

Carbon Nanotube- and Graphene-Based Conductive Inks for Smart Textiles: Sources, Processing, Properties, and Future Prospects

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Abstract

In the last few decades, smart textiles have drawn the attention of human beings for their unique and substantial properties. With the increased demand for wearable smart textiles, comfortable, soft and flexible devices are required that will not compromise the comfortability of garments. A very efficient way to make such garments is to use conductive ink. Conductive ink can be applied in garments for imparting electronic properties to the base textile material like fiber, yarn, and fabric. Conductive inks are applied in the field of transparent electrodes as cheap, sensors, labels of biological sensing, marking of radiofrequency, super capacitor electrodes, photonics, transistors etc. This study attempts to review the information of carbon nanotubes, and graphene-based conductive inks-their sources, preparation processes, properties, applications, limitations and prospects.

Keywords: Carbon nanotubes, Graphene, Conductive inks, Smart textiles

1. Introduction

About 27,000 years ago textiles were invented (Adovasio et al., 1996). The basic object of textiles to indemnify their wearer from the outer environment. In the past, leaves, barks of trees, skin of animals were used for protection from the environment and to sustain in cold regions. Humans have been using conventional textiles for their needs for several centuries. Recently a new revolution was observed in textiles, which is to integrate electronic elements in the textile material which can sense as well as respond to the state of the environment. This type of textile can be defined as smart textiles (Dejene, 2025). It can also observe its wearer and can take necessary steps according to observation. Due to the invention of flexible electronics, smart textiles are getting more popular with versatile applications. In 1883 illuminated headbands were used in the ballet La Farandole (Guler et al., 2016). This is the beginning point of smart textiles. With the invention of tiny scale and multifunctional electronic parts at a cheap rate, advancement of material science and invention of new methods for integrating electronic components into garments, a good deal of prosperity has been noticed. By inventing conductive polymer in 1977 Hegger won the Nobel Prize (Shirakawa et al., 1977). One big revolution was observed after the invention of conductive

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textiles. Due to the gigantic scale of smart textiles, it is sub classified into three classes as passive smart textile, active smart textile and ultra-smart textile (Cochrane et al., 2016). Passive smart textiles can only sense the signals from the outer environment. Only one sensor is integrated into these types of textiles. These types of textiles are sensitive to color, shape, electrical as well as thermal resistivity, for instance, Ultraviolet protective clothing, garments finished with plasma, optical sensors containing garments etc. Some other examples can also be given such as bullet proof, anti-static, anti-odor, antimicrobial garments. These types of textiles are considered the first generation of smart textiles. Active smart textiles are considered as the second generation of smart textiles. Like a passive smart textile, it can sense the outer environment furthermore it can respond according to sensation. It contains sensors like passive smart textiles, moreover it contains actuators. Shape memory polymers, phase change materials, fabrics heated with electricity are some examples of it. Ultra-smart textiles are the latest generation of smart textiles. It is the most modern technology of smart textiles. It can acclimatize with the changing outer environment. Like an active smart textile, it contains both sensors and actuators and furthermore microprocessors. It integrates some electronic devices which can dig up incentives, examine them and change themselves according to incentives. Sport jackets, spacesuits, wearable computers, smart socks are some examples of it. Smart textiles (electronic textiles) are also called E-textiles. For versatile application of smart textiles, it is required to ensure better integration of electronics devices with textiles. For fulfilling the requirements of integration semiconductor components were added with fiber during spinning (Hughes-Riley et al., 2018). Conductive fibers and functional yarns are used during fabrication of textiles (Garcia and Wood Conroy, 2010). Smart textiles are developing with the development of fiber engineering, nanotechnology, materials science and conductive ink (Kazani et al., 2017). Printing of textile fabric with conductive ink array circuit is akin to the printed circuit board (PCB) manufacturing process. It is possible to produce circuits on textile fabric with conductive ink by using a textile printing machine is the easiest method. For printing textile material with conductive ink, generally two types of conventional techniques are used, one is inkjet printing and another one is silk screen. Inkjet printing is comparatively easy and friendly to acquire higher precision. This printing can be printed on a resolution scale of 50 microns. For electrical and electronics silkscreen layout is preferable (Sauer et al., 2004). The conductive ink comprises conductive parts like silver, copper, gold, carbon nanotubes, graphene and PEDOT: PSS etc. The conductive ink is classified into two classes, first one is particle and second one is nanoparticle. Furthermore, nanoparticles are divided into two, one is organic and another is non-organic. In ink, different types of solvent are used. Mostly water is used as a solvent in case of organic conductive ink (Stoppa and Chiolerio, 2014). In this paper ultra-smart textiles are discussed which are produced by using conductive ink such as containing one of silver, carbon nanotubes, graphene and PEDOT: PSS.

Although several reviews exist on conductive inks and smart textiles, this review uniquely focuses on a critical comparison between CNT- and graphene-based inks from the perspective of textile integration. It emphasizes ink formulation strategies, printability on fibrous substrates, mechanical durability, wash stability, and scalability. Additionally, this

work highlights commercialization bottlenecks and provides comparative performance metrics relevant to wearable and washable e-textiles.

2. Carbon nanotubes (CNTs) based Conductive Ink

By a planar atomic sheet structure containing carbon atoms attached by covalent bonds like graphene, carbon nanotubes start their journey to origin (Fig. 1). Commonly the length to diameter ratio of CNTs is 1000 or higher than this. Honeycomb lattices with sp^2 bonds are found in CNTs. Like a graphite, one atom is attached with three adjacent and forms a structure without seam.

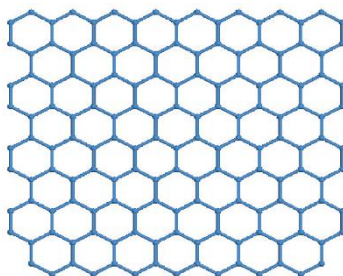


Fig. 1 Layer of a grapheme

Three different chiralities are found in CNTs structure (Hamers et al., 2003).

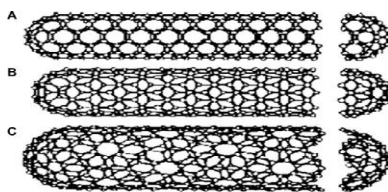


Fig. 2 (A) Armchair structure, (B) Zig-zag structure, (C) Chiral structure.

From the open part of the tube the variation of the structure can be easily understood. The tubular macromolecules containing carbon hexagons in a converging style in which two sides are closed by a structure like fullerene can be defined as single-wall carbon nanotube (SWNT) (Fig. 3). SWNT can be considered an excellent conductor.

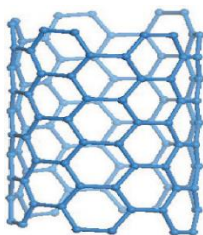


Fig.3 Single-wall carbon nanotube

On the other hand, more graphene nanotubes around the center of SWNT can be defined as multi-wall carbon nanotubes (MWNT) (Fig. 4). Layer to layer distance is about 3.3 Å.

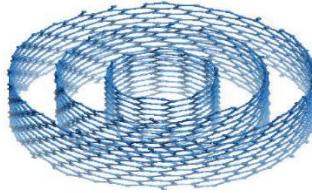


Fig. 4 Multi-wall carbon nanotube

Moreover, depending on hexagon rings alignment and diameter of carbon nanotubes, it can be divided into two- semi-conductive and metallic.

2.1 Properties

Due to containing several rare features, Carbon nanotubes (CNTs) are getting more popular. The side walls of both types of CNTs are chemically inert and hydrophobic. CNTs bear electrical properties. Along with electrical properties it also showed excellent thermal and mechanical properties (mechanical strength in terms of young's modulus ~ 1 TPa and tensile strength of ~ 150 GPa). The value of axial thermal conductivity for SWNTs is up to 3,500 W/m·K and for MWNTs up to 3,000 W/m·K (Salvetat-Delmotte and Rubio, 2002; Ruoff et al., 2003).

2.1.1 Photoluminescence spectroscopy

SWCNTs are divided into bundles, and the Van der Waals force is observed in the nanotubes. The bundles should be segmented into individuals for observing photoluminescence signals. Ultra sonication treatment is popularly used for this separation. The π -electrons system is derived from p -electrons contained by Carbon atoms of CNTs. Van Hove singularities of concentrated electronic condition of π -electrons system is presented in Fig. 5.

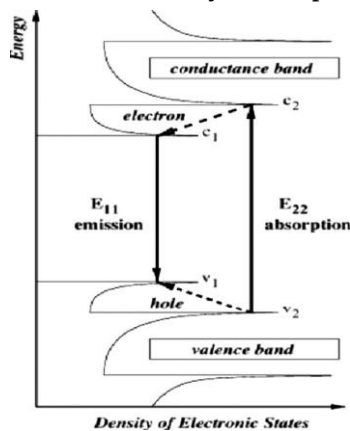


Fig. 5 Photoluminescence excitation for semiconducting SWCNT

In the figure, E_{11} transitions represent possible photoluminescence emission. Non radiative transitions of electrons and holes are presented by dashed lines. E_{11} and E_{22} represent the optical transitions. Thus, photoluminescence techniques find diameter, nature (conductive or metallic) and geometry.

2.1.2 X-ray photoelectron spectroscopy (XPS)

For observing chemical structure, this technique is used. Due to chemical reactions, the structure change of CNT walls is widely observed. Integration of nitrogen can be also observed by it (Droppa et al., 2002).

2.1.3 Electron microscopy

Scanning and transmission electron microscopy is generally used for determining structure, size as well as shape of any kind of nanomaterial.

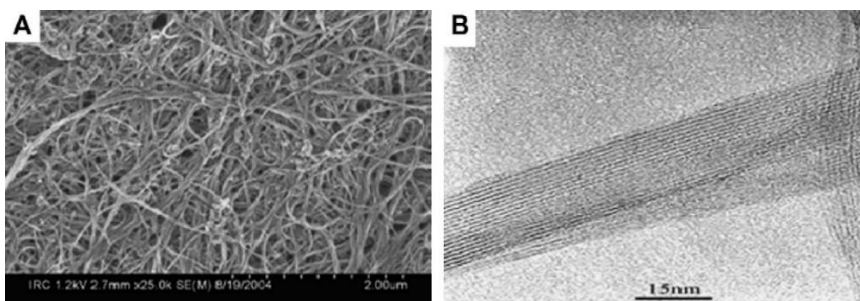


Fig. 6 (A) SEM of SWCNT, (B) TEM of SWCNT

2.1.4 X-ray diffraction

It provides information about impurities, layer to layer spacing and structural strain. The XRD pattern of graphite and CNTs are almost the same.

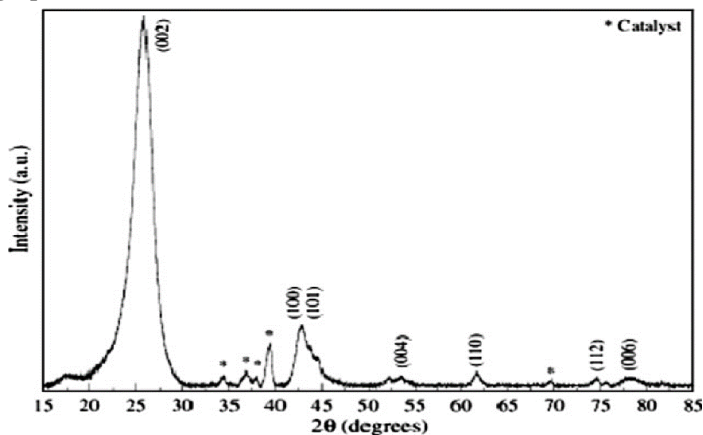


Fig. 7 XRD pattern of MWCNT

2.1.5 Neutron diffraction

It is basically used for measuring structural features like length of bond probable distortion of hexagonal network (Burian et al., 2004). Neutron diffraction is affected by polydispersity of CNT diameters, bundle tubes, CNT bundle size (Rols et al., 1999).

2.1.6 IR spectroscopy

It is commonly used for determining the impurities in a sample. It can show the structural change of CNTs and understand the compound mixed with CNTs. In research, the properties of MWCNTs were determined which were modified by amino compounds (Saito et al., 2002).

2.2 Ink preparation

For preparing carbon nanotubes ink the main obstacle is to dispersion of carbon nanotubes. Proper surface tension and viscosity should be controlled precisely. Due to the presence of van der Waals forces among carbon nanotubes it is very difficult to disperse carbon nanotubes in a liquid. It is possible to disperse carbon nanotubes by using organic solvents, dispersants or by functionalizing sidewall (Song et al., 2009; Okimoto et al., 2010; Gracia-Espino et al., 2010). The surface tension of the ink should be low for proper ejecting through the nozzle.

2.3 Applications

Nowadays CNTs replace indium tin oxide (ITO) for producing transparent electrodes as a cheap source. CNTs not only reduce the cost but also make the device flexible. It is used in case of sensors (Li et al., 2003; Fu et al., 2010), labels of biological sensing (Abera and Choi, 2010), marking of radiofrequency (Yang et al., 2009), photonics (Avouris et al., 2008), transistors (Artukovic et al., 2005) and in case of many other applications. There are several printing processes used for accumulating CNTs on substrates. Screen printing (Li et al., 2003a), aerosol printing (Ha et al., 2010), transfer printing (Zhou et al., 2006), and contact printing (Liu and Choi, 2009) are commonly used for accumulating CNTs on different substrates. Among all, inkjet printing is prominently used for printing textile material.

Mass printing processes were used for applying MWCNT on polyester and polyamide microfilaments-based textile substrate as well as on coated paper for producing electrically conductive layers which can be used as heating arrangement (Fischer et al., 2011).

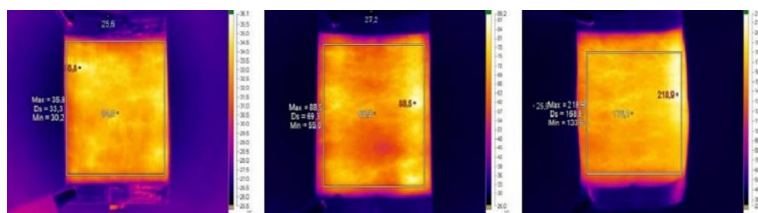


Fig. 8 Thermal images of printed CNT layers on textile substrate, number of layers: 3 (left), 8 (middle), 20 (right)

In a study, the CNT, binder and water were mixed, and coating paste was formulated. Microscopic view of coating paste is shown in figure 9.

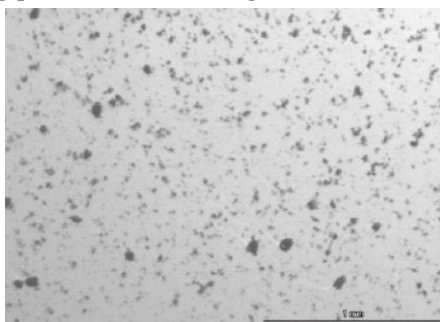


Fig. 9 Agglomerates of CNTs

Thickener was added for adjusting the viscosity of coating paste. Coating paste was applied on a multi-layered structure. Microscopic images of the textile structure are shown in figure 10.

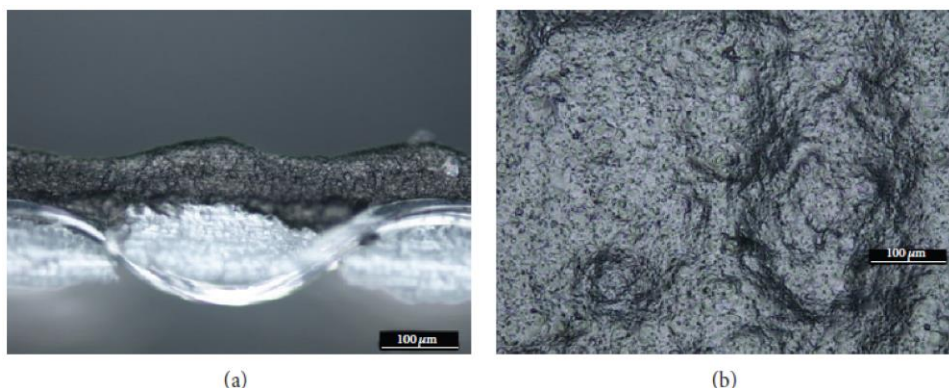


Fig. 10 A cross section (a) and surface (b) of textile structure coated with coating paste (Govaert and Vanneste, 2014)

3. Graphene based Conductive Ink

Graphene is a new addition in the field of nanocarbons but getting more popular for its excellent thermal, electrical, mechanical as well as optical characteristics. Cost of Graphene-based inks is comparatively low, environment friendly and showed excellent electrical conductivity. It is applied in the field of photovoltaic (Jeon et al., 2017), sensors (Zirkl et al., 2011; Dua et al., 2010) and thin-film transistors (Lim et al., 2012; Kong et al., 2012).

3.1 Preparation

There are two ways of graphene and its derivatives preparation, one is bottom-up and another is top-down (Huang et al., 2011; Huang et al., 2012). In case of bottom-up process graphene

is produced from carbon sources and in case of top-down process graphene is produced from bulk material. Different methods have been developed for obtaining graphene like thermal reduction (Mao et al., 2010), photochemical reduction (Ng et al., 2010), chemical reagent reduction (Fan et al., 2008), sonolysis (Vinodgopal et al., 2010), photothermal reduction (Cote et al., 2009). From the mentioned method chemical reduction of graphene oxide (GO) is widely used. Due to hydrophobicity of graphene, it is very hard to disperse in water to apply as ink. For improving dispersing ability and electrical conductivity it is required to modify graphene by integrating functional groups either by non-covalent approaches or by covalent bonding (Liu et al., 2012; Lonkar et al., 2015).

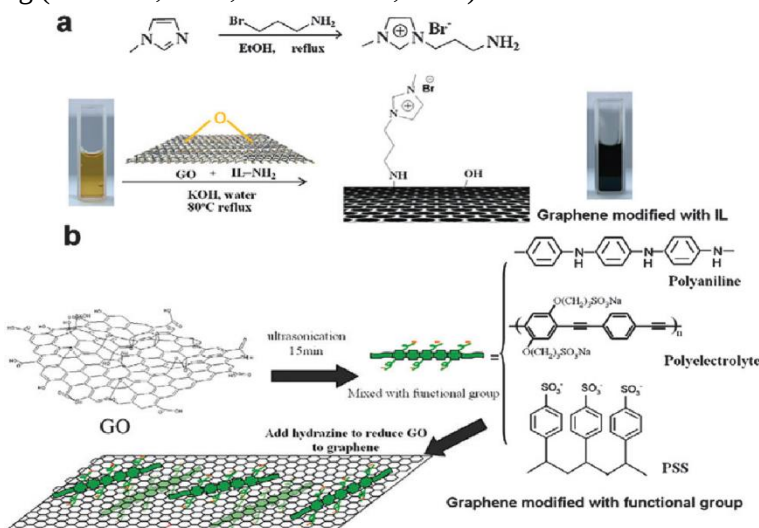


Fig. 11 Graphene functionalization: (a) graphene functionalized by ionic liquid and (b) graphene modified by different functional groups of polyaniline, polyelectrolyte and PSS anions.

3.2 Graphene ink formulation

The ink needs high surface tension as well as low viscosity for easy passing through the nozzle. There are two ways for producing graphene inks, first one is to use stabilizers for ultrasonic exfoliation of pristine graphene sheets, second one is to use stabilizers for chemical reduction of GO. There are different types of stabilizers such as polymers (PVP, PVA), surfactants, polycyclic aromatic hydrocarbons and ethyl cellulose. For ink formulation organic solvents like dimethyl sulfoxide, ethanol and terpeneol or water are used as prime solvents.

Scientists reported a simple method for formulating stable graphene inks. Graphene was collected from graphite flakes in DMF after that a little amount of ethyl cellulose was put on the above-mentioned solvents. Then the DMF was exchanged by terpeneol by distillation. For preparing dispersion suitable for inkjet printing ethanol was added for enhancing surface tension as well as viscosity (Li et al., 2013).

Another researcher proposed a method for producing graphene ink by using ethyl cellulose (EC) powder/graphene at normal temperature. With ethanol and EC graphite was exfoliated and graphene inks were produced. The steps of graphene ink production shown below figure 12 (Secor et al., 2013).

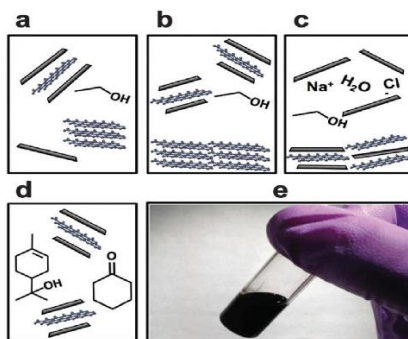


Fig. 12 (a) Graphene is exfoliated from graphite powder in ethanol/EC, (b) Without large graphite flakes, (c) Graphene/EC flocculation with salt, (d) Graphene/EC powder dispersed on 85: 15 cyclohexanone/terpineol and produced ink, (e) Graphene ink in vial (Secor et al., 2013).

3.3 Properties

3.3.1 Fourier Transform Infrared (FTIR) Spectroscopy

In this technique, absorption spectra are formed when molecules of a substrate absorb IR. Three incidents can occur when absorbing IR. Firstly, rotational transition may occur, secondly vibrational transition, thirdly electrons can transfer to higher energy levels; it can be referred to as electronic transitions.

3.3.2 Raman spectrum

In this technique, energy of photons is transferred up or down due to interaction with laser light. This transformation gives a message about vibrational modes of the system. It is a very important tool for determining the behavior of protons and electrons in graphitic materials. Fig. 13 represents a typical figure of a GO.

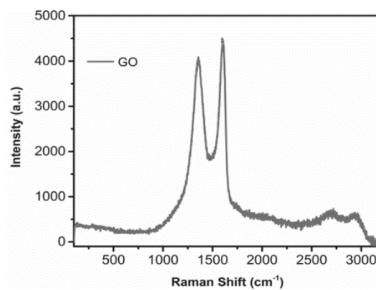


Fig. 13 Typical figure of a GO (Raman spectrum) (Song et al., 2019)

3.3.3 Atomic Force Microscopes (AFM)

Before taking AFM images, for obtaining proper size of graphene sheets in ink, the solution was centrifuged and then filtered. After taking a big amount of AFM images, generally the graphene size is found at 400 nm.

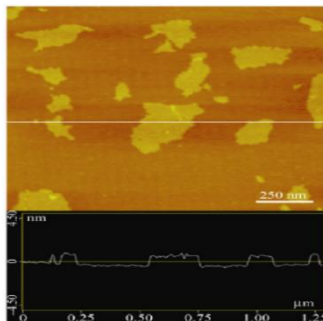


Fig. 14 Typical AFM images (top) and cross-sectional profiles (bottom) of GO sheets in GO ink (0.1 mg/mL) (HuangL et al., 2011)

3.4 Applications

By using wet-spinning process, at first time graphene fiber was produced (Yu et al., 2011). A structure of supercapacitor electrodes was developed by using graphene-MnO₂. At scan rate 2 mVs⁻¹, it shows the value of specific capacitance up to 315 Fg⁻¹ (Figure 15) (Meng et al., 2013).

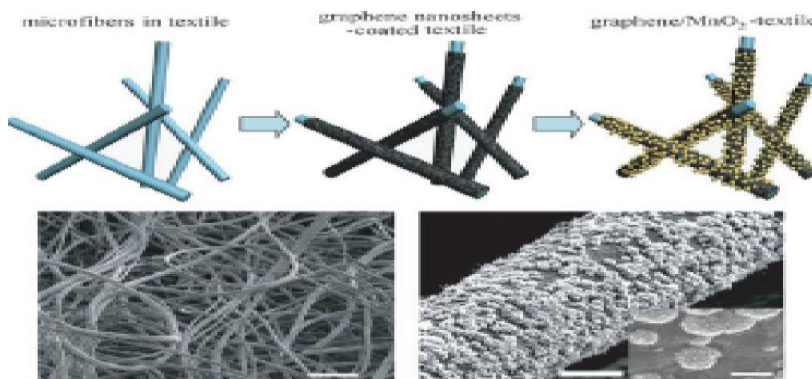


Fig. 15 Super capacitor electrode

Fabric showed excellent flexible properties when applied graphene on it. Pad-dry-cure method was used for functionalizing the periphery of cotton fabric by applying graphene nanoplate (0.05–0.4 weight %) for protecting it from UV rays. UPF value 356.74 was obtained when applied 0.4 weight% graphene on fabric (Meng et al., 2013).

By applying conventional emulsion polymerization technique cross-linked styrene/divinylbenzene nanoparticles functionalized with hydroxyl containing 1 weight% hydroxyethyl methacrylate were prepared. By using modified Hummer's method Graphene oxide (GO) was produced. PVA was used for preparing reduced graphene oxide (rGO). Inkjet printer was used for printing purposes. The all over process is shown in figure 16 (Karim et al., 2017).

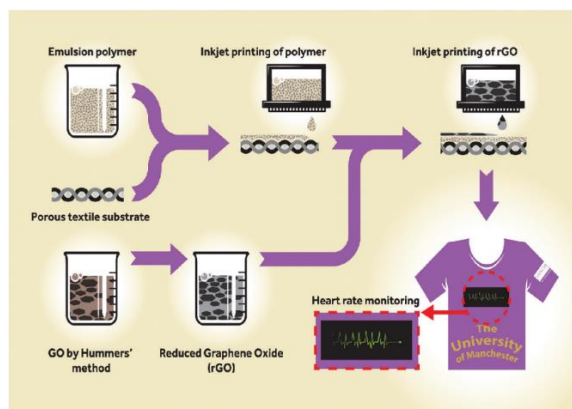


Fig. 16 Graphene based e-textile manufacturing with inkjet-printer (Karim et al., 2017).

4. Limitation and recommendations

Carbon nanotubes (CNTs) and graphene-based conductive inks offer exciting opportunities for the development of smart textiles, but they also come with certain limitations. Let's explore these limitations, potential solutions, and the prospects of these materials in the context of smart textiles. One of the main challenges with CNTs and graphene-based inks is their high production costs, which limit their widespread adoption. However, ongoing research and advancements in manufacturing techniques are aimed at reducing the cost of production. New methods such as scalable synthesis and printing processes are being explored to make these materials more cost-effective.

Achieving a homogeneous dispersion of CNTs or graphene in the ink matrix can be challenging. Aggregation or agglomeration of these materials can affect the ink's stability and hinder its performance. To address this, researchers are developing functionalization techniques that modify the surface properties of CNTs and graphene, enabling better dispersion and ink stability. Surface modifications such as chemical functional groups or polymer coatings can enhance compatibility with ink medium.

CNTs and graphene-based inks may face limitations in terms of flexibility and durability when applied to textiles. However, strategies such as blending these materials with polymers or other additives can improve their flexibility and enhance the mechanical properties of the resulting conductive films. Additionally, optimizing the ink formulation and deposition techniques can ensure better adhesion and durability on textile substrates.

Scaling up the production of CNTs and graphene-based inks for industrial applications remains a challenge. However, advancements in large-scale synthesis techniques, such as chemical vapor deposition for graphene and aerosol-based methods for CNTs, are being explored to enable mass production. Furthermore, collaborations between academic institutions, industry, and government agencies can drive the development of standardized manufacturing processes and quality control measures.

5. Conclusion

Despite the limitations, CNTs and graphene-based conductive inks hold great promises for smart textiles and wearable electronics. Integration of CNTs and graphene-based ink enables the development of smart textiles with enhanced sensing functionalities, such as strain, temperature, and gas sensors. These materials offer high sensitivity and fast response times, making them ideal for various applications in health monitoring, sports performance, and environmental sensing.

CNTs and graphene have excellent electrical conductivity and can serve as components for energy harvesting and storage devices. Smart textiles incorporating these materials can generate and store energy from body movements or environmental sources, enabling self-powered wearable electronics.

CNTs and graphene-based inks can be used to fabricate conductive patterns, interconnects, and electrodes on textiles, enabling the integration of electronic components into wearable devices. This opens opportunities for flexible and lightweight sensors, displays, and communication systems embedded in clothing.

CNTs and graphene-based inks can enable multifunctional smart textiles with properties such as electromagnetic shielding, antibacterial properties, and controlled drug release. These materials offer the potential to transform conventional textiles into intelligent and interactive systems.

Efforts have been made to report the definition, classification of smart textile, description, preparation, properties and application conductive inks in smart textile. Two methods were described for preparing graphene based conductive ink. A method for preparing carbon nanotube based conductive ink was also described. For applying carbon nanotube as conductive ink, proper surface tension and viscosity should be maintained. Commonly length to diameter ratio of CNTs should be more or equal to 1000. More research is required for industrial-scale production.

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