivity properties of NR.

Isoprene rubber (IR) has applications that are similar to those of NR; e.g., it is typically used to manufacture the deep-sea cables insulating materials [90]. This is because IR has good electromagnetic interference (EMI) shielding property, which is important for rubber application in the electronics sector, and has been found to be proportional to electrical conductivity properties, i.e., the higher the electrical

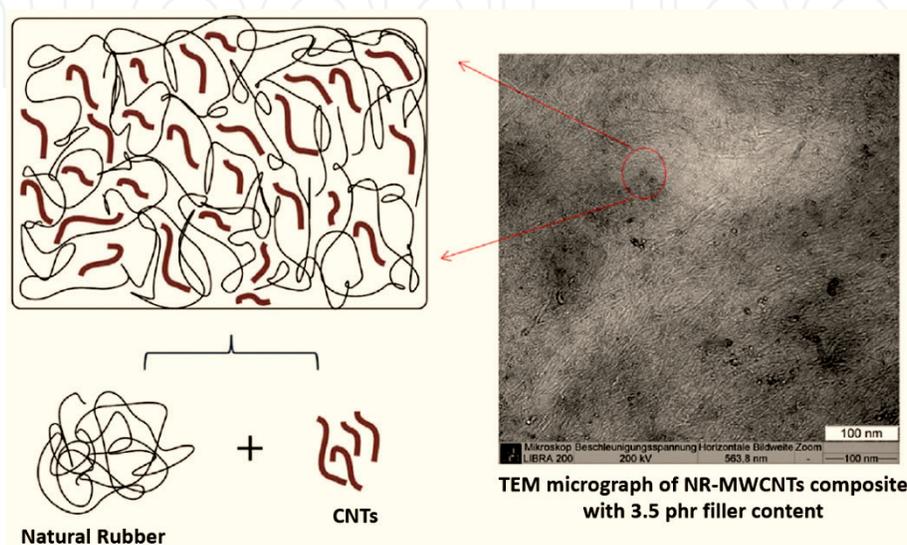


Figure 4. Schematic representation of uniformly dispersed MWCNTs into NR matrix [33].

conductivity values, the higher the EMI shielding effectiveness [17]. IR is also employed in manufacturing the anti-vibration mounts, drive couplings and bearings; therefore, it is the best rubber especially for electronics that are exposed to high vibrations and pressure, including washing machines, engine seals and belts, as well as machines and instruments that supply power through mechanical forces [17, 90]. Wang et al. [17] have shown that the electronics rubber materials that are made of IR and CNTs exhibit high flexibility, and outstanding mechanical, electrical conductivity and EMI shielding properties, and this is mainly because CNTs are one-dimensional (1D) materials in comparison to the traditional filler (i.e., CB) that is zero-dimensional. They explained that the CNTs properties were successfully explored because of the excellent uniform dispersion of these nanomaterials into the IR matrix, and the SEM micrograph, including those of CB, can be seen in **Figure 5**. While CNTs exhibited an outstanding uniform dispersion into the IR matrix even at high content (20 wt.%), some small spherical or cluster-like aggregations for CB-IR composites were observed.

5.2 Ethylene propylene diene monomer rubber-CNTs composites

Ethylene Propylene Diene Monomer (EPDM) rubber is well-known for its outdoor applications. Considering the fact that it is the most water-resistant rubber and capable of resisting failure (abrasion, tear and degradation) during harsh weather conditions, i.e., ozone, UV ray, low-high temperature, steam and flame exposure, it is regarded as the best rubber for making seals, hoses, isolators, gaskets, roll covers, tubes, wires and cables especially for outdoor electronics, including power transformers, distribution automation devices, street lights, engines and cameras [90]. CNTs have been shown to significantly further improve the properties of EPDM for potential electronics applications. Bizhani et al. [69] prepared the EPDM-MWCNTs composite foams, as illustrated by **Figure 6**, to make an industrially scalable lightweight rubber material. Based on the cryofracture surfaces of the EPDM-MWCNTs composites that were visualized using SEM, as can be seen in **Figure 7**, to study the nature of MWCNTs dispersion in the EPDM matrix, the authors claimed to have successfully achieved EPDM-MWCNTs composite foams with a good interfacial interaction.

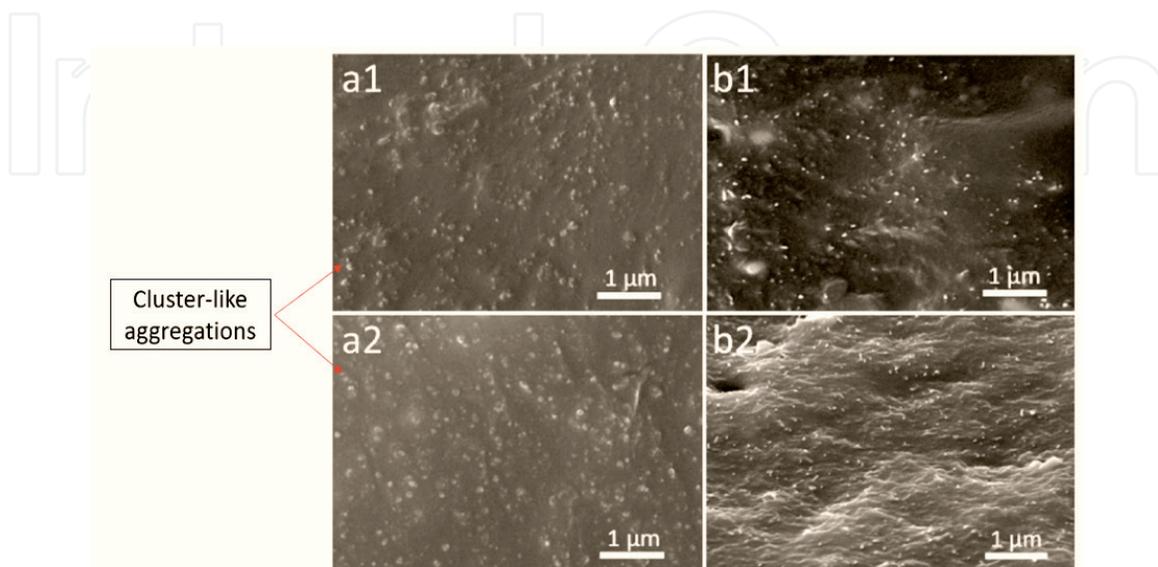


Figure 5. SEM micrograph of IR-based composites: (a1) IR-CB composite with 10 wt.% CB content, (b1) IR-CNTs composite with 10 wt.% CNTs content, (a2) IR-CB composite with 20 wt.% CB content and (b2) IR-CNTs composite with 20 wt.% CNTs content [17].

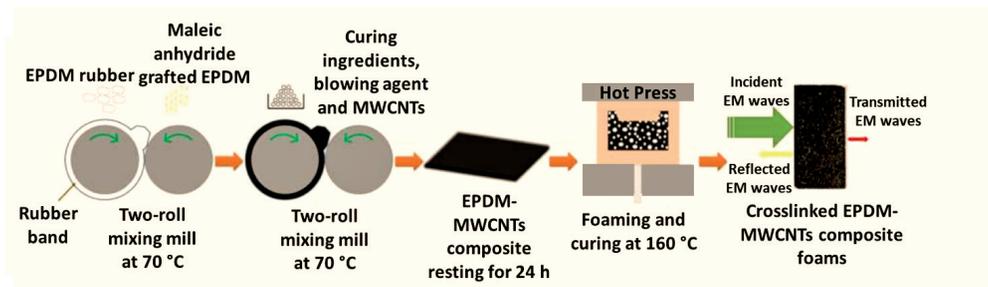


Figure 6. Preparation of vulcanized EPDM-MWCNTs composite foams [69].

Due to the development of a good three-dimensional (3D) interconnected network between MWCNTs and EPDM matrix, the authors [69] found that MWCNTs enhanced thermal conductivity (up to 0.2 W/m·K), electrical conductivity (up to 2.7×10^{-4} S/cm) and EMI shielding efficiency (up to 45 dB) properties of the EPDM-MWCNTs foams, and that they do not significantly deteriorate with continuous deformation through bending, as can be seen in **Figure 8**. These properties, as well as observed high flexibility and lightweight wave absorber capability, of the EPDM-MWCNTs composite foams indicate that these foams may be best suited for several applications, including lightweight portable devices like cell phones.

Researchers [6, 70, 71] have also used high-shear mixing method to prepare the EPDM-MWCNTs composites and found that MWCNTs significantly increased the UTS and M, while reducing the ϵ_b of EPDM, due to both the good uniform distribution of MWCNTs within the EPDM matrix and the effective EPDM-MWCNTs bonding. Chougule et al. [6] explained that functional groups that are randomly orientated on the MWCNTs surface typically impacts both the level and type of interfacial bonding between MWCNTs and the EPDM matrix. MWCNTs were found to cause the swelling behavior of EPDM to decrease while increasing its electrical conductivity [6, 70]. Additionally, other authors [70] obtained the strain response that showed piezoresistivity behavior under deformation, and this indicates that the EPDM-MWCNTs composite materials have a great potential for being used as flexible strain-sensitive materials. This has also been shown by Haj-Ali et al. [91].

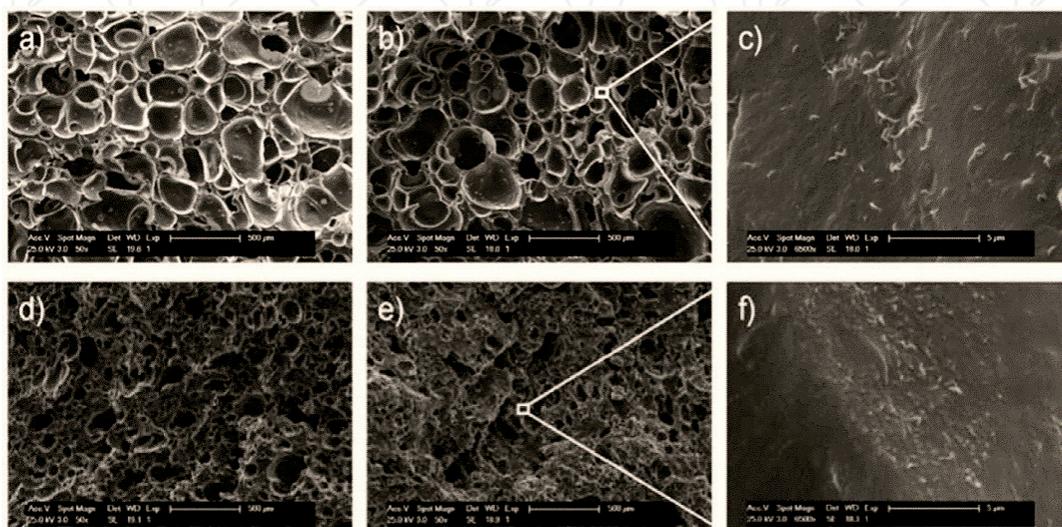


Figure 7. SEM micrograph of EPDM-MWCNTs composite foams: (a) with 0, (b) with 2, (d) with 6 and (e) with 10 phr MWCNTs content. (c) and (f) are micrograph of filler within the morphology [69].

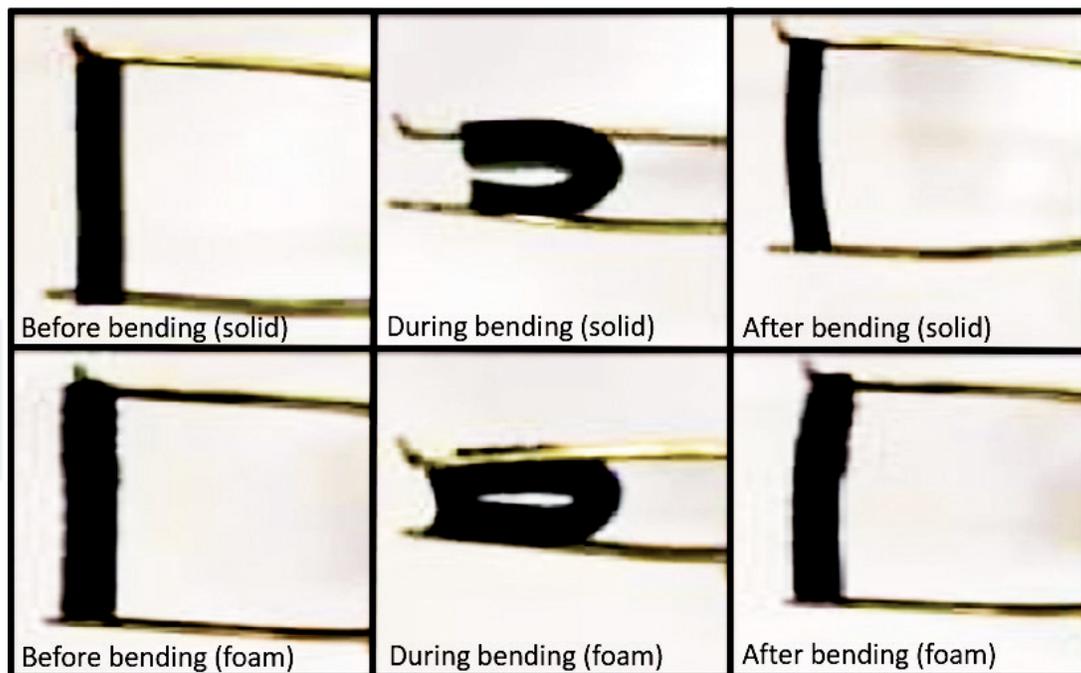


Figure 8.
Vulcanized solid and foamed EPDM-MWCNTs composites before and after bending [69].

5.3 Silicone rubber-CNTs composites

Silicone rubber (Q) is considered an inorganic–organic hybrid polymer because of the inorganic silicon–oxygen main chain with two organic groups bonded to each silicon center. As can be seen in **Table 3**, silicone rubbers can be divided based on their pendant group structure, which function to improve the natural properties of Q. Its major application is in the electronics sector mainly because of its non-toxicity, aging resistance at high temperatures, good flame resistance, chemical stability, electrical insulating and weatherability [90, 92].

CNTs are typically incorporated into Q to mainly enhance the mechanical properties, surface hydrophobicity, thermal and electrical conductivity of the resulting Q-CNTs composites. These composites, particularly the ones with liquid Q, have been reported to be very useful in high-voltage indoor and outdoor systems, and CNTs function by extending the service life of Q in such applications [92, 93]. Since Q-CNTs composites are heat, fire and chemically resistant with excellent electrical conductivity and weatherability, they can ideally be employed in the cable, potting and encapsulation sector, particularly as sealing and insulating materials for energy transmission and distribution, electronic utility systems, high frequency communications, wearable electronics, soft robotics, health and sport performance monitoring, oil drilling and automotive electronics [74, 75, 92–95]. Additionally, silicon-CNTs micropatterns can be fabricated and used in biomedical and chemical sensors, tissue engineering, drug screening, and optical devices [92]. Yanagizawa et al. [96] have prepared Q-CNTs composites and studied the effect of these nanomaterials on their water repellency. The authors observed an increase in contact angle on incorporation of just 1 wt.% of CNTs content, and that CNTs significantly improved repellency. This study aimed mainly at making the CNTs-based rubber roofing materials that are resistant to snow build-up in regions of high snowfall [5]. Li et al. [15] and Bannych et al. [75] observed a rapid increase in electrical conductivity of the Q-MWCNTs composites, after which it slowly changed when the MWCNTs content was above 1 wt.%. They reported that these trends are due to MWCNTs having a large aspect ratio, and therefore a small quantity in the rubber matrix can form an

Type	Pendant group	Chemical formula
MQ	Methyl	CH ₃
PMQ	Phenyl	C ₆ H ₅
VMQ	Vinyl	CH ₂ =CH
PVMQ	vinyl, phenyl	CH ₂ =CH, C ₆ H ₅
FMQ	Trifluoropropyl	CF ₃ CH ₂ CH ₂
FMVQ	Vinyl, trifluoropropyl	CH ₂ =CH, CF ₃ CH ₂ CH ₂

Table 3.
Different types of silicone rubbers [90].

effective effectual conductive path. Typically, the electrical conductivity of the insulator is less than 10^{-8} S/m while that of the conductor is approximately 10^5 S/m [15, 97]. Hence, Q-MWCNTs composites are suitable to be used as semiconductor materials [15]. Based on this suggestion, the authors [15] also conducted the Seebeck coefficient test and found that the Seebeck coefficient of the Q-MWCNTs slightly decreases with an increase in MWCNTs content, and it decreases with an increase in electrical conductivity (see **Figure 9**). The thermal conductivity was found to increase with an increase in MWCNTs content. Although some aggregates of MWCNTs are seen within the Q matrix in the SEM micrograph of the prepared Q-MWCNTs composite (see **Figure 10**), an increase in electrical and thermal conductivity has been attributed to uniform distribution of MWCNTs in Q matrix and the good interfacial bonding between MWCNTs and Q matrix [15]. Bright filament-like substances that appear on this SEM micrograph are due to MWCNTs.

Kim et al. [50] successfully prepared, *via* melt mixing method, a highly stretchable and conductive Q-SWCNTs gel composite as illustrated in **Figure 11**. The authors firstly produced the SWCNTs gels by grounding, using a mortar and pestle,

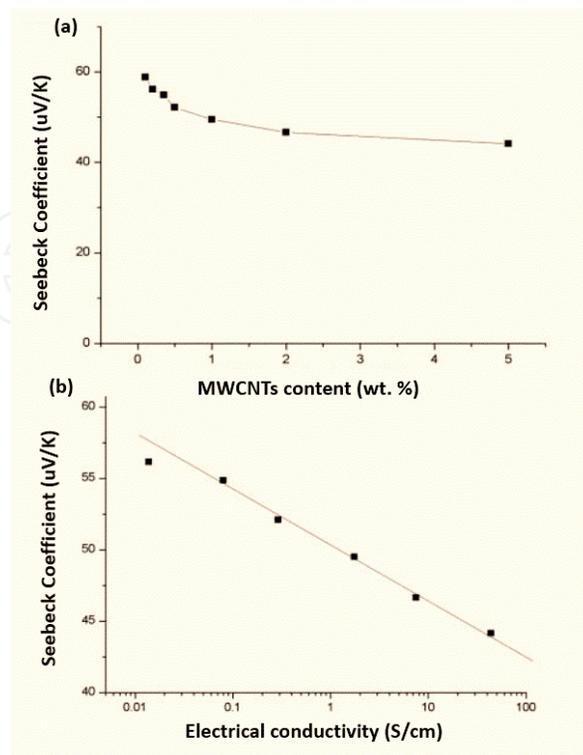


Figure 9.
Seebeck coefficient results: (a) dependent on the MWCNTs content and (b) dependent on electrical conductivity [15].

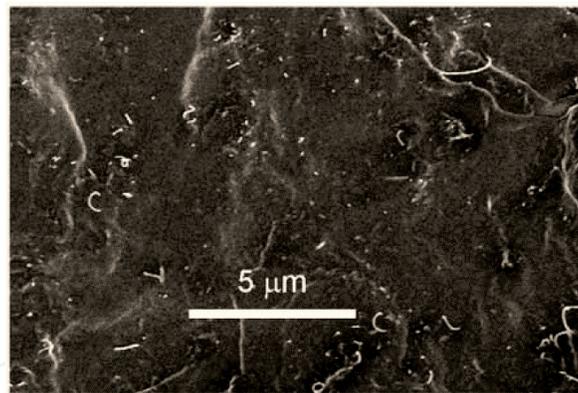


Figure 10.
SEM micrograph of Q-MWCNTs composite with 1 wt.% MWCNTs content [15].

the SWCNTs with a room temperature ionic liquid (IL) called 1-butyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide. Using an ultrasonic bath, gels were immersed in a solvent (toluene) for about 1 h to produce a solution of about 1 mg/ml concentration, after which it was homogenized with Q by stirring for about 3 h. The electrodes were made by spraying the Q-SWCNTs-IL solution onto acrylic elastomeric substrates through a contact mask and drying the samples on a hot plate for 3 h and in an ambient temperature vacuum for 24 h. The Q-SWCNTs composite electrodes were also treated with nitric acid by placing them for 30 min in a saturated acid vapor environment of 70°C, after which they were subjected to a vacuum oven at 25°C for 24 h.

The uniform dispersion of conducting SWCNTs within the Q matrix was shown by the microstructural analyses presented by **Figure 12**. This led to obtaining a composite material that is stretchable for three times its length, while maintaining its high electrical conductivity (18 S/cm) even after prolonged and continuous deformation. Hence, such materials can be best suited for wearable and stretchable conductors and strain sensors that need a constant conductivity when an intense deformation is applied [36, 50, 98].

Katihabwa et al. [73] and Shang et al. [72] also studied the effect of CNTs, particularly the multi-walled type, on the mechanical, thermal and electrical properties of the Q. Although different composite preparation methods were employed, the authors achieved a uniform dispersion of MWCNTs into the Q matrix which consequently led to the enhancement of the studied properties with an increase in the MWCNTs content in the composites. Shang et al. [72] also studied the relative resistance change (R/R_0) of the Q-MWCNTs composites when applied to elastic

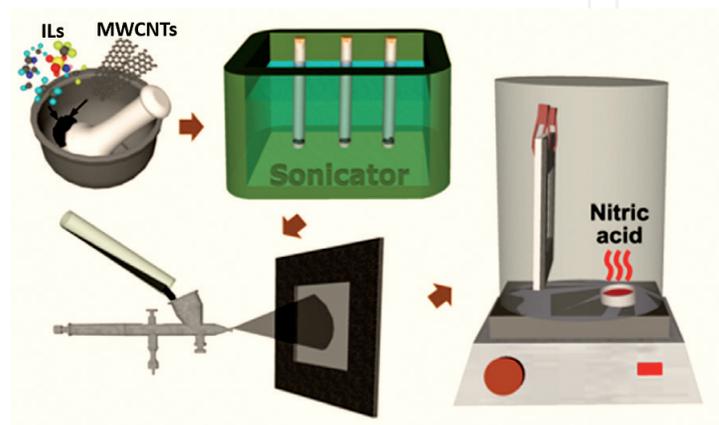


Figure 11.
Schematic representation of the preparation of Q-SWCNTs composite electrodes [50].

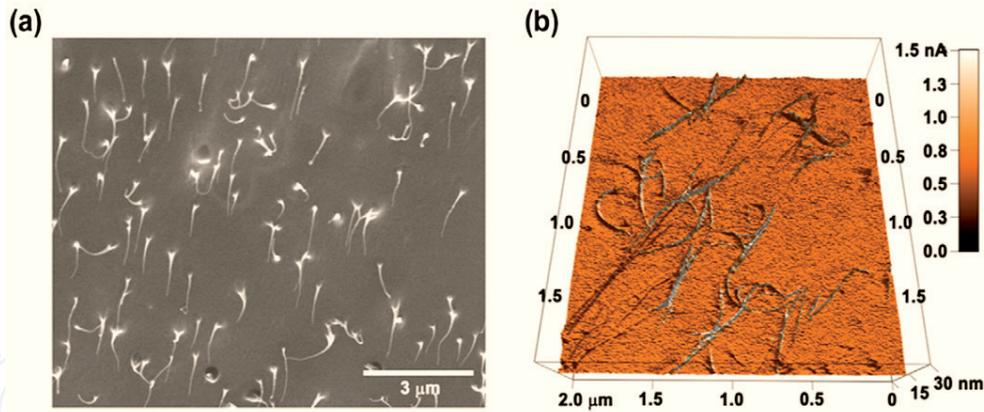


Figure 12.

Microstructural analyses of fractured regions of Q-SWCNTs composite sample with 4 wt.% SWCNTs content: (a) SEM micrograph and (b) atomic force microscope (AFM) micrograph (Amplitude represented in height difference, and current represented in color gradient) [50].

deformation (bending and twisting) of different angles. It can be seen in **Figure 13** that as the MWCNTs content reached 8 wt.% (and higher), the R/R_0 change values were smaller, meaning that there was a firm and continuous MWCNTs conducting network within the rubber matrix [72]. Hence, the prepared Q-MWCNTs composites have a great potential in the field of conductive elastomer or pressure sensors [72]. The uniform distribution of MWCNTs within Q matrix was achieved regardless of the filler content, as can be seen in **Figure 14(a)-(d)**, and this was reported to be due to the employed compatibilizer, i.e., chitosan salt, that increased the interactions between MWCNTs and Q matrix [72].

5.4 Styrene butadiene rubber-CNTs composites

Styrene butadiene rubber (SBR) has generally been employed in electronics sector because of its good elasticity and resistance to radiation, abrasion, aging, weather and ozone. However, its properties are strongly reliant on the reinforcements [90].

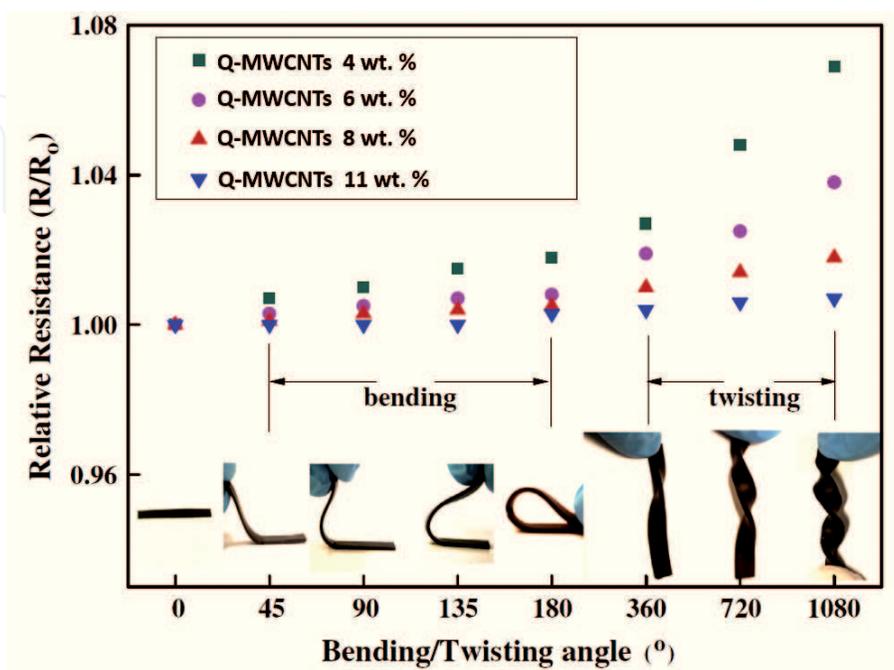


Figure 13.

Relative resistance change of the Q-MWCNTs composites at different deformation angles [72].

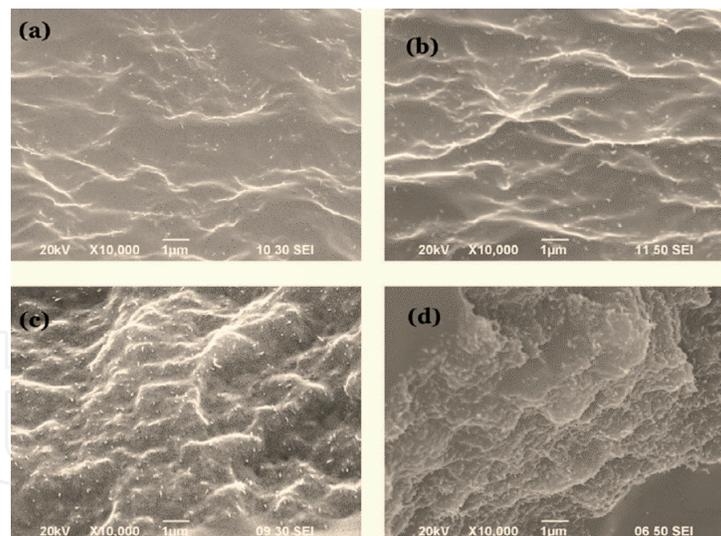


Figure 14. SEM micrograph of Q-MWCNTs composites: (a) with 4, (b) with 6, (c) with 8 and (d) with 11 wt.% MWCNTs content [72].

Researchers [24, 41, 76, 77, 83] have reinforced the NR-SBR blend, and the results showed that MWCNTs increased the UTS, M, storage modulus, and thermal stability of the blend. This could be due to good dispersion of MWCNTs into the blend and a strong interaction between MWCNTs and the matrices of both rubbers, indicating that MWCNTs have good affinity for both NR and SBR. Gao et al. [24] also reported an improvement of the abrasion resistance of the NR-SBR (80–20 phr) blend with 5 phr MWCNTs content, which was due to the synergistic effect of MWCNTs. The authors [24] showed, by SEM and TEM (*see* **Figure 15(a)** and **(b)**), that MWCNTs can come into contact with each other and that they were uniformly distributed within NR-SBR blend matrix; meaning that a good interface cohesion between MWCNTs and the blend matrix was successfully achieved, which consequently led to obtained properties. Additionally, it was reported that, based on TEM micrograph in **Figure 15(b)**, MWCNTs can form the bridges between the CB aggregations if the CNTs/CB hybrid filler is used for rubber reinforcement [24].

Rather than blending rubbers, Liu et al. [78] mixed a reduced graphene oxide (rGO) with MWCNTs to form rGO-MWCNTs hybrid filler, which was used to prepare SBR-rGO-MWCNTs composites, as illustrated in **Figure 16**. According to the authors [78], the employed hydrothermal step facilitated the prevention of restacking of graphene sheets and agglomeration of MWCNTs, and therefore

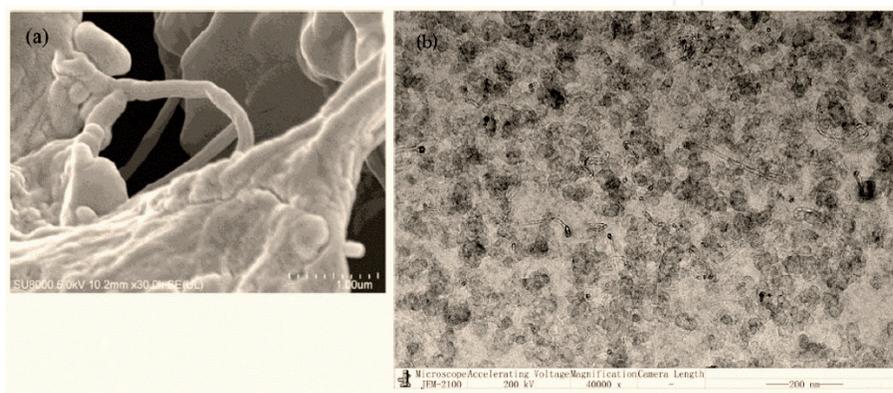


Figure 15. Electron microscope micrographs: (a) SEM image of NR-SBR-MWCNTs composite with 5 phr filler content and (b) TEM image of NR-SBR-MWCNTs-CB composite with 5 phr MWCNTs and 27.5 phr CB content [24].

obtaining a significant increase in electrical conductivity and thermal stability of the SBR-rGO-MWCNTs composites with an increase in the rGO-MWCNTs hybrid filler content. The distribution of rGO-MWCNTs hybrid filler within SBR matrix can be seen in **Figure 17**. MWCNTs content of 10.4 wt.% gave about 3.62 S/cm of electrical conductivity of the composite, of which this was 14 orders of magnitude higher than that of unreinforced SBR. Additionally, it was reported that SBR-rGO-MWCNTs composites can retain high electrical conductivity mostly under low tensile strain. Due to the one- and two-dimensional interconnected network, formed through MWCNTs bonding with rGO sheets, SBR-rGO-MWCNTs composites can be tailored for electronics that require highly conductive and stretchable rubber materials [78].

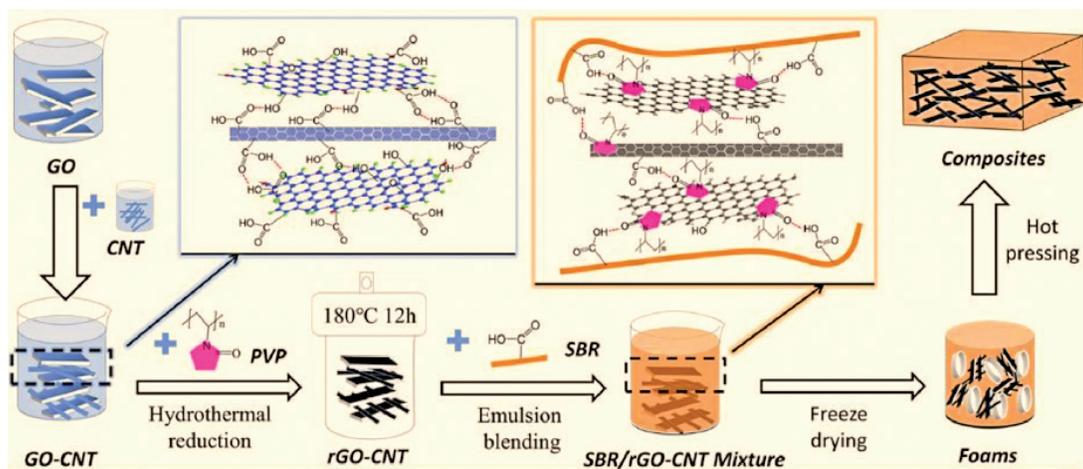


Figure 16. Fabrication of rGO-MWCNTs hybrid and SBR-rGO-MWCNTs composites [78].

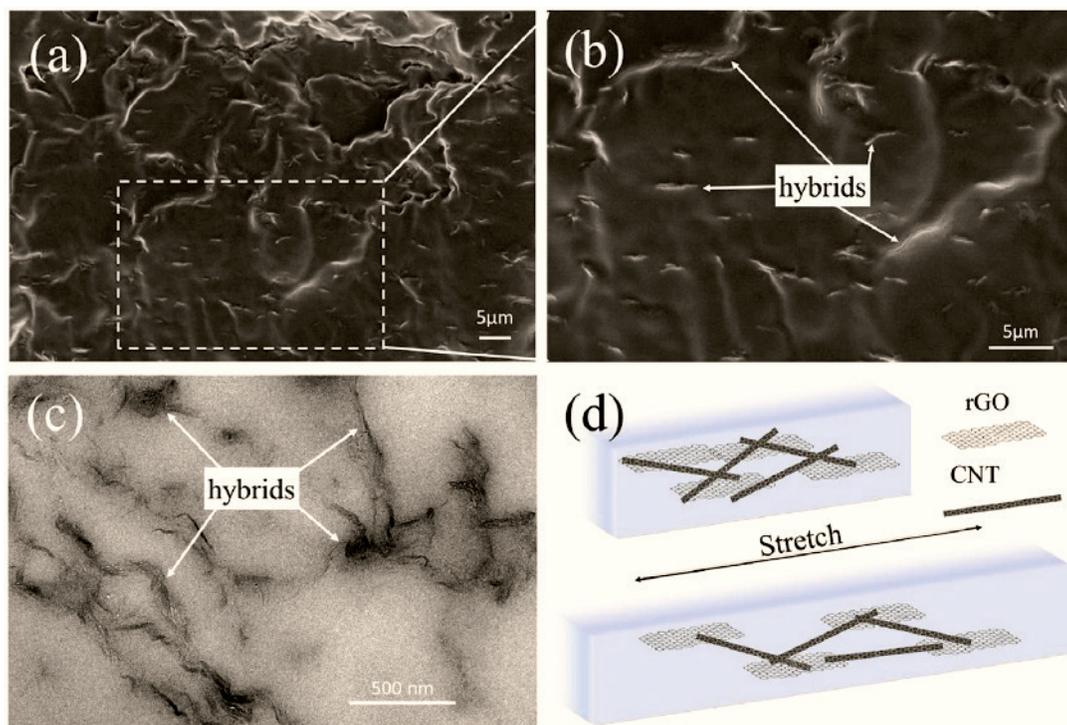


Figure 17. (a, b) SEM and (c) TEM images of SBR-rGO-MWCNTs composite and (d) schematic representation of the redistribution of the filler hybrids within SBR composite under elastic deformation [78].

5.5 Nitrile butadiene rubber-CNTs composites

Nitrile butadiene rubber (NBR) is commonly used in making hoses, joints, sealing and roll covering materials. However, there is a growing demand for its use in electronics applications. NBR can be blended with other rubbers to improve its thermal stability and gas resistance, as well as weatherability [90]. Similar to Kim et al. [50], Wang et al. [82] also employed an ionic liquid (IL) (1-aminoethyl-3-methylimidazolium bis((trifluoromethyl) sulfonyl) imide), as well as polydopamine (PDA) and (3-Aminopropyl) triethoxysilane (KH550) to functionalize the carboxylated MWCNTs, therefore overcoming their agglomeration, and to promote the stronger carboxylated NBR-MWCNTs (XNBR-MWCNTs) interfacial bonding, which facilitated the formation of the best mechanical and damping properties (see **Figure 18**). The effect of functionalizing MWCNTs before they are incorporated into rubber, for the fabrication of nanomaterials-based rubber composites, were visualized by SEM micrograph, and this can be seen in **Figure 19(a)-(h)**. Agglomerated MWCNTs are clearly seen in **Figure 19(a)**, which might have indicated that pristine MWCNTs disperses poorly in XNBR matrix. Substantial crack orientations that are seen in **Figure 19(a)** are the representation of a reduction in toughness of the XNBR-pristine MWCNTs composite and explains an obtained reduction in damping (loss $\tan \delta$) properties [82]. PDA decreased the size and amount of MWCNTs agglomeration in XNBR matrix (see **Figure 19(c)**), and this was due to hydrogen bonding between MWCNTs-PDA and XNBR matrix [82, 99]. The authors [82] reported that MWCNTs-KH550 (**Figure 19(e)**) and MWCNTs-IL (**Figure 19(g)**) had relatively stronger interaction with XNBR, and therefore the large-scale agglomeration was prevented. XNBR-MWCNTs-KH550 (**Figure 19(e)**) and XNBR-MWCNTs-IL (**Figure 19(g)**) composites showed only small cracks in the cross-section of XNBR, which indicated that the XNBR toughness was improved by functionalizing MWCNTs before they were incorporated into rubber matrix. Storage modulus of the prepared XNBR-MWCNTs composites was seen to increase by 80% (from 1392 to 2488 MPa) with the incorporation of 2.2 wt.% CNTs-KH550, while the UTS increased from 0.32 to 0.68 MPa by 110% with 3.0 wt.% CNTs-IL.

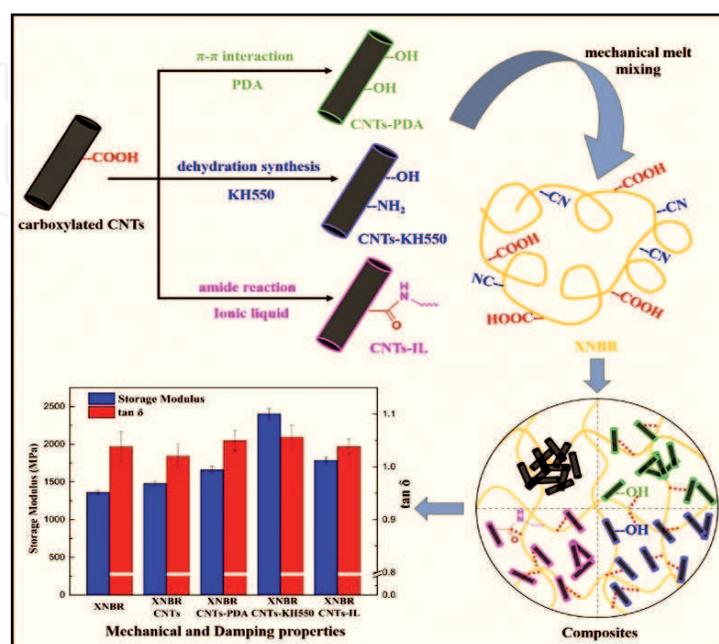


Figure 18. Functionalization of MWCNTs and preparation of XNBR-MWCNTs composites with their studied mechanical and damping properties [82].

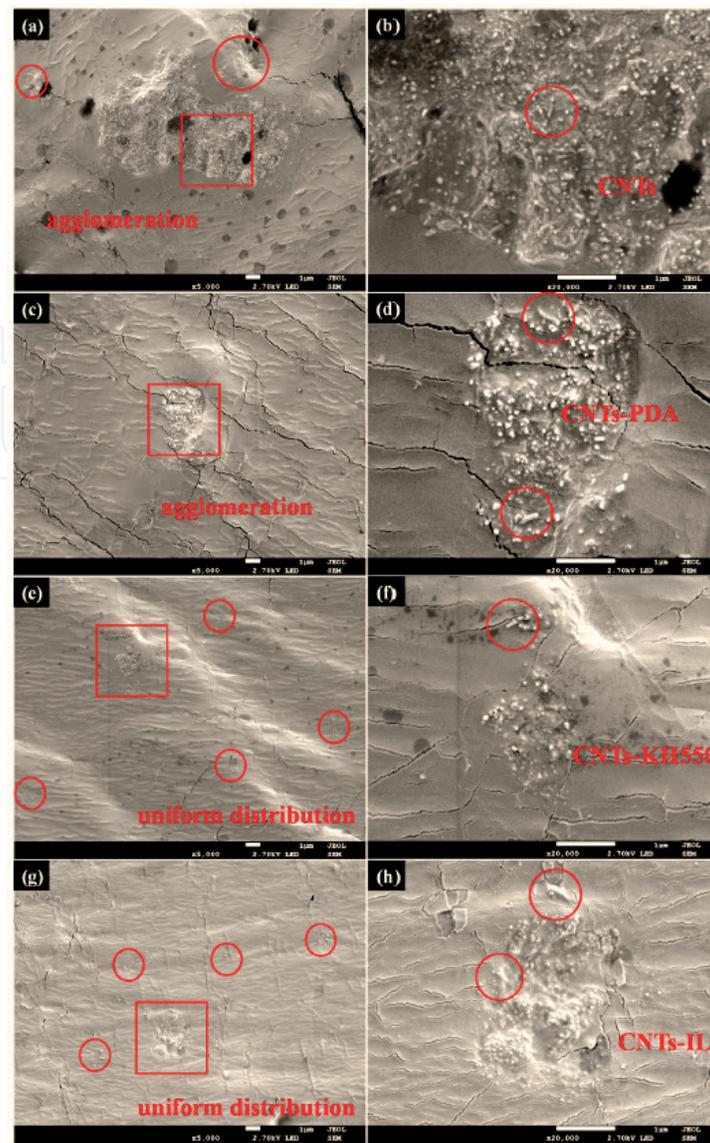


Figure 19.

Cross-sectional SEM micrograph of XNBR-MWCNTs (3 wt.%) composites with different functionalizing materials: (a, b) XNBR-MWCNTs, (c, d) XNBR-MWCNTs-PDA, (e, f) XNBR-MWCNTs-KH550 and (g, h) XNBR-MWCNTs-IL [82].

Although other researchers have reported good results of CNTs-based composites that constitute the pristine CNTs, the authors [82] have reported that the failure of damping performance that is often encountered in the field of composites, due to agglomeration of CNTs, can be suppressed by functionalizing the MWCNTs prior the fabrication of MWCNTs-based rubber composites, and this will widen the scope of the application of NBR-MWCNTs composites in the field of electronics.

Shao et al. [79] observed an improvement in UTS and hardness to about 39% and 101%, respectively, after adding CNTs into NBR, while the ϵ_b maintained at high CNTs contents over 100%. They also observed a significant reduction of volume resistivity and an increase in dielectric constant and dielectric loss with an increased CNTs content in the NBR-based nanocomposites. Additionally, the electrical percolation threshold obtained was low (1.5 pph). These results indicated the development of three-dimensional conductive networks in the composites, which meant that NBR-CNTs nanocomposites can provide insulation service to electronics [7, 79]. The presence of MWCNTs in the prepared, *via* melt and high-shear mixing methods, NBR-MWCNTs composites reduced the swelling behavior while increasing the crosslink density of the composites; consequently increasing their UTS, M, hardness, abrasion resistance and electrical conductivity (with corresponding low percolation

threshold value), and significantly decreasing their volume resistivity [6, 18, 80, 81]. These results have been attributed to uniform distribution of MWCNTs into the NBR matrix and the superior interfacial bonding between MWCNTs and the host matrix (NBR) which are due to large aspect ratio of the MWCNTs.

5.6 Fluorocarbon rubber-CNTs composites

Fluorocarbon rubbers, particularly the fluoroelastomer (FKM/FPM), are popular in electronics sector mainly because of their good heat, chemical and abrasion resistance; and does a perfect job for sealing the electronics materials. Seo et al. [100] have proposed a solvent-free encapsulation method to produce the FKM-SWCNTs composite layers that can help protect the electronic components from the physisorbed moisture, water/oxygen molecules. They suggested that this method, with elastomeric poly(vinylidene fluoride-co-hexafluoropropylene) (e-PVDF-HFP) film lamination, can potentially provide the cost-effective, large-area processable and highly dependable SWCNTs-based thin-film transistors in electronics applications. To study the hydrophilicity and hydrophobicity of each layer of SWCNTs-based thin-film transistors, the contact angle measurements of deionized (DI) water on the poly-L-lysine (PLL) solution-treated layer, SWCNTs-deposited layer, and e-PVDF-HFP layer were examined as illustrated by **Figure 20(b)**. The layer that was pre-treated with PLL solution appeared to be hydrophilic with an angle of 22.3° , and this was attributed to the hydroxyl and amine groups on the surface of the layer [100]. Due to the annealing process, an angle of this layer increased after the deposition of SWCNTs. However, the substrate retained the hydrophilicity with an angle of 57.9° . On the other hand, the e-PVDF-HFP layer was hydrophobic with an angle of 95.9° , and this was reported to be due to low surface energy that was developed by fluorine atoms in the FKM. As can be seen in **Figure 20(a)**, the hydrophobicity of e-PVDF-HFP layer can therefore be a permanent barrier to moisture, water/oxygen molecules to hamper the physisorption on the surface of SWCNTs and the oxide layer [100].

Hiao et al. [85] prepared, as shown in **Figure 21**, the porous conductive fluororubber-SWCNTs composites that were said have great stability for pressure sensing applications in electronics sector. They homogenized the pristine SWCNTs with a foaming agent called N,N-dinitrosopentamethylenetetramine (DPT) into 20 ml of methyl ethyl ketone (MEK) using a magnetic stirrer. Dispersion of SWCNTs in MEK was ensured by further sonicating and homogenizing (at 200 W for 10 min) the SWCNTs/DPT mixture, which was then mixed with about 3 g of Daiel-G801 fluororubber. The resultant SWCNTs/G801 suspension (fluororubber-SWCNTs composite) was stirred for 5 h with the aim of dissolving the fluororubber. After

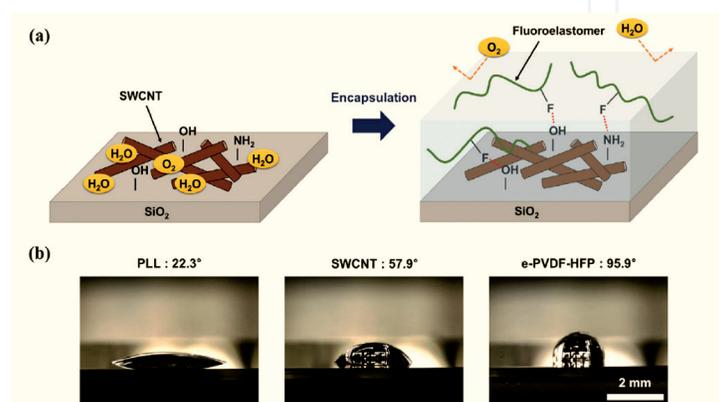


Figure 20. (a) Effect of encapsulating a layer of SWCNTs-based thin-film transistors. (b) Optical images of contact angle measurements on different layer surfaces [100].

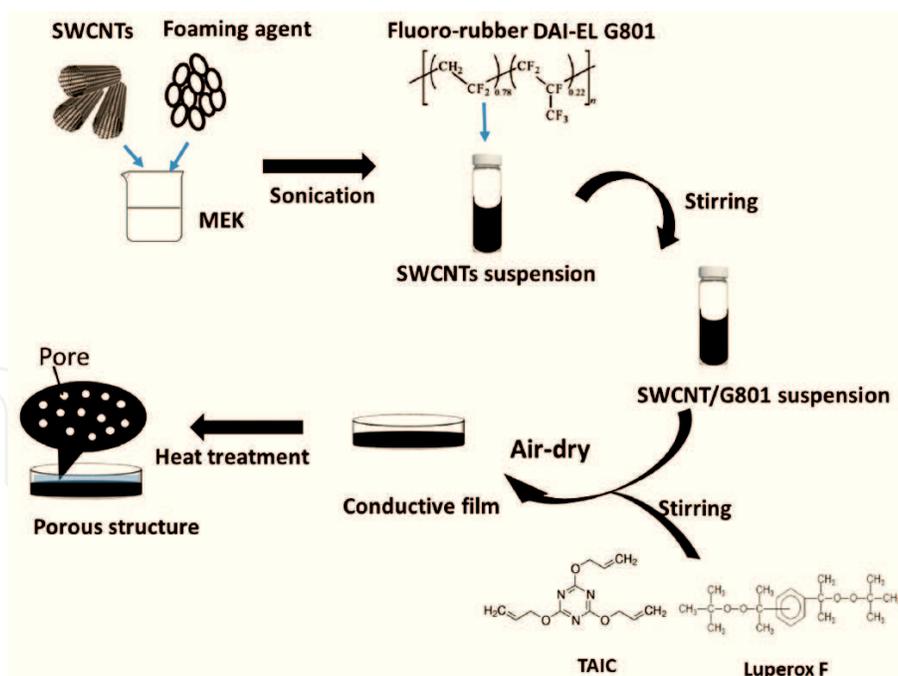


Figure 21. Preparation of porous conductive fluororubber-MWCNTs composites [85].

being air-dried at ambient temperature for 24 h, the fluororubber-SWCNTs mixture, containing the vulcanizing agents (0.15 g of Luperox F and 0.39 g of triallyl isocyanuric acid), was vulcanized on a hot press machine at about 160°C for 15 min and 180°C for 1 h. The foaming process was initiated by increasing the temperature of the press to about 210°C to obtain a porous structure after about 1 h. To print the conductive silver plates for sensing electrodes ($0.5 \times 0.5 \text{ cm}^2$) on polyimide films, a dispenser was employed. For 30 min, a sensor of the same size, pressed between the two printed electrodes, was baked at about 200°C. The prepared porous pressure-sensitive fluororubber-SWCNTs composites showed the electric resistance variation to compressive stresses with a sensitivity that is as high as 4.31 MPa^{-1} when the foaming agent (i.e., DPT) was incorporated into the composite; therefore showing a great potential for fast assembly into the printed electronic circuits and utilization in dynamic pressure monitoring applications in the electronics sector [85].

The properties of FKM have been seen to improve after the incorporation of MWCNTs [6, 60, 61]. Shajari et al. [61] made stretchable electronics materials, *via* high-shear mixing method, with FKM and CNTs. The authors observed the high electrically conductive network, with corresponding ultralow percolation thresholds of about 0.45 phr and 1.40 phr CNTs content. These results indicated that the prepared FKM-CNTs composites have a wide range of strain sensitivity. At the first strain conductivity plateau, FKM-CNTs composites gave high sensitivity with a gauge factor of 1010 at about 23% strain for 0.6 phr nanotubes content, and of 6750 at 34% strain for 1 phr content. At the second strain conductivity plateau, the composites gave high sensitivity with high gauge factor of about 4×10^4 at about 78% strain for 1.5 phr content, and the composite with 2 phr content corresponded to much higher strain of about 100% with gauge factor of 1.3×10^5 . The ϵ_b of FKM-CNTs composites was found to be as high as 430% and up to about 232% strain sensitivity. These stretchable and conductive FKM-CNTs composite materials are said to be best suited for wearable electronics, including stretchable sensors and light-emitting diodes (LEDs), as well as human motion monitoring electronics [61]. Yang et al. [60] chemically modified the surface of the MWCNTs

to get MWCNTs-COOH, MWCNTs-NH₂ and MWCNTs-A1120 filler products, as shown in **Figure 22**. MWCNTs-COOH was prepared by incorporating pristine MWCNTs into a premixed acid solution of H₂SO₄ and HNO₃ with a volume of ratio of 3:1, after which it was ultrasonically stirred for about 2 h at a temperature of 60°C and washed alternately with deionized water and dehydrated ethyl alcohol. The last step involved the filtration with suction and drying of MWCNTs-COOH in a vacuum oven at 80°C for about 12 h. For MWCNTs-NH₂ preparation, MWCNTs-COOH, N-(3-dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride (EDAC), and 4-Dimethylaminopyridine (DMAP) were ultrasonically mixed for about 30 min with dehydrated ethyl alcohol, after which ethylenediamine (EDA) was added and stirred for about 24 h. The resulted product was washed alternately with deionized water and dehydrated ethyl alcohol to remove excess reagents, and therefore achieving pure MWCNTs-NH₂. The last functionalized MWCNTs product (i.e., MWCNTs-A1120) was prepared by adding a silane coupling agent A-1120 into a premixed solution of deionized water and dehydrated ethyl alcohol at a weight ratio of 1/4 with mixing for 1 h, after which pristine MWCNTs ethyl alcohol dispersion was added into the mixed solution, followed by further mixing for about 3 h.

Comparing all prepared functionalized MWCNTs products in **Figure 22**, MWCNTs-A1120 was reported to have a relatively better interfacial interaction with FKM matrix; therefore, it showed uniform distribution within the matrix, and hence the FKM-MWCNTs-A1120 composite had relatively the best tensile properties (UTS increased by 16.58% compared to that of neat FKM; the ϵ_b was maintained above 111% with 0.5 wt.% MWCNTs-A1120 content). The nature of dispersion of MWCNTs-A1120 into FKM matrix is shown by fracture surfaces of FKM-MWCNTs-A1120 composites in **Figure 23**. At 1 wt.% and 3 wt.% MWCNTs-A1120 contents, MWCNTs-A1120 are seen to be uniformly distributed within the FKM matrix. As the content was increased, MWCNTs-A1120 began to contact each other, forming a conductive network. A dramatic agglomeration began to take place at a loading of 7 wt.% MWCNTs-A1120, which can be seen in **Figure 23(d)** (red dashed frames).

Uniform distribution of MWCNTs-A1120 within FKM matrix is attributed to the carbon-carbon double bond ($-C=C-$), formed in FKM molecular chain during vulcanization process, that chemically bonded to the amino group on MWCNTs-A1120, resulting to a strong interface between MWCNTs-A1120 and FKM matrix [60, 101]. This phenomenon also explains why the composites

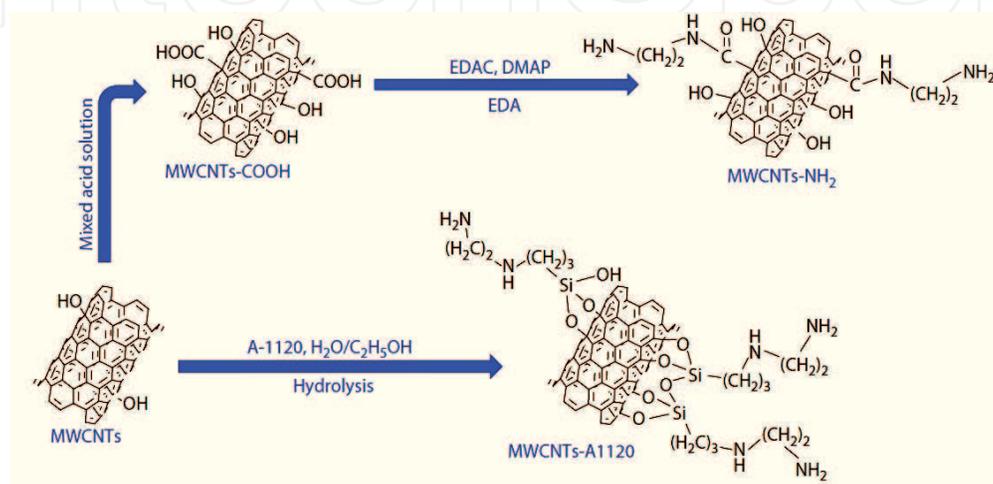


Figure 22. Schematic representation of surface modification of MWCNTs [60].

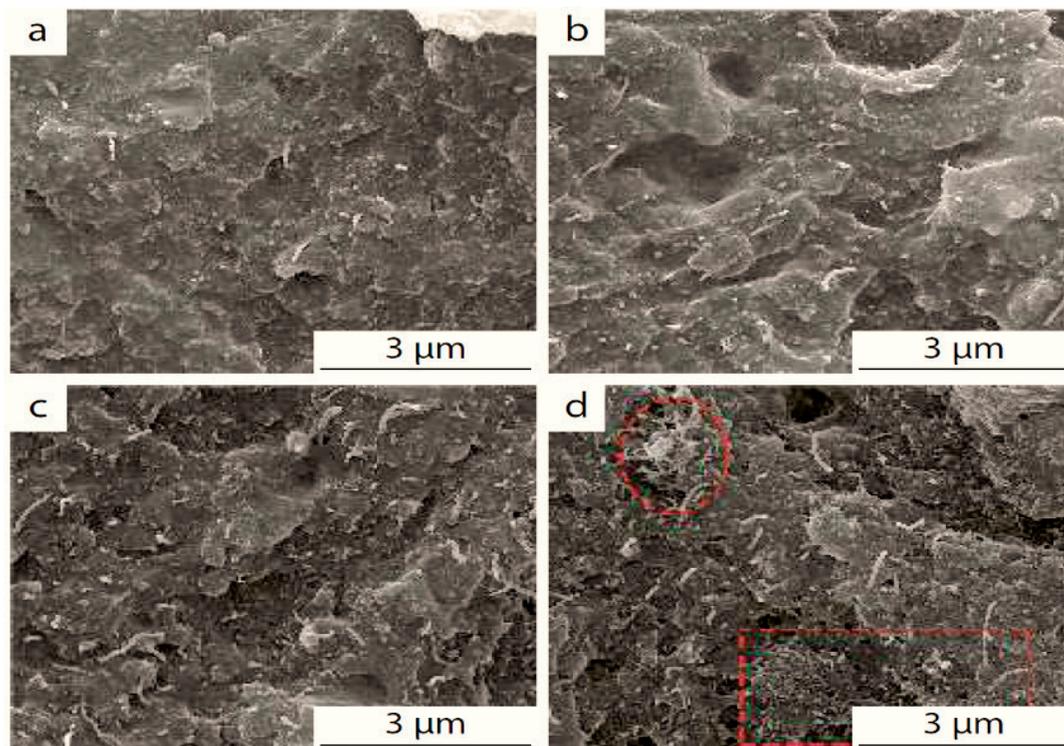


Figure 23. SEM micrograph of FKM-MWCNTs-A1120 composites: (a) with 1, (b) with 3, (c) with 5 and (d) with 7 wt.% MWCNTs-A1120 content [60].

containing amino MWCNTs (i.e., MWCNTs-NH₂ and MWCNTs-A1120) had outstanding tensile properties [60]. In addition to tensile properties, MWCNTs-A1120 seemed to enhance the electrical properties of FKM-MWCNTs-A1120 composite. As the loading of MWCNTs-A1120 was increased, the dielectric constant and dielectric loss of the FKM-MWCNTs-A1120 composite also increased, and the volume resistivity decreased. When the doping concentration of MWCNTs-A1120 reached 5 wt.%, the dielectric constant and dielectric loss of FKM-MWCNTs-A1120 composite increased significantly, and the volume resistivity got reduced. The authors [60] reported that since a conductive network can be formed by MWCNTs-A1120 product, the doping concentration of 5 wt.% MWCNTs-A1120 can be taken as the percolation threshold of the FKM-MWCNTs-A1120 composite. Nonetheless, as the tensile deformation increased, the dielectric constant and dielectric loss of the composite decreased, and the volume resistivity increased. This may be an indication that the tensile deformation can increase the spacing of the conductive filler or even destroy the conductive network structure, and hence influence the electrical properties of the materials [60]. Therefore, the great potential applications for FKM/MWCNTs-A1120 composites include flexible dielectric and flexible conductive materials [60].

6. Overview of rubber-CNTs composites for electronics

Electronics rubbers generally have some good and poor properties, and their good properties are due to their stable backbone structure and are the main factors that prompted the initial interest for the use of rubber in electronics. For instance, rubbers like NBR, EPDM and FKM are known to have high lifetime or life expectancy, with FKM having relatively the highest, as it has been shown by their hardness and compression set properties which were studied on their O-ring seals after aging for five-years at various temperatures [14]. With the current transition from CB to CNTs

due to superior properties and performances of the latter relative to the former, literature has reported that rubber-CNTs composites are currently the major application area for CNTs in the reinforcement of rubbers. Since rubber materials for electronic components need to be precisely shaped to provide proper insulation, electric shock absorption, chemical and thermal resistance and high mechanical strength, and yet be easy to use in the manufacturing and installation of the end product, several researchers have reported that CNTs are the future reinforcing materials for rubbers used in electronics, in partial or complete replacement of conventional fillers, due to the fact that rubber compounds containing CNTs have relatively ultralight weight, extremely high flexibility, and superior chemical, thermal and mechanical properties [2, 8, 35, 49, 102]. Owing to their extremely large surface area, CNTs are expected to enhance the matrices of the electronics rubbers and thereby enhancing their poor properties and further improving their good properties to produce rubber-CNTs composite materials with excellent properties [2, 34, 36, 49].

7. Conclusions

It is notable that rubber-CNTs composites present an array of possibilities for their use in electronics industry. Rubbers such as EPDM, Q, SBR, NBR, FKM/FPM, IR and neoprene are commonly used and often reinforced with conventional fillers to strengthen, insulate and seal the electronic components, including electronic circuits, wires and cables; and thereby protecting them from exposure to high-stress and extreme environments, which normally causes problems that may lead to catastrophic breakdown of the electronic machines and instruments [5–7, 9, 16]. Although the present study found that carbon black (CB) is widely used to reinforce electronics rubber materials, there is also some evidence on the use of CNTs as rubber reinforcing materials, even for rubber materials used in the electronics industry.

The studies about reinforcement of electronics rubbers using CNTs showed that even though CNTs are used only in minute quantities compared to CB, they result in outstanding properties and performances of composites. Several researchers strongly believe that rubber-CNTs composites are futuristic materials mainly for the electrical and thermal insulation since CNTs (both SWCNTs and MWCNTs) can form a conducting network within the composite material at contents that are above the certain minimum value called the percolation threshold.

The properties and performances of the rubber-SWCNTs and rubber-MWCNTs composites are often dependent on the rubber reinforcement quality which is defined by the extent of dispersion of CNTs in the rubber matrix, level of CNTs wetting with rubber, and degree of interfacial bonding between CNTs and the rubber matrix [8, 13, 21, 38]. Surface modification of CNTs and optimization of the ratio of CNTs to the amount of rubber seems to be the main factors that can possibly lead to high exploitation of the properties and performances of rubber-CNTs materials in the electronics application. The suitability of rubber-CNTs composites for the manufacturing of electronics rubber materials is based on that their properties meet most of the specifications for these materials, i.e., rubber-CNTs composites are flexible and light in weight for easy installation and are capable of resisting prolonged vibrations, high strain/pressure and most substances that may cause rubber fatigue and cracks, and therefore expose the electronics to damage [2, 6, 7, 16, 45]. Additionally, rubber compounds reinforced with CNTs can be made to be relatively cost-effective for the manufacturing and maintenance of the CNTs reinforced electronics rubber materials [5, 39, 87]. The performance of SWCNTs is comparable to that of MWCNTs in both the functional and structural composites. However, rubbers like Q are typically expensive, and

therefore their nanocomposites often constitute MWCNTs rather than SWCNTs because the former is relatively much cheaper to fabricate than the latter. The preference of conductive SWCNTs filler over MWCNTs by some researchers is due to that the former can result in relatively better homogeneity during the fabrication of rubber composites especially for pressure-sensitive rubber materials. For conductive MWCNTs, it is suggested that the MWCNTs are functionalized before they are incorporated into the rubber matrix, especially for flexible dielectric and flexible conductive materials.

Currently, the preparation of rubber-CNTs composites seems to be dominantly done at the laboratory scale. Therefore, when scaling up for mass production for CNTs reinforced electronics rubber materials in the future, the main challenge that the researchers could potentially still face is to come up with practical ways of overcoming the parameters that typically affect the properties and performance of CNTs in the prepared rubber-CNTs composites.

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Conflict of interest

The authors declare no conflict of interest.

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