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Recent advance in using eco-friendly carbon-based conductive ink for printed strain sensor: A review

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A R T I C L E I N F O

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A B S T R A C T

Printed electronics specifically printed strain sensor is emerging as a way forward for wearable application because of its flexibility and sustainability. Many efforts have been made to ensure the eco-friendliness of synthesized carbon-based ink to reduce the electronic waste. Carbon based fillers such as carbon nanotube have been widely used because of high electrical conductivity and excellent mechanical properties. However, the production of carbon-based fillers towards the environment still needs to be attended due to the involvement of hazardous fossil-based precursors that may harm the environment. Besides, the involvement of binders such as polyvinyl chloride (PVC), synthetic solvents and additives in the synthesis of the carbon-based conductive ink can impact serious health and environmental issues. Hence, the usage of natural precursors for green synthesis of carbon and the incorporation of biopolymer binder which are environmentally friendly and renewable need to be considered as an alternative to produce eco-friendly conductive ink. This review article presents the progress in green synthesis of the carbon-based filler, recyclability of the ink and material selection for the ink composition from biopolymer binder, solvent and additives that are eco-friendly. The performances of the carbon-based conductive ink are discussed in terms of the percolation theory and tunneling effect that form the conductive pathway in microscopic level in stretching and relaxing phenomena for printed strain sensor applications. The rheological properties of the printed ink such as viscosity, surface tension and adhesion properties to the chosen substrate also plays crucial role depending on the chosen printing technique of the printed strain sensor. The highlight of this paper is it also correlates the performance of the printed strain sensor in terms of its sensitivity using different eco-friendly carbon-based conductive ink with different printing techniques.

1. Introduction

Recent years demonstrated an increasing trend of printed strain sensors for wearable applications such as human motion monitoring (Lian, 2022); (Hwangbo et al., 2023), soft robotics (Hu, 2022) and healthcare monitoring (Du, 2020). Printed strain sensor grasps huge attention in wearable application due to its simple process, cost-effectiveness, and its customizability for flexible and stretchable devices. Fig. 1 shows the potential usage of printed and flexible strain sensors for various applications from health care monitoring (pulse wave detection, wound healing monitoring), fitness and sport (smart headband and sweat detection), internet of things (smart wearables, printed RFID tags) to agriculture (printed strain sensor for plant growth monitoring).

Several printing techniques have been widely implemented for printed strain sensor fabrication such as screen printing, inkjet printing and 3D printing. Selection of conductive printable ink is crucial for printed electronics. There are four main elements in the conductive ink which consist of conductive filler, polymer binder, solvent and additives. Each printing technique required different conductive ink properties which includes viscosity, filler concentration and the ink adhesion towards chosen flexible substrate. This addressed the importance of material selection towards ink formulation as it will impact the printed sensor performance. Materials selection in formulating the printed ink

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need crucial attention to ensure the ink properties met printing requirements and produce good strain sensor performance. Current traditional printed strain sensors fabrication used metallic components (silver and copper) as the filler (Mohammed Ali, 2018) (Liu et al., 2018) and show promising performance in terms of gauge factor for various applications (Liu, 2018); (Wang et al., 2019). However, metal-based printed strain sensors were facing several drawbacks such as non-flexible due to its rigidity, low stretchability and low sensitivity, resulting to its incompatibility for wearable application (Kanoun et al., 2021). Copper and silver are among popular metals to be used for their excellent electrical conductivity. However, copper resulted to oxidation issues while silver showed unstable performance for conductive ink (Jeong, 2008); (Tran et al., 2018). Plus, high sintering temperatures required by metal-based ink limit its usage towards flexible substrate. Hence, carbon conductive ink was chosen as a promising alternative due to their high electrical conductivity and excellent mechanical properties (Yi et al., 2022); (Klemens, 2000).

Hazardous binder, solvent and additives such as polyvinyl chloride (PVC) (Yazdani et al., 2016) tetrahydrofuran (Thiyagarajan et al., 2019) were also used in the current conductive ink that led to environmental concerns (Sanchez-Duenas, et al., 2023) (Htwe and Mariatti, 2022). The incorporation of toxic chemicals in the ink formulation such as organic solvent and additives emits harmful volatile organic compounds (VOCs) that can harm both humans and environment (Lukman Hekiem et al., 2021). To overcome this issue, the emergence of greener alternatives using eco-friendly conductive carbon-based materials started to be implemented in printed strain sensor fabrication. Materials selection in formulating the printed ink need crucial attention to ensure the ink properties met printing requirements and produce good strain sensor performance. Therefore, selection of binder and additives should not only hold good properties but also need to avoid toxic and harmful chemicals. Some of eco-friendly binder examples are polydimethylsiloxane (PDMS) (Fu et al., 2019); thermoplastic polyurethane (TPU) (Xiang, 2019), cellulose (Franco et al., 2020) and chitosan (Camargo et al., 2022); (Maizal Hairi et al., 2022).

Hence, the aim of this paper is to review various types of carbon polymer composite ink composition that are eco-friendly specifically for printed strain sensor application. The review involves the ink synthesis technique, filler- binder ratio including the selection of organic solvent and additives for eco-friendly ink. This review articles presents the progress in green synthesis of the carbon-based filler, recyclability of the ink and material selection for the ink composition from biopolymer binder, solvent and additives that are eco-friendly. The performances of the carbon-based conductive ink are discussed in terms of the percolation theory and tunneling effect that form the conductive pathway in microscopic level in stretching and relaxing phenomena for printed strain sensor applications. The highlight of this paper is it also correlates the performance of the printed strain sensor in terms of its sensitivity using different eco-friendly carbon ink with different printing techniques.

The organization of the paper is as follows. Section 2 focused on material selection for eco-friendly conductive carbon-based ink for printed strain sensor which covers ink components. Each of these ink components, which are carbon filler, binder, solvent and additives were covered in detail in each subsection. Next, the ink properties which includes basic percolation theory, electrical conductivity of fabricated ink, rheological properties of the ink and ink characterization were discussed in Section 3. Recent research works reported on eco-friendly carbon-based materials were compiled in this section. Section 4 described the piezoresistive theory of printed strain sensor. The performance of fabricated printed sensors using eco-friendly carbon conductive ink have also been discussed in this section. Lastly, possible recommendations and future directions were discussed.

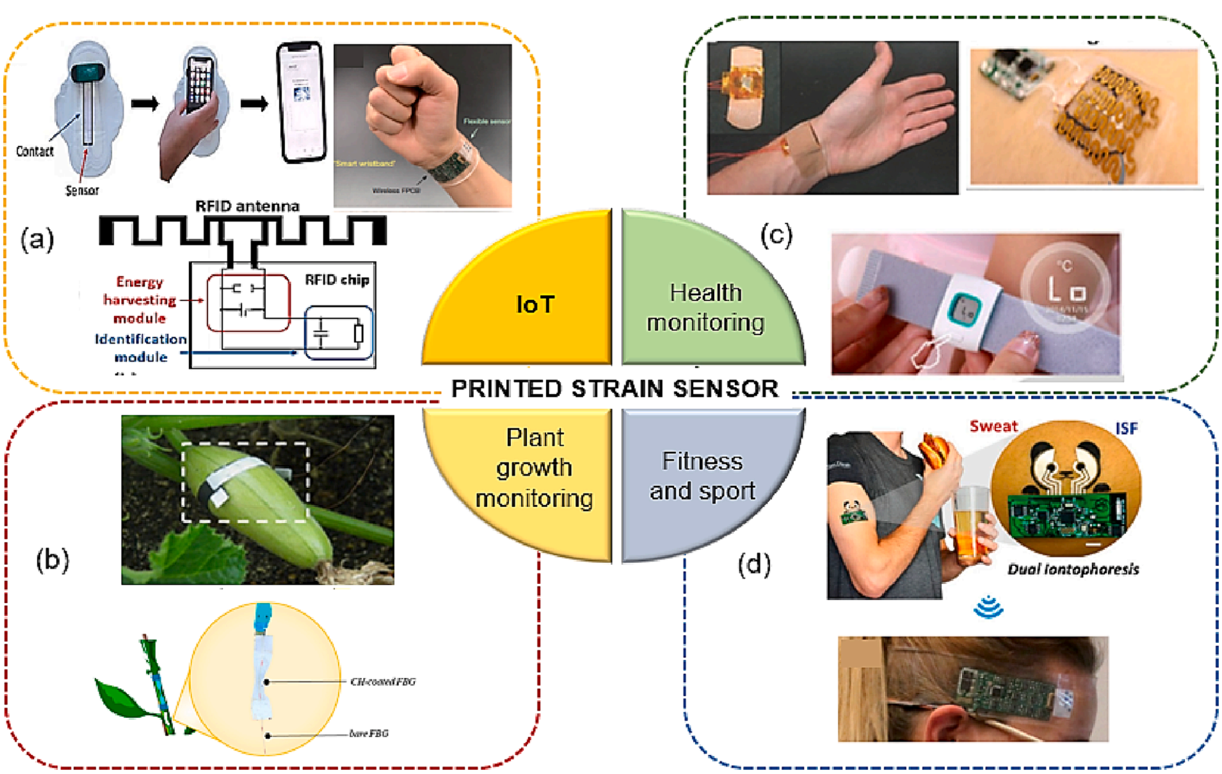

Fig. 1. Printed strain sensor application (a) IoT application which includes printed graphene for RFID application (Conti, 2023); (Wang et al., 2023), Smart wristband (Khan et al., 2019) (b) Printed strain sensor for plant growth monitoring (Lo Presti, 2021); (Tang, 2019) (c) Pulse wave detection (Li et al., 2021), wound healing monitoring (Lin, et al., 2022), body temperature monitoring (Lin, et al., 2022) (d) Printed strain sensor for sweat detection, smart headband (Khan et al., 2019).

2. Composition of eco-friendly carbon-based conductive ink

Conductive inks play a vital role in printed electronics, specifically printed strain sensors due to recent demands for wearable applications. However, there are challenges in terms of the usage of toxic chemicals in the conductive ink synthesis and complex fabrication process. Generally, printable conductive ink fabrication was classified into four main elements which are conductive filler, solvent, binder and additives. Selection of material for fabrication of printed conductive ink was crucial as different ink properties were required depending on printing techniques and its application. Printed conductive ink consists of a mixture of conductive fillers uniformly disperse in a certain amount of binder, additives and solvents. Fig. 2 illustrates the components of the printable ink which consist of conductive filler, binder, solvent and additives. For conductive fillers, most of the previous work reported on metal-based, carbon-based, metal nanoparticles (NP) and nanowires. This paper will focus on carbon based conductive filler such as carbon nanotube, graphene and carbon black. For binder, selection of polymer binder such as chitosan and cellulose was significant in binding the particles element together and determine the ink rheological properties according to printing techniques. Organic and inorganic solvents play important roles to control the solubility for the binder that will allow the ink to flow easily. Two types of conductive carbon-based conductive ink will be discussed such as water-based and solvent-based ink. Additives in the conductive ink acts as the dispersants, surfactant or as an adhesion promoter such as gum arabic and sodium dodecyl sulphate (SDS).The important properties of each of the ink components such as viscosity, surface tension and adhesion properties also are some of crucial parameters that need to be taken into account as shown in Fig. 2. Some examples of fillers, binders, solvents and additives used in the previous works were also illustrated respectively.

2.1. Carbon-based conductive filler

Conductive filler is responsible for determining the electrical properties of the ink. The selection of conductive ink is highly dependent on the conductivity and durability of the design upon bending for flexible electronics. There are two common types of conductive filler which are metal-based, carbon-based, metal nanoparticles (NP) and nanowires. Nanomaterials based conductive ink has emerged as promising material for conductive ink due to its good conductivity. However, metal-based filler are less favorable due to its fragile and rigid properties, which does not offer great performance for printed flexible wearable sensors (Park et al., 2015). Various conductive polymers have been studied as an alternative for printable conductive ink such as poly(3,4- ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) (Alshammari et al., 2014) and polyaniline (PANI) (Lin et al., 2017) (Tong, 2018), polypyrrole (PPy) (Li, 2018) because no annealing is needed (Htwe and Mariatti, 2022). However, the conductivity for conductive polymer is still very low compared to metal-based ink. Among the various selection of conductive ink, carbon-based ink has promising potential for flexible electronics application. Carbon-based conductive filler comes in several varieties such as graphene (Franco et al., 2020) (Pan, et al., 2018), graphite, carbon nanotube (CNT) (Akindoyo et al., 2021) and carbon black (Lian, 2022). The utilization of carbon-based conductive filler is in demand compared to metal-based as it offers interesting properties such as good conductivity (Yi et al., 2022) and mechanical properties (Yi et al., 2022); (Kim, 2017).

Graphene consists of two-dimensional (2D) carbon atoms bonded by molecular bonds. The usage of graphene has been popular among numerous areas such as sensors, biomedical engineering, nano and flexible electronics (Nag et al., 2018). Graphene was one of favored conductive carbon-filler used due to its great mechanical, thermal and electrical properties (Htwe and Mariatti, 2022); (Franco et al., 2020). Pan et al. (Pan, et al., 2018) prepared graphene-based ink using non-toxic solvent which was Cyrene. The synthesized ink was reported to

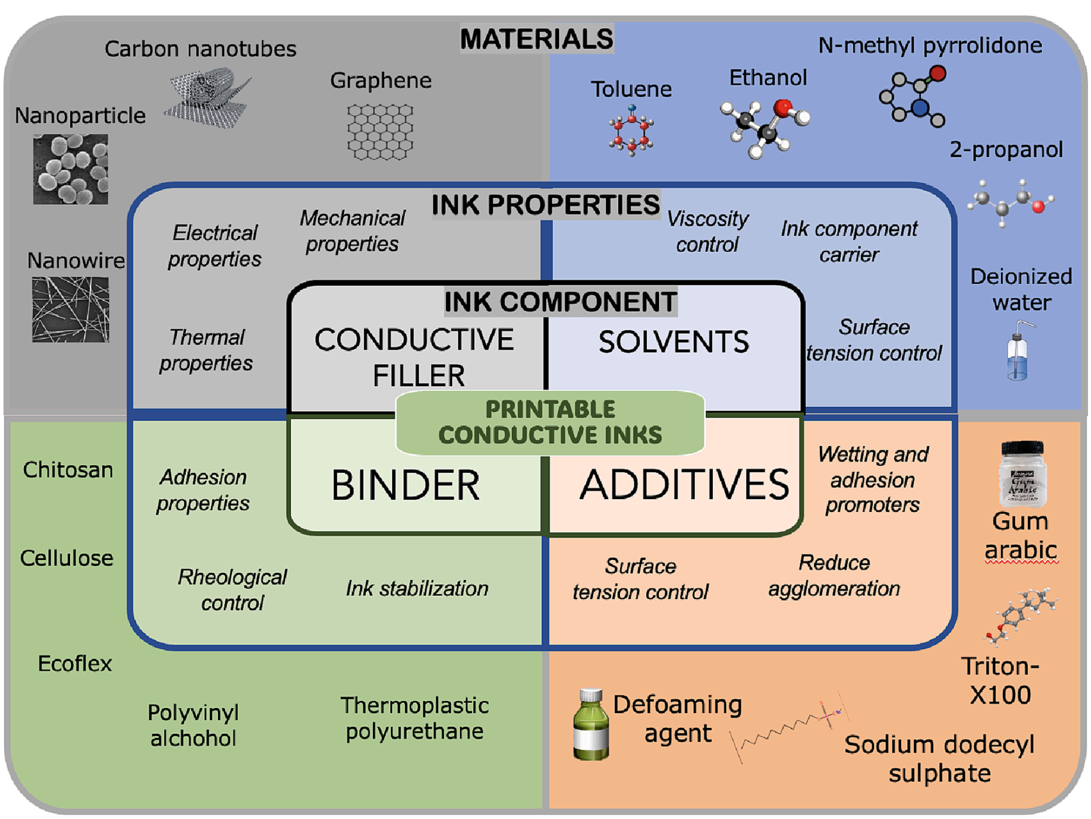

Fig. 2. Ink composition for printable conductive ink fabrication from the component, properties and examples of the materials used.

have 7.13 x 10^4 S/m conductivity. He et al. (He, 2019) prepared graphene nanoplatelets (GNP) ink printed on plastic and paper via screen printing technique. Printed GNP exhibited excellent electrical conductivity at 8.81 x 10^4 S/m. However, the GNP ink preparation in this work required complicated process and longer annealing time for printed GNP to dry which at 80℃ for 2 h. Franco et al. (Franco et al., 2020) developed graphene combined with carboxymethyl cellulose for printed electronics. The printed ink recorded sheet resistance at 197 Ω/□. This work reported to have low annealing temperature of 100℃ for only 5 min, which was excellent in eliminating the need for high temperature and long duration for annealing process compared to previous works (Pan, et al., 2018); (Bellani, 2019).

Meanwhile, graphite is a layered planar structure that consist of rings of six carbon atoms arranged in a widely horizontal sheets, where one carbon atom forms covalent bonds with the other three neighbors. (Khan et al., 2019). It is well known for having good thermal and electrical conductivity, primarily along its planes. There are strong covalent bond that exist within the sheets but weak interaction between their layers, which allow them to easily slide passing each other (Boikanyo et al., 2018). This weak interaction is known as Van der walls bond. Nonetheless, these bonds have no direct impact in making graphite to have a good electrical conductivity, making graphite one of choice for printed conductive ink. Zhang et al. (Zhang, 2022) fabricated highly anisotropic graphite framework (HAGF)/PDMS composite for high performance strain sensor. This sensor recorded conductivity at 45 S/m with gauge factor (GF) between 1.3 and 379, indicating promising performance for strain sensor application (Zhang, 2022). In 2017, Weigu et al. (Li et al., 2017) reported a 3D graphite/PDMS composite strain sensor which exhibited GF of 52 at 100 % strain. However, the graphite was required to undergo chemical vapor deposition (CVD) process before being incorporated with PDMS, which marked a time consuming and complicated process.

The implementation of CNT as fillers attracted vast researchers in flexible strain sensor fabrication. CNT consists of graphene sheet that been rolled up into various lattice directions, and the number of sheets rolled will determine the final properties of the CNT materials (Khan et al., 2019). CNT can be referred as a single-wall carbon nanotube (SWCNT) and multi-wall carbon nanotube (MWCNT). SWCNT occurred due to the rolling of a single sheet of graphene, while MWCNT occurred due to the rolling of multiple graphene sheets (Khan et al., 2019). CNT is known to have a strong chemical bond as it is bonded by sp2 hybridization bond, which is higher than diamond which has sp3 bond (Boikanyo et al., 2018). This strong covalent bond resulted to an excellent mechanical property of CNT having Young Modulus between 250–950 GPa and great tensile strength of 11–63 GPa (Rizvi, 2014). Kim et al. (Kim, Dec. 2017) reported value of tensile strength at 0.85 GPa and Young Modulus of 34.65 GPa as properties for MWCNT reinforcement in composite system. Deng et al. (Deng et al., 2011) obtained SWCNT Young Modulus ranging from 530 to 700 GPa as effective value to be used in composite reinforcement (Dai and Sun, 2016). These great mechanical properties offer attractive and promising results in optimizing CNT as filler in polymer-based composite in achieving stretchable and sensitive strain sensor. However, the challenge is to control the stability of CNT dispersion in water as the nanoparticles often appear to agglomerate quickly due to their strong van der Waals attraction (Htwe and Mariatti, 2022).

Another active element that is expanding in carbon-based ink is carbon black (CB). CB is usually used in various products such as tires, plastics, colors pigmentation, toners and printing inks (Liu et al., 2018). Basically, carbon black has good conductivity as it has large surface area and also has a volume resistivity about 0.1 to 10 Ω cm (Liu et al., 2018). Generally, when CB is in contact with another binder, the electrical conductivity is reported to be improved as a large number of conductive channels are formed. The conductive network also was not easy to ruin as the higher the CB structure, the easier the access to form network space. This can be supported by paper published by Bhagavatheswaran et al. (Bhagavatheswaran, 2015) who successfully constructed a strong filler-filler by using synergetic effect of micro-silica fume and CB as the active element for the printing ink. Due to all of these good conductive properties, CB is widely used in fabricating a strain sensor (Lian, 2022); (Liu et al., 2018); (Hu, 2021).

Table 1 tabulates the comparison between several carbon-based fillers that have been widely used in the printed strain sensor fabrication using carbon-based ink.

2.1.1. Green synthesis of carbon-based conductive fillers

As mentioned previously, selection of conductive filler is important as it is responsible in determining the electrical properties of the synthesized ink. However, the production of carbon-based fillers such as graphene and CNT towards the environment still needs to be attended in achieving an eco-friendly carbon-based conductive ink. The usage of natural precursors which are environmentally friendly and renewable need to be considered as an alternative for fossil-based precursors which are commonly used in the production of carbon-based fillers. Carbon-based filler can be produced either through laser ablation/evaporation, arc discharge method, electrolytic methodologies and chemical vapor deposition (CVD) (Rahman, 2019; Mubarak et al., 2014; Qasim et al., 2023). Parsons et al. (Parsons et al., 2015) studied the impact of CNT production towards environment using selected categories suggested in International Reference Life Cycle Data System (ILCD) which covers global warming, ecotoxicity, human toxicity cancer effects and respiratory inorganics. As the result, this work reported major environmental impact which is more than 92 % of percentage contribution by CNT production, which each detail was further tabulated in Table 2 (Parsons et al., 2015). This can further be supported by Temizel-Sekeryan et al. (Temizel-Sekeryan et al., 2021) which stated that CVD method used to produce CNT contributed highest impact towards all ILCS categories except for ozone depletion.

In producing carbon-based filler such as graphene and CNT, the involvement of precursor is crucial as it will determine the growth of the carbon-based filler (Saputri, 2020). However, the usage of fossil-based precursors such as methanol, toluene, propane and ethylene can pose a significant disadvantages such as non-renewable, high cost and harmful to health (Saputri, 2020); (Paul and Samdarshi, 2011). To combat this issue, the usage natural precursors in carbon-based filler production were explored widely among researchers (Roshni and Ottoor, 2015; Bernd et al., 2017; Asnawi et al., 2018; Hu et al., 2016). Natural precursors will fulfil the aim to provide greener alternatives for cheap raw materials. Plus, natural precursors are renewable, which have the potential for industrial production as the source will not be depleted. Improvised processes of CVD method and spray pyrolysis method has been implemented to achieve environmental friendly production of graphene or CNT using natural precursors (Paul and Samdarshi, 2011); (Szabó et al., 2010); (Hamid et al., 2017). For instance, high quality graphene grown on silicon substrate using natural extract from tea tree plant (*Melaleuca alternifolia*) was produced by Jacob et al. (Jacob, 2015). In this work, a low cost PECVD process was used and no catalyst was used within graphene developing process (Jacob, 2015). Other than tea

Table 1
Characteristic comparison between several carbon-based conductive filler (Yi et al., 2022) (Klemens, 2000) (Balandin, 2011) (Azizi et al., Dec. 2019).

Type of conductive carbon filler	Young Modulus (GPa)	Tensile strength, (GPa)	Poisson ratio	Resistivity, (10^{-2} μΩm)
Graphene	1000	130	0.36	0.01
Carbon nanotube (MWCNTs)	950	10–60	0.34	0.05–0.5
Graphite	100–400	0.013–0.07	0.31	0.3–6
Carbon black (CB)	80	0.002–0.024	0.30	–

Table 2
Percentage contributed by CNT production towards multiple environmental impact categories recommended by ILCD (Parsons et al., 2015).

Environmental impact category recommended by ILCD	Percentage (%) contributed by CNT production
IPCC global warming [kg CO_2-eq]	94
Ecotoxicity for aquatic fresh, USEtox [CTUe]	98
Human toxicity cancer effect, USEtox [CTUh]	99
Respiratory inorganics [kg PM2,5-eq]	92

tree extract, soybean oil was used by Seo et al. (Seo, 2017) as precursor in synthesizing single to few layers of graphene through CVD process. From these previous works, it can be deduced that production of carbon-based filler can be obtained through methods that meet the eco-friendly requirement.

Substituting fossil-based precursor with oils in producing CNT is also one of the method used in achieving eco-friendly CNT production (Qasim et al., 2023). Hamid et al. (Hamid et al., 2017) successfully produced CNT using coconut oil as precursor substituting methanol. In this work, coconut oil were mixed with nickel chloride ($NiCl_2$) at 900℃ using spray pyrolysis method (Hamid et al., 2017). Fig. 3 demonstrated the Raman analysis which I_D/I_G ratio calculated for methanol recorded at 0.9 (Fig. 3(a)) while coconut oil recorded at 0.93 (Fig. 3(b)) (Hamid et al., 2017). This result indicates that successful production of CNT can be achieved by substituting fossil-based precursor with natural precursor. In addition, Kumar et al. (Kumar et al., 2011) reported to produce MWCNT with inner diameter between 15 nm to 30 nm using neem oil as precursor mixed with ferrocene mixture at 825℃ (Kumar et al., 2011). Hence, the usage of carbon-based as conductive filler in synthesizing the eco-friendly carbon-based conductive ink for printed strain sensor can be achieved and needs further research in this area.

2.1.2. Recyclability of the carbon-based conductive ink

Although an abundance of efforts has been made to ensure the eco-friendliness of synthesized carbon-based ink, the recyclability of printed ink or electronic waste (e-waste) issues still need to be addressed. This is because e-waste contribute to substantial pollutants towards environment (Williams et al., 2021). Williams et al. (Williams et al., 2021) reported recyclable water-based CNT printable inks using nanocellulose as polymer binder. In this work, the authors successfully demonstrated good recyclability having density of printed recycled CNT which only differs by ∼ 10 % compared to printed new CNT ink as shown in Fig. 4(a) (Williams et al., 2021). This can be further supported by UV–Vis analysis showing minimal loss of CNT dispersion with recovery yields at 95 ± 6 % for printed CNT ink as shown in Fig. 4(b) (Williams et al., 2021). This reduction may occur due to incomplete removal of CNT leaving residues from substrate during the recycling process. However, the conductivity of recycled ink may slightly deteriorate as the density of dispersed CNT is lower compared to new ink. Besides, Zhang et al. (Zhang et al., 2020) achieved almost identical conductivities for all 3 recyclable PI-MWCNT ink compared to new ink, as shown in Fig. 4(c). This work incorporated MWCNT as fillers with polyimine vitrimer (PI) as binder, which is also eco-friendly binder materials (Zhang et al., 2020). Hence, these work proved that the printable carbon-based inks are capable of being recycled at the end of their service life, especially the carbon fillers component. This is significant as recycling carbon fillers can greatly reduce the needs of production of carbon filler, reduce cost and thus reducing the environmental impact. This is in line with Eckelman et al. who stated that the production of carbon fillers such as CNTs gives higher impact towards environment compared to the impacts of releasing them at the end of their service life (Eckelman et al., 2012).

2.2. Biopolymer binder

Selection of biopolymer binder is significant in binding the particles element together and determine the ink rheological properties according to printing techniques. Typically, biopolymer binders were usually insulators. Therefore, it is important to obtain optimum filler/binder ratio as it will influence electrical, mechanical and desired rheological properties (Franco et al., 2020).

Chitosan (CS) is one of the biopolymers that attracted researchers to be incorporated into printed ink formulation. It is a linear polysaccharide and was produced by alkali deacetylation of chitin obtained from exoskeletons of edible marine crustaceans such as shrimps and crabs. CS exerts significant physical properties that provide antimicrobial (Je and Kim, 2012), great biocompatibility, non-toxic (Park et al., 2015). Liu et al. (Liu et al., 2018) successfully fabricated CS/carbon black ink composite for human motion monitoring. The bending and unbending test of human arm displayed stable signal and good sensitivity, which indicated by successful re-establishment of conductive network upon multiple bending (Fig. 5(a)). The fabricated strain sensor showed good sensing repeatability of 200 bending-recovery cycles without apparent resistance change, as shown in Fig. 5(b) (Liu et al.,

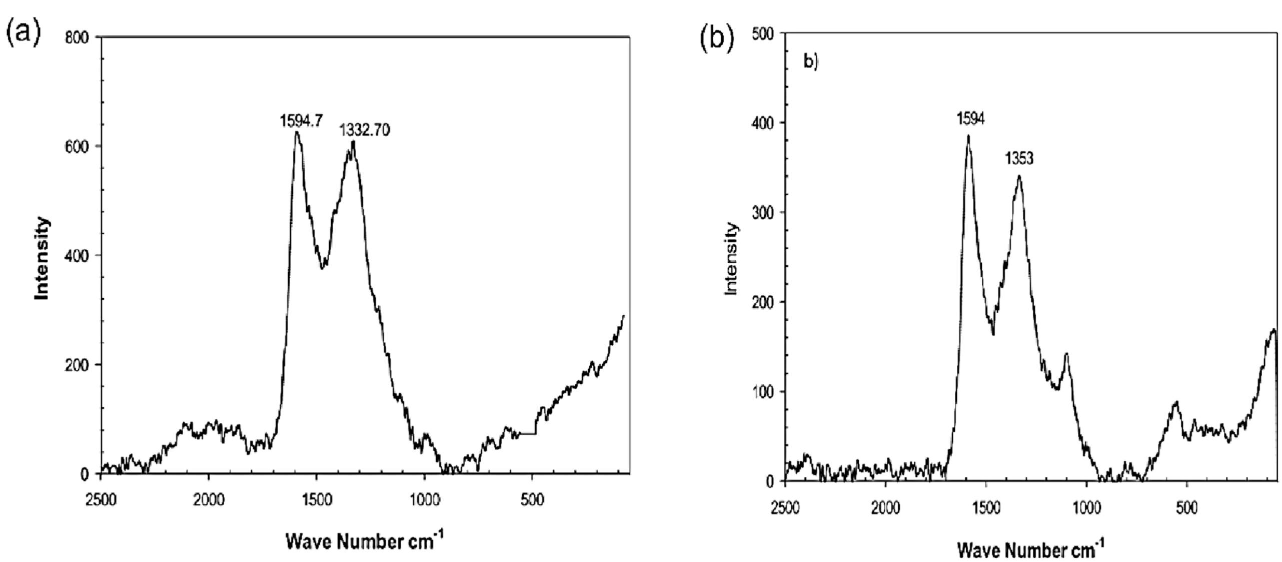

Fig. 3. Raman spectra of CNT synthesized using (a) methanol as precursor (b) coconut oil as precursor (Hamid et al., 2017).

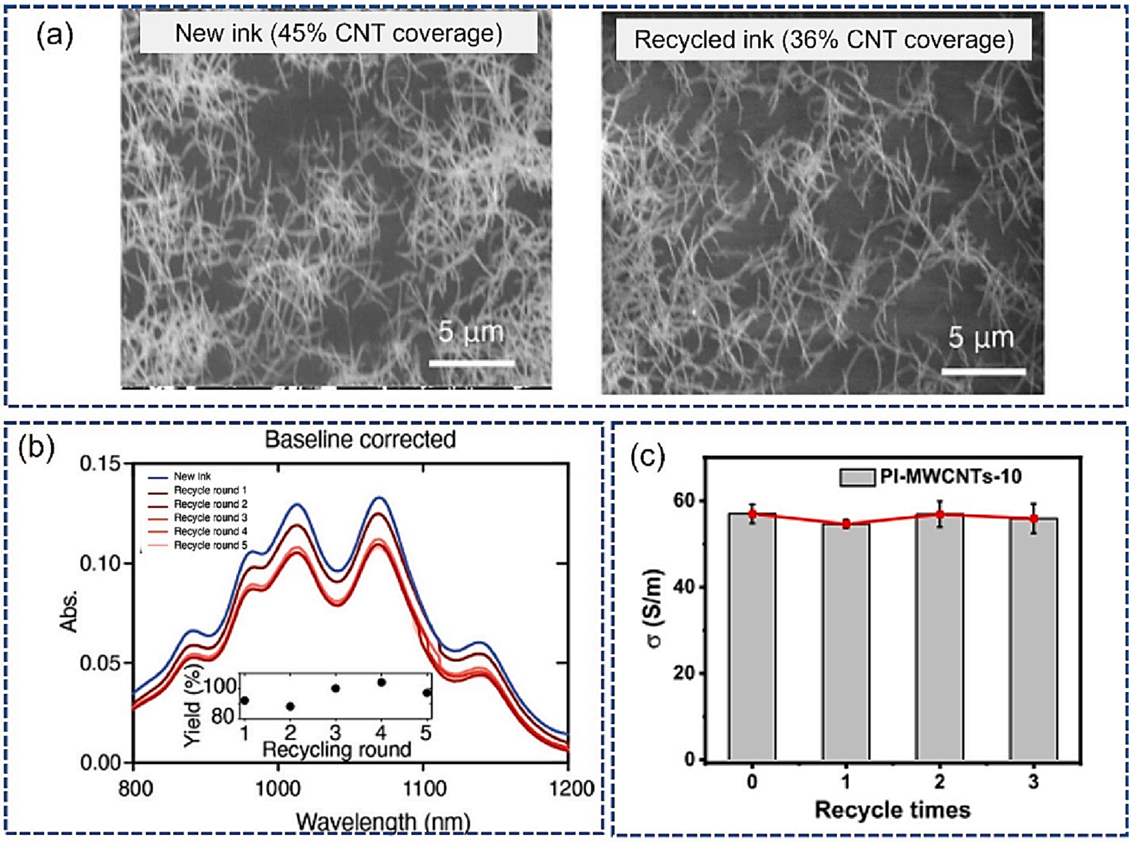

Fig. 4. Recyclability test on printable CNT ink (a) SEM images of CNT dispersibility in new printed CNT and recycled CNT ink (b) UV–Vis analysis comparison result towards new CNT ink and 5x iteration of recycled CNT ink (Williams et al., 2021) (c) Electrical conductivity of new and recycled PI-MWCNT ink (Zhang et al., 2020).

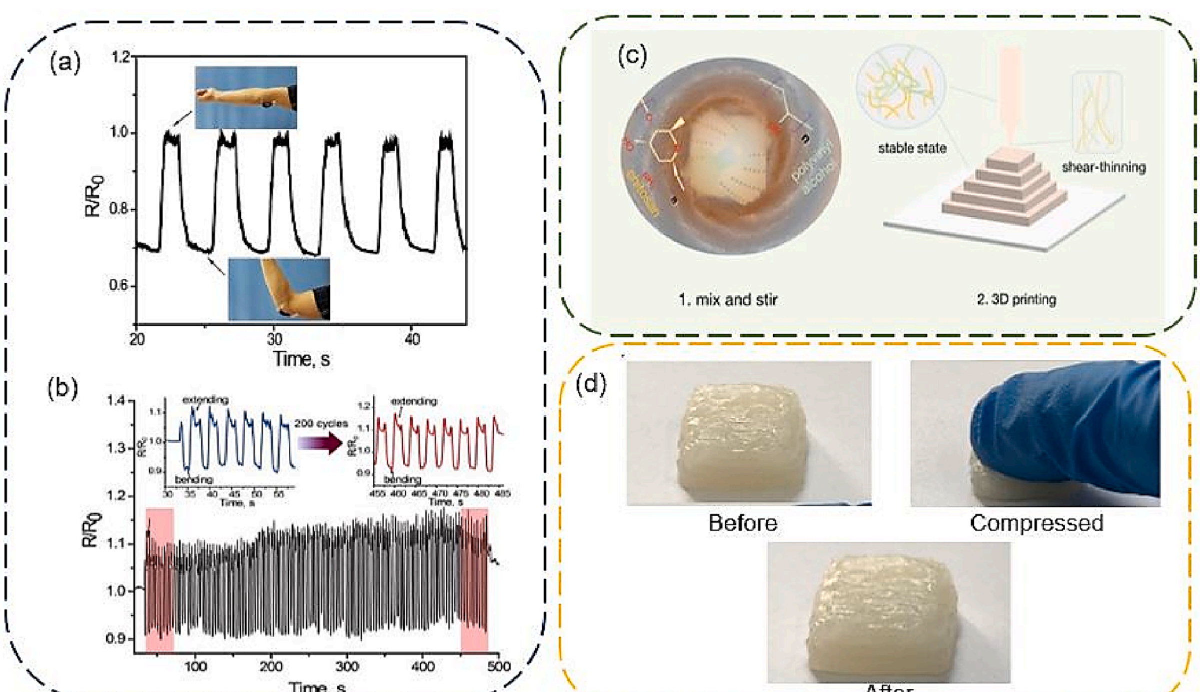

Fig. 5. (a) Stable signal obtained from CS/carbon black ink composite for towards human arm bending (b) 200 cycles of cyclic test of CS/carbon black ink composite during bending (Liu et al., 2018) (c) Schematic illustration of CS assisted by PVA ink synthesis (d) Compressive recovery test towards 3D printed CS hydrogel (Hao and Maimaitiyiming, 2022).

2018). The incorporation of CS/graphene ink composite was also reported by Hao et al. (Hao and Maimaitiyiming, 2022) which developed CS/graphene composite hydrogel ink which demonstrated excellent strain detection (>400 %) and good compressive recovery via 3D printing. The CS hydrogel was reported to have smooth printing without clogging the nozzle at room temperature. However, polyvinyl alcohol (PVA) was required in this work to improve the rigidity of the printed CS hydrogel (Hao and Maimaitiyiming, 2022) as shown in Fig. 5(c) and Fig. 5(d).

Another biopolymer binder that has been utilized in printed ink formulation is carboxy methyl cellulose (CMC). CMC is derived from natural cellulose, and it is readily soluble in water or alkaline medium (Brandley et al., 2018). Due to its unique properties such as thickening, stabilizing and binding properties, it is suitable to be employed as binder to formulate conductive printable ink as it can provide desired ink viscosity according to the printing technique and application (Franco et al., 2020); (Brandley et al., 2018). Besides, shellac has been widely used as biodegradable binder alternative for petroleum-derived binders. Shellac is water insoluble that able to enhance flexibility of printed sensor due to its natural plasticizers properties (Poulin et al., 2021). Poulin et al. (Poulin et al., 2021) fabricated conductive carbon ink dispersed ink shellac polymer. This work reported the highest electrical conductivity of 1000 S/m indicating integration of shellac does not deteriorate electrical properties of carbon particles. However, this work exhibited low strain detection reported at only 6 % strain (Poulin et al., 2021), indicating low stretchability for stretchable strain sensor applications.

2.3. Solvent

Solvent on the other hand provides solubility for the binder that will allow the ink to flow easily. Generally, there are two types of conductive carbon-based conductive ink which are water-based and solvent-based ink. The composition of water-based and solvent-based were illustrated in Fig. 6.

2.3.1. Water-based ink

Water-based ink or also known as aqueous ink, uses water as its primary solvent. The ink contains dye, pigment or can also be combination between both dye and pigment. Water-based ink started to grasp attention as it acts as a green alternative towards human and environment. Décor printing and flexible plastic packaging are some of the applications that used water-based ink (Saidina et al., 2019). It is worth noting that water-based ink usually requires co-solvent and additives which are important for its curing. This is due to some of its drawbacks such as volatility of the ink in an open system such as screen printing and inkjet printing application. Water-based ink has high tendency to dry onto the substrate, commonly on non-absorbent substrates (Saidina et al., 2019). Therefore, integration of additives such as surfactant and dispersant are crucial as it can improve the wetting properties required according to each printing technique. He at al. (He et al., 2022) compared performance various dispersant towards MWCNT-based ink stability which consist of polyvinylpyrrolidone (PVP), cellulose nanocrystal (CNC) and chitin nanocrystal (ChNCs). ChNCs demonstrated the most stable dispersion within MWCNT having dispersion efficiency of 91.1 % compared to other dispersants (Fig. 7(a)). Plus, Fig. 7(b) demonstrated ink viscosity for ChNCs/MWCNT composite ink was suitable for screen printing applications as it is in range of 1–10 Pa.s, and proved that presence of MWCNT did improved the ink viscosity (He et al., 2022). In 2021, Htwe et al. (Htwe and Mariatti, 2021) reported effect of surfactant towards wetting properties of ink on PET substrate. All surfactant used in this work recorded good wetting properties as it is below 90° as shown in Fig. 7(c). However, graphene assisted with PVP surfactant showed lowest contact angle at 16°, indicating best adhesion towards PET substrate compared to other surfactants. This resulted in great printed traces and graphene/PVP assisted ink displayed great flexibility as illustrated in Fig. 7(d).

2.3.2. Solvent-based ink

Solvent-based ink mainly consists of pigment and alcohols. It can be categorized into two which are aggressive solvent-based inks and eco-solvent based inks. Aggressive-solvent based ink used chemical solvent that emits harmful volatile organic compound (VOC) such as toluene and benzene (Pellis et al., 2019). Meanwhile, eco-solvent based ink used mild chemical solvent such as tetrahydrofuran (Thiyagarajan et al., 2019); (Pellis et al., 2019). However, eco-solvent based ink is still facing environmental issues as it is considered hazardous and harmful to humans and the environment. Joo et al. (Joo et al., 2022) demonstrated fabrication of flexible strain sensor using several different solvents

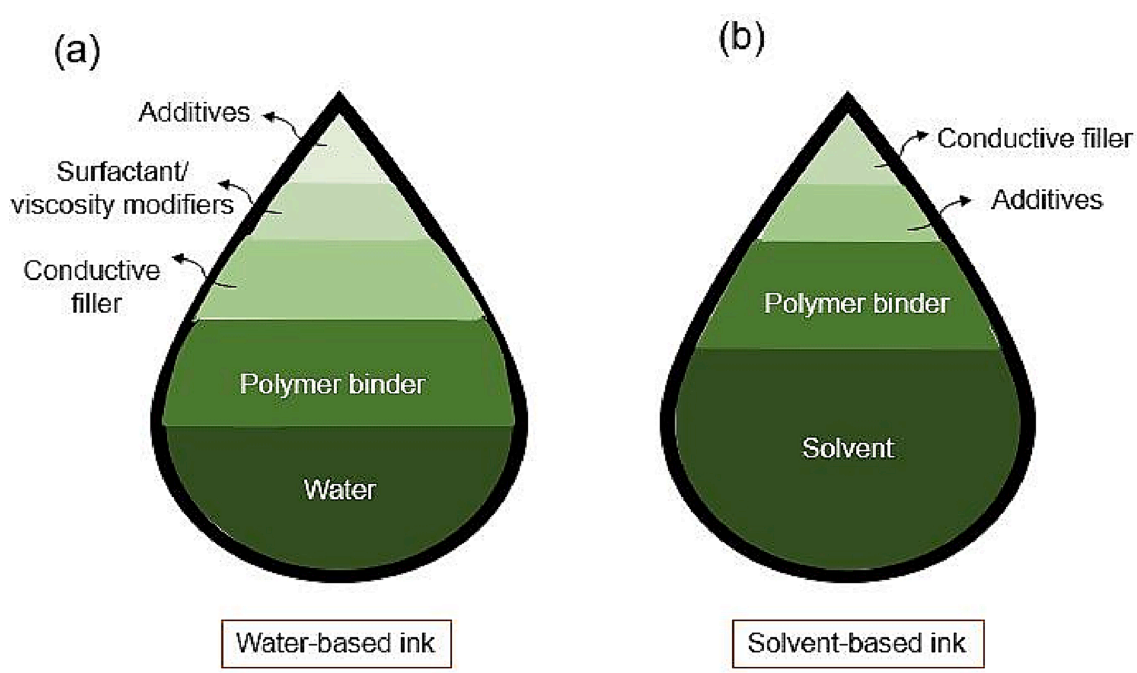

Fig. 6. Schematic illustration of ink composition (a) Water-based ink (b) Solvent-based ink (Sanchez-Duenas, et al., 2023).

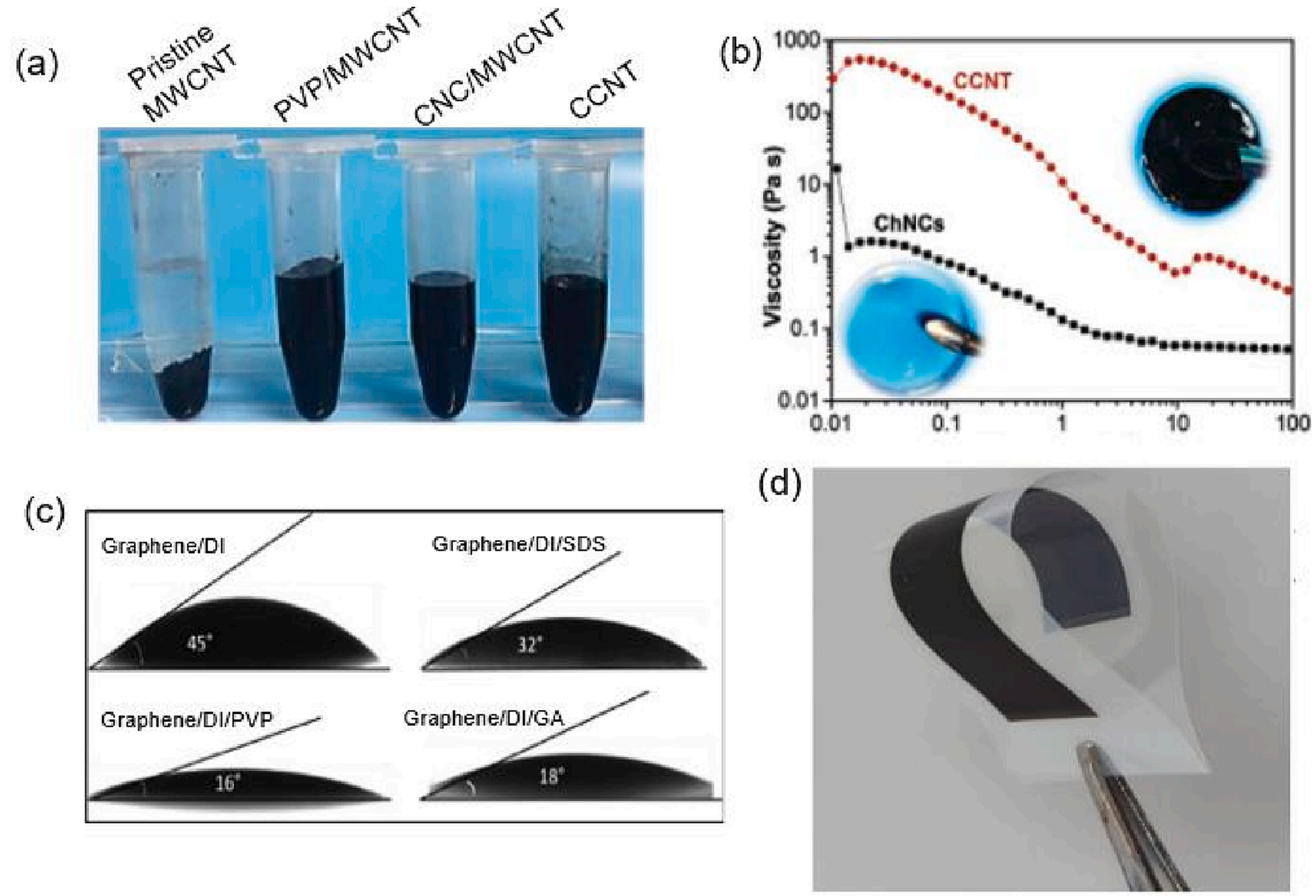

Fig. 7. (a) Ink stability of different dispersant used for water-based MWCNT ink (b) Rheology analysis towards ChNCs and CCNT composite ink (He et al., 2022) (c) Contact angle analysis towards different surfactant assisted water-based graphene ink (d) Printed graphene/PVP ink bent for nearly to 90° (Htwe and Mariatti, 2021).

which include isopropyl alcohol (IPA), chloroform, toluene, 1-methyl-2pyrollidone (NMP) and hexane. NMP was reported to have most stable dispersion of MWCNT as compared to other solvents (Fig. 8.(a)) and recorded lowest sheet resistance at 419 Ω/□ (Joo et al., 2022). However, IPA solvent was chosen in this work as acrylic mold was used for the strain sensor fabrication. This is because NMP was reported to melt the acrylic throughout the sensor fabrication. Plus, TEM images in Fig. 8 (b) illustrated severe entanglement of MWCNT in hexane, toluene and NMP compared to IPA (Joo et al., 2022).

2.4. Additives

Additives play the role in giving the ink their healing and stretching properties. In developing the ink for printed sensor, the viscosity of the ink must be low to attain a good printing quality (Hu, 2022). Ecoflex (Szabó et al., 2010), ethanol (Hamid et al., 2017) and fluorine surfactant (Jacob, 2015) are some examples of additives that have been used to improve the ink properties. The selection of additives to be used will depend on the type of inks formulated as they provide different functions which cover them to act as the dispersants (Fu et al., 2019), surfactant (Jacob, 2015) or as an adhesion promoter (Hamid et al., 2017).

Secor et al. (Secor et al., 2013) demonstrated stable graphene conductive ink for printed electronics using ethyl cellulose (EC) as stabilizing agent. The printed graphene ink assisted by EC exhibits conductivity of 2.5 x 10^4 S/m annealed at 250°C for 30 min. In 2017, Secor et al. (Secor, 2017) then further investigated on the effect of stabilizer using nitrocellulose (NC) in graphene ink synthesis to compare with EC stabilizer used in previous work. A Scotch™ tape test was conducted to compare the effect of EC and NC as stabilizer towards the adhesion of printed graphene ink. As shown in Fig. 9(a), printed graphene/NC ink does not leave any visible residue while residue was observed for printed graphene/EC. To support this result, water sonication test was conducted for both annealed graphene/EC and graphene/NC on glass by immersing both printed ink in water in ultrasonic bath (Secor, 2017). This test was conducted to investigate the ability of printed graphene ink to withstand harsh conduction. As shown in Fig. 9, delamination and disintegration of annealed graphene/EC film was observed within 10 s of ultrasonication condition. In contrast, graphene/NC film shows minimal film breakage or delamination in 60 s, indicating great adhesion between graphene film-glass as no residue left on the tape (Secor, 2017). This work concluded that utilization of NC as stabilizer showed better results in terms of adhesion and electrical properties compared to using EC.

3. Performance of carbon-based conductive ink

The performance of carbon-based conductive ink will be discussed in terms of the percolation theory and tunneling effect that form the conductive pathway in microscopic level. For specific application such as strain sensors, the carbon fillers will undergo orientation as it will experience stretching and relaxing phenomena. This section will discuss on the different optimal carbon filler loading/weightage for different printing techniques after stretching and relaxing phenomena.

3.1. Percolation theory and tunneling effect

Percolation is one of the fundamental theories that are involved in the electrical conductivity in a filler/polymer composite. The changes in electrical conductivity occurred due to the integration of conductive filler into a polymer matrix, resulting in the transition from insulator to

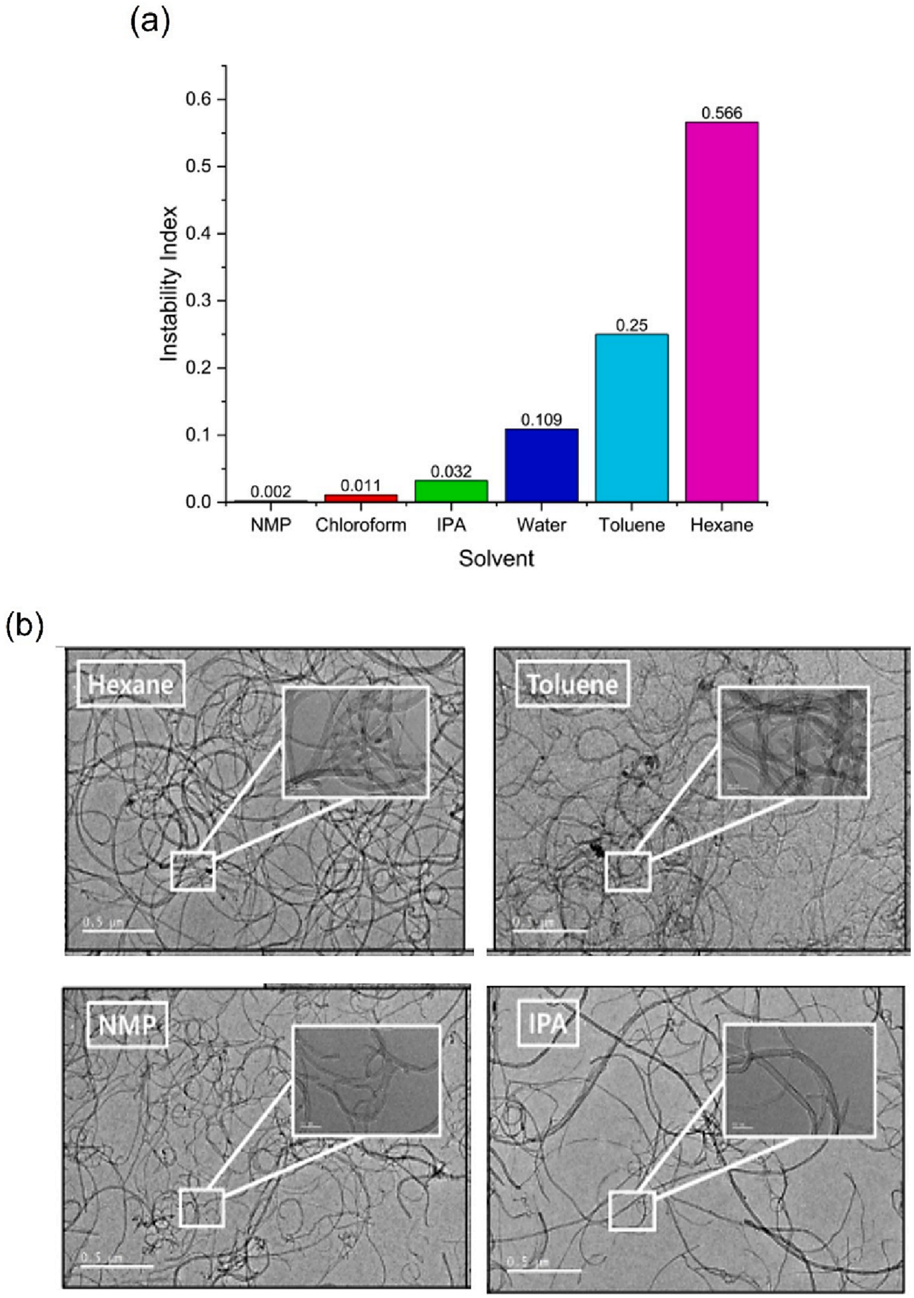

Fig. 8. (a) Instability index of MWCNT ink dispersion stability analysis in different solvents (b) TEM images of MWCNT dispersion in NMP and IPA solvent (Joo et al., 2022).

conductor. This insulator-conductor transition relates with the two key mechanisms involved in filler/polymer composite which are, the non-ohmic condition and the ohmic condition, as shown in Fig. 10(a) (Choi et al., 2019). Firstly, the non-ohmic condition occurs in the early stage before the percolation takes place. The electrical pathway is generated due to the presence of tunnelling effect between reinforced fillers that are close to each other within the polymer matrix. Next, the ohmic condition happens when the dispersed fillers are directly connected with each other, resulting to the formation of network that allow currents to flow easily (Choi et al., 2019). In addition, the integration of the active filler into the polymer matrix will create significant difference between electrical conductivity of the filler/polymer composite which follows the percolation theory (Kanoun et al., 2021).

The schematic diagram of the percolation process that occurs between conductive fillers and polymer matrix is shown in Fig. 10(b). Initially, the conductive fillers started to disperse randomly inside the polymer matrix. Therefore, there are no electrical pathway exist as the conductive fillers were physically disconnected with each other. As the amount of conductive fillers reinforced increase, the distribution of fillers inside the polymer matrix seems to be partially connected due to the occurrence of inter-particle contacts between fillers (Khan et al., 2021). This will result to formation of conductive pathway that allow current to flow through the polymer matrix. Lastly, abundance of conductive paths was constructed as the conductive fillers were continuously added into the polymer. This is because the continuous addition of fillers will allow the fillers to disperse uniformly and result to the fillers to be fully connected with each other (Choi et al., 2019).

As discussed before, the formation of conductive pathway occurs due to the percolation effect. Therefore, it is important to note that there are two key mechanisms that were involved in the percolation phenomena,

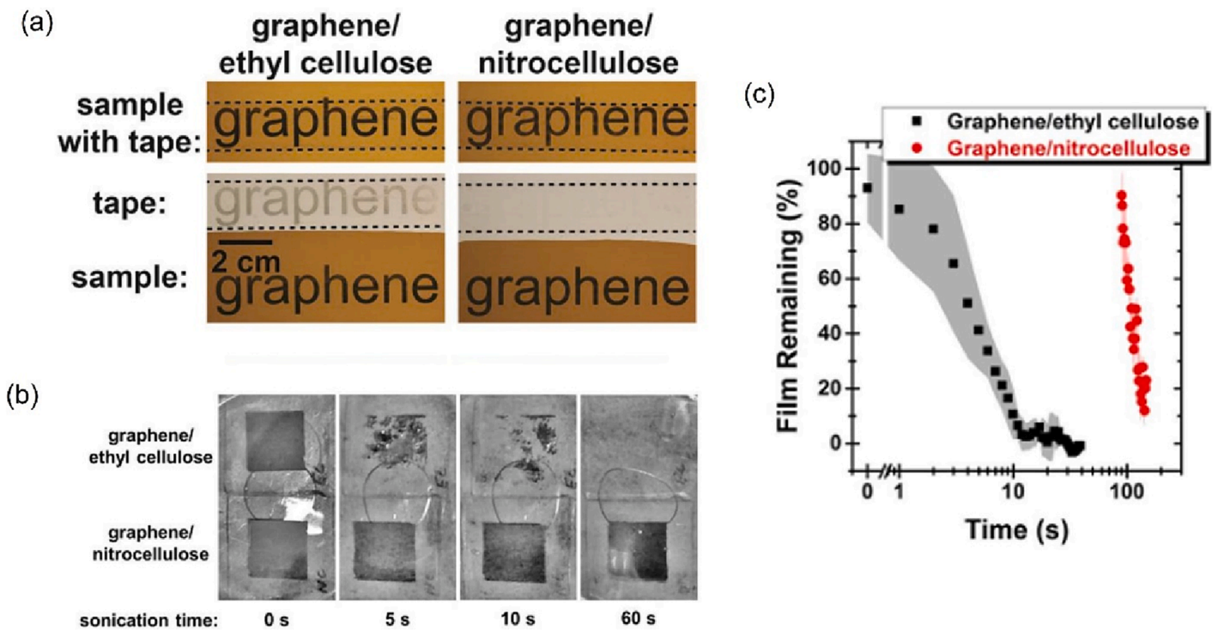

Fig. 9. (a) Scotch^{TM} test tape towards printed graphene ink using EC and nitrocellulose as stabilizer (b) Water-sonication test for graphene/EC and graphene/NC annealed on glass (c) Percentage of remaining graphene/EC and graphene/NC film during ultrasonication process (Secor, 2017).

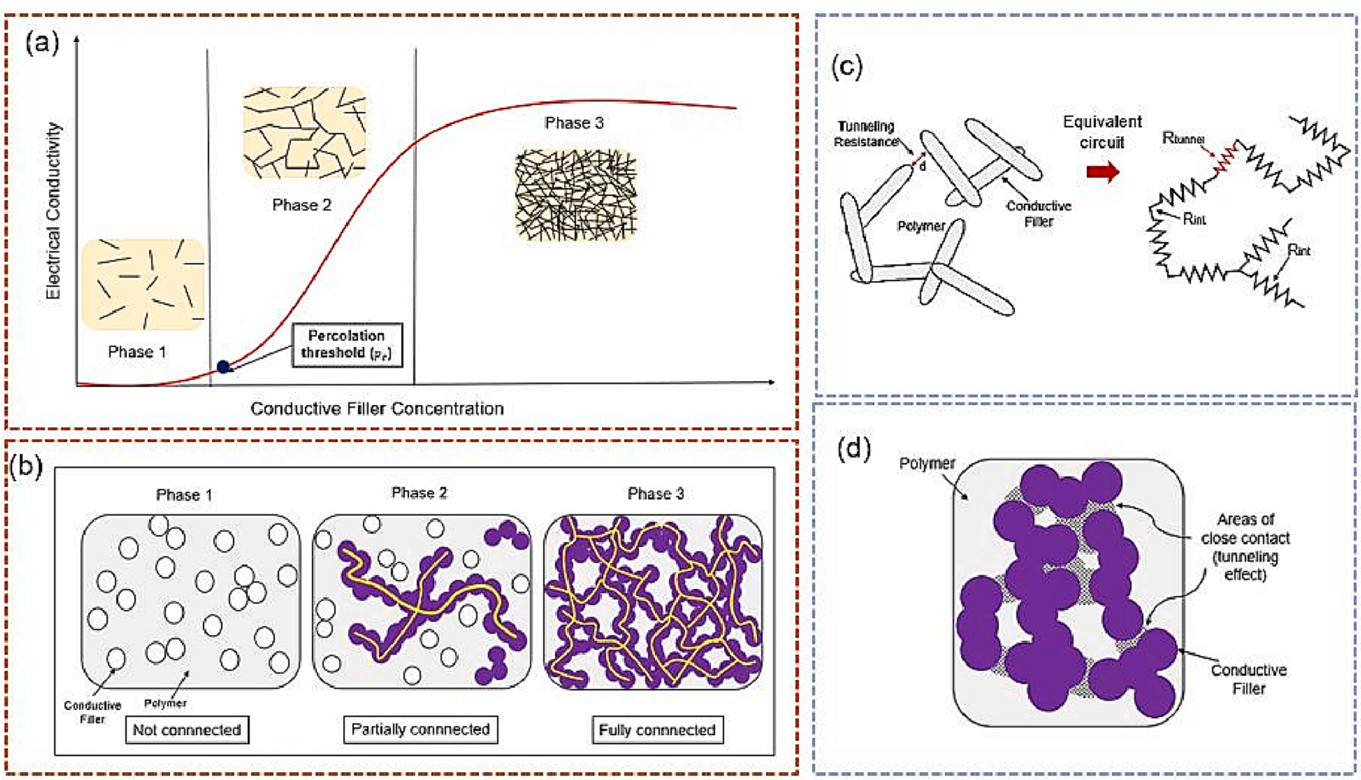

Fig. 10. Schematic illustration of (a) Percolation phenomena (b) Percolation process between conductive filler in polymer matrix (c) Difference between direct contact mechanism and tunneling resistance (d) Tunneling effect phenomena.

which are the direct contact between fillers and the tunnelling effect. According to Singla et al. (Singla et al., 2021), electrical conductivity of filler/polymer composite depends on combination of two resistance which are the intrinsic resistance (R_{int}) and the intertube resistance. R_{int} is the resistance present due to the structure of the filler itself. Meanwhile, intertube resistance refers to the resistance that exists due to the direct contact between two neighboring fillers and also due to the tunnelling resistance (R_{tunnel}). The existence of R_{tunnel} is due to the tunnelling effect that occurs when the area of contact between fillers are separated by a small distance of *d* (Singla et al., 2021). According to tunnelling effect, transportation of electric charge can happen if the gap (d) between fillers is sufficiently close enough towards each other. However, the amount of filler dispersed within the polymer matrix needs to be adequate enough for the electron tunnelling to occur (Liu et al., 2018). Plus, the presence of this R_{tunnel} will affect the electrical conductivity of the filler/polymer composite (Khan et al., 2021)– (Oliva-Avilés et al., 2011). The difference between R_{int} and R_{tunnel} is portrayed in Fig. 10(c) and Fig. 10(d).

The R_{tunnel} can be mathematically expressed using Simmons' formula as shown in Eq. (1) (Hu, 2010);

$$R_{tunnel} = \frac{V}{AJ} = \frac{h^2 d}{Ae^2\sqrt{2m\lambda}} \exp\left(\frac{4\pi d}{h}\sqrt{2m\lambda}\right) \quad (1)$$

where J refers to tunnelling current density, V is the electrical potential difference, e referring to quantum of electricity, m is the electron mass, h the Planck's constant, d is the tunnelling distance between fillers, λ the barrier height of polymer used, and A is the cross-sectional area of the tunnel gap.

As mentioned before, the amount of filler dispersed within the polymer matrix needs to be adequate enough for the electron tunnelling to occur (Hu, 2021). Therefore, it is worth noting that the carbon fillers will undergo orientation as it will experience stretching and relaxing phenomena. Different materials and printing techniques require different optimal filler loading/weightage. For instance, the loading of metal-based filler required for screen printing is between 50 % to 80 % fillers within the ink composition due to its high ink viscosity requirement (Qin et al., 2023). In contrast, optimum metal-based filler loading required by inkjet printing method is only at 30 % to 40 % filler loading within the ink composition, much lower compared to screen printing method (Qin et al., 2023). This differs for carbon-based filler loading where it is only limited to 2 % to 6 % carbon filler loading for inkjet and 3D printing method. Meanwhile, carbon filler loading for screen printing method only required maximum up to 10 % filler loading (Qin et al., 2023). Carbon-based filler required only low filler loading due to their hydrophobicity and tends to agglomerate easily (Qin et al., 2023). Hence, the close relationship between filler loading which covers the orientation of fillers and performance of strain sensor should be studied. This is because the stretching of printed strain sensor will lead to disconnection of electrical pathways between neighboring fillers (Kantarak, 2020). However, the electrical conductivity between neighboring fillers is still available due to occurrence of tunneling effect, as shown in Fig. 11(a). As mentioned before, transportation of electric charge can happen if the gap between fillers is sufficiently close enough towards each other (Hu, 2021). This can be supported by Amjadi et al. (Amjadi and Park, 2015) who investigated on the relative changes of resistance upon strain which correlated with the orientation of CNT fillers within the CNT-Ecoflex nanocomposite ink. As shown in Fig. 11 (b), the CNT-Ecoflex nanocomposite strain sensor broke and losses its conductivity at 1380 % strain (Amjadi and Park, 2015). This happens due to the huge gap of CNT fillers which restricted tunneling effect to occur. Before achieving 1380 % strain, the CNT reorientation eventually changes upon stretching. However, the strain sensor is not broken, and electrical conductivity is still in presence due to the percolation and tunneling resistance between neighboring CNTs. This concluded that percolation and tunneling effect correlate with strain sensor performance, hence require close study between these two parameters.

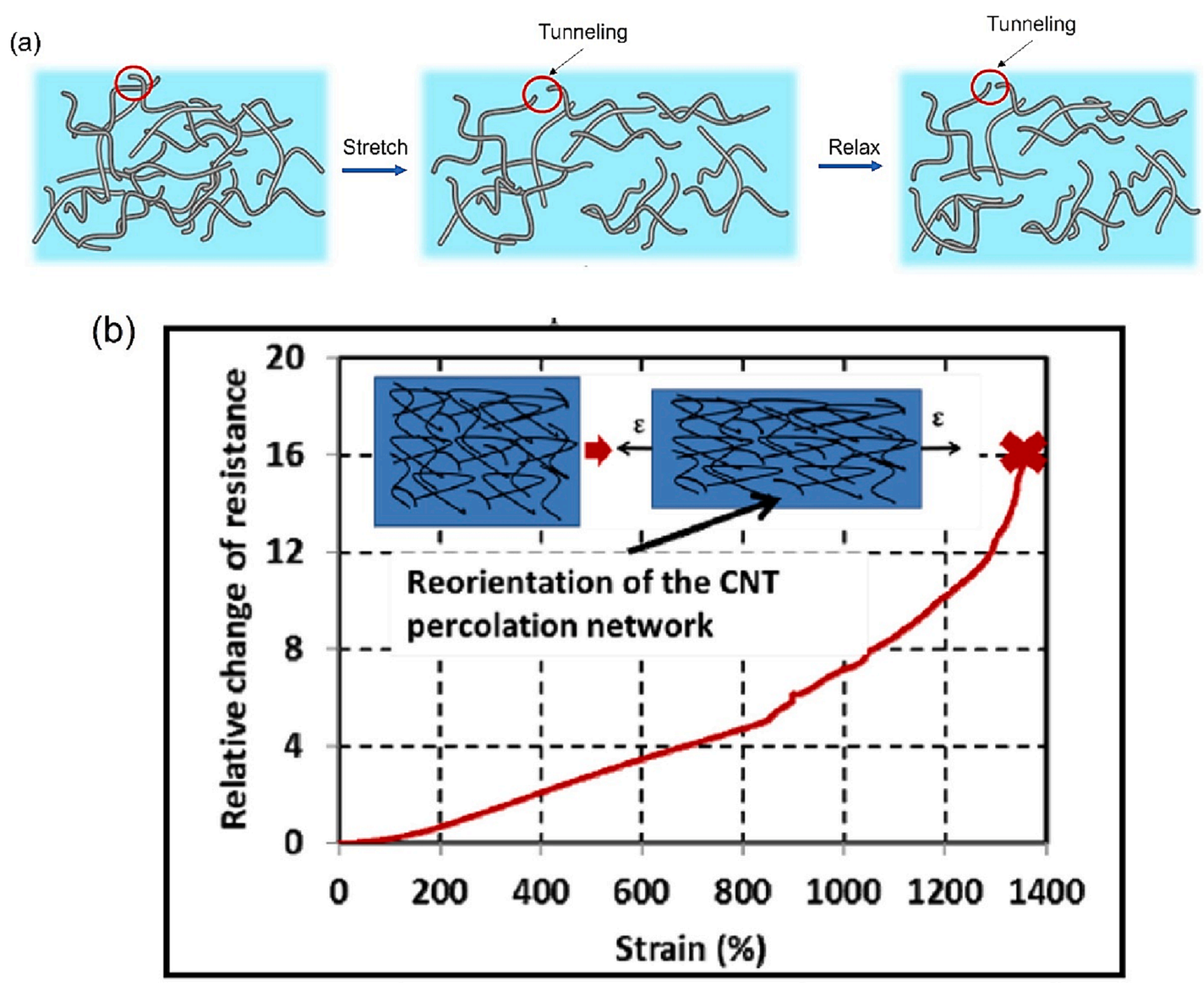

Fig. 11. (a) Schematic illustration of percolation and tunneling effect of carbon fillers during stretching and relaxing (Kantarak, 2020) (b) Strain measurement of CNT-Ecoflex nanocomposite and reorientation of CNT percolation (inset) (Amjadi and Park, 2015).

3.2. Electrical conductivity of conductive carbon-based ink

In fabricating carbon conductive ink for printed strain sensor, the polymer binder properties also play an important role as it will lead to significant impact on electrical conductivity as well as the ink stability. This is because the polymer binder properties will affect the orientation of the conductive fillers which will contribute to lower percolation threshold (Kanoun et al., 2021). Htwe et al. (Htwe and Mariatti, 2021) compared the presence of various surfactant which are sodium dodecyl sulphate (SDS), polyvinyl pyrrolidone (PVP) and gum Arabic (GA) towards water-based graphene ink composite. Based on their findings, graphene ink assisted with PVP surfactant exhibited the highest electrical conductivity (>30 S/m) compared to SDS and GA, which were < 15 S/m. Plus, PVP surfactant demonstrated the most stable ink properties after being stored for one month, having consistent electrical conductivity recorded compared to SDS and GA (Fig. 12(a)). This can be supported by the least sedimentation obtained by PVP-assisted graphene ink compared to other surfactants as shown in Fig. 12(b). This is because PVP consist of pyrrolidone ring and a vinyl group, which makes them able to firmly knitted with the graphene sheets after sonication (Fig. 12(c)) that contribute to excellent dispersion stability (Htwe and Mariatti, 2021).

In addition, optimum filler/binder ratio also needs to be considered in fabricating carbon conductive ink. This is because larger filler content resulted to larger conductive paths, increasing the electrical conductivity of the printed sensor. Franco et al. (Franco et al., 2020) varied filler/binder ratio from 10:90 to 90:10 of graphene/cellulose composite and recorded increasing electrical conductivity upon increasing graphene weightage. Firstly, 10:90 graphene/cellulose weightage recorded 1.03u S/m and the conductivity rose up to 1.11 S/m 30:70 graphene/cellulose composite. A 90:10 graphene/cellulose composite ratio showed slight increase of electrical conductivity recorded at 55.55 S/m, indicating the stabilization of graphene dispersion surpassing the percolation threshold (Franco et al., 2020). This corresponds with the SEM images illustrated in Fig. 11(d) indicating increasing conductive path upon larger graphene weightage. The same finding was reported by Poulin et al. (Poulin et al., 2021) who dispersed conductive carbon (graphite/carbon black) particles in shellac, which act as biopolymer binder. This work reported a sharp increase of conductivity from 100 S/m to 1000 S/m at filler/binder ratio of 1.5 (Poulin et al., 2021). Therefore, optimum amount of filler/binder ratio is crucial as it will govern the electrical conductivity and performance of printed sensors.

3.3. Rheological properties of carbon-based conductive ink

Rheological properties are crucial in printed ink fabrication. Viscosity and surface tension are some of important rheological properties of fabricated ink that need to be evaluated before applying for printed electronics. Rheological properties can be divided into two which are Newtonian fluid and non-Newtonian fluid. A Newtonian fluid behaviour have linear relationship between viscosity and shear rate, while non-Newtonian fluid viscosity varies depending on the shear rate as shown in Fig. 13(a). In fabricating printed ink for stretchable piezoresistive strain sensor, the fluid behaviour is crucial as it reflect the ink viscosity and will affect the adhesion between deposited ink towards the substrate.

Different fabrication process requires different rheological properties. For instance, strain sensor fabricated using inkjet printer require viscosity between 0.001 to 0.02 Pa•s, while viscosity range for carbon-based screen-printed strain sensor is in range of 1 to 10 Pa•s, both recorded at shear rate 100 s^{-1} (Menon et al., 2017); (Dybowska-Sarapuk, 2018). Menon et al. (Menon et al., 2017) fabricated a MWCNT screen print ink and obtained viscosity of 2.09 Pa•s at shear rate 100 s^{-1}. Bougot et al. (Bougot et al., 2013) studied on the dispersion of carbon nanotubes (CNT)/chitosan composite for inkjet printing and obtained ink viscosity of 0.002 to 0.018 Pa•s. However, this paper changes the polymer matrix from chitosan to sodium dodecyl sulphate (SDS) due to ejection failure of the CNT/chitosan ink from the nozzle as chitosan macromolecular chain seals the nozzle (Bougot et al., 2013). This indicate that selection of materials give impact towards the fluid behaviour of ink towards different printing techniques. This can be supported by He et al. (He et al., 2022) who reported on integration of MWCNT within chitin nanocrystal (ChNCs) polymer matrix did improved the ink viscosity as depicted in Fig. 13(b). This happened due to the high aspect

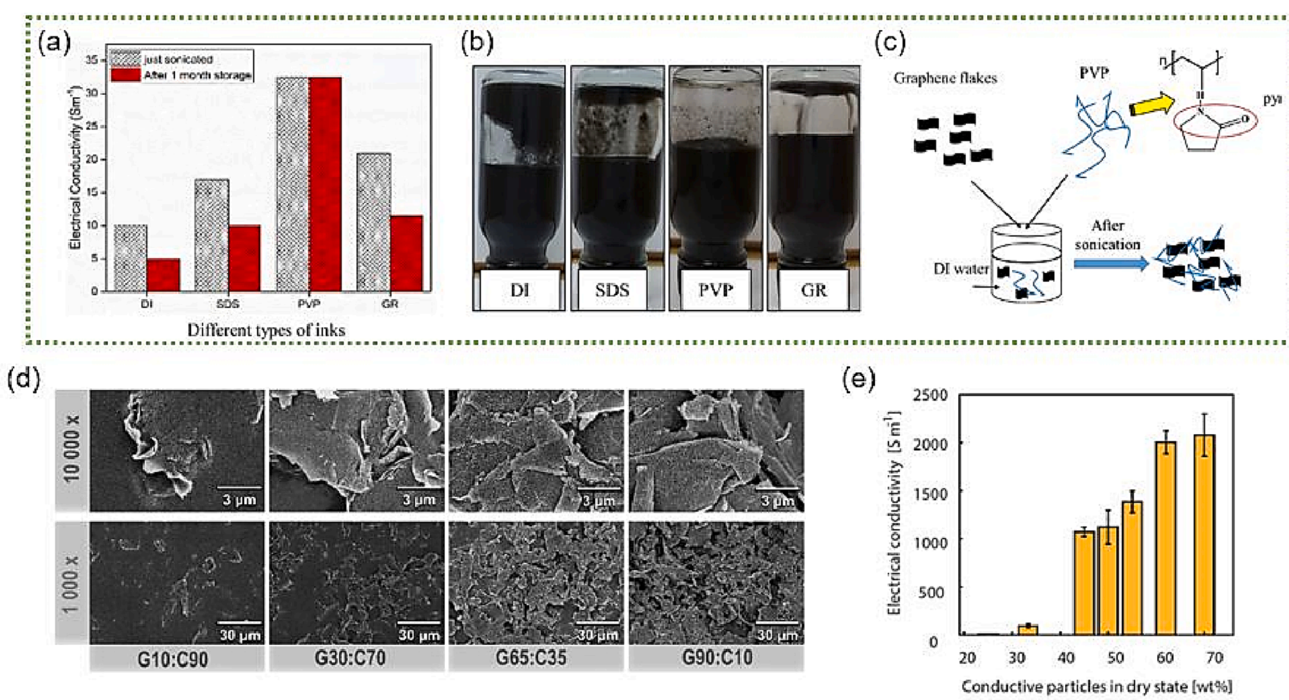

Fig. 12. (a) Schematic illustration on ink component distribution and electrical percolation upon solvent evaporation (b) Electrical conductivity upon different filler weightage (Poulin et al., 2021) (c) SEM images of prepared ink with varies filler/binder ratio (Franco et al., 2020).

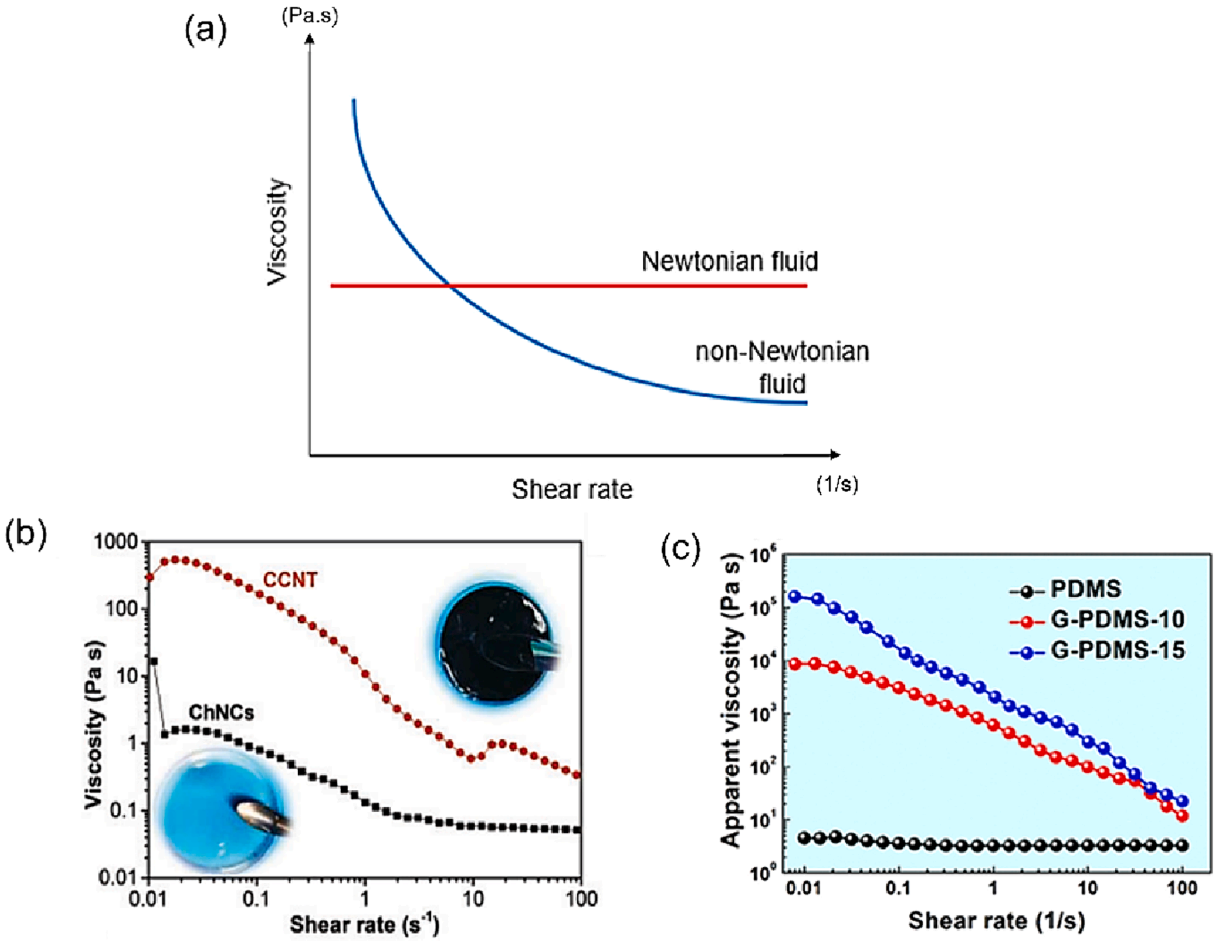

Fig. 13. (a) Newtonian and non-Newtonian fluid behavior (b) Viscosity vs shear rate comparison result of chitin and chitin/MCWNT ink (He et al., 2022) (c) Viscosity vs shear rate for graphene-PDMS composite with various filler content (Ma et al., 2019).

ratio of MWCNT and interaction that occurred between MWCNT and ChNCs. Besides, amount of conductive filler added within the ink synthesis will also impact the ink viscosity. Ma et al. (Ma et al., 2019) evaluated on the ink viscosity towards graphene-PDMS composite ink with varies graphene content at 10 wt% and 15 wt%, denoted as G-PDMS-10 and G-PDMS-15, as shown in Fig. 13(c). In this study, the authors reported that G-PDMS-15 recorded the highest ink viscosity at shear rate 100 s^{-1} compared to G-PDMS-10 and pure PDMS (Ma et al., 2019).

Another factor to be considered in synthesizing the carbon-based/polymer composite ink for printed electronics is the wetting behaviour of ink towards the substrate. The interaction between the ink droplets and the substrate will impact the adhesion between the printed ink towards the substrate. A good wetting behaviour will result in contact angle $< 90^\circ$ while contact angle of $> 90^\circ$ suggest poor wetting behaviour, as shown in Fig. 14. A good wetting behaviour can also be indicated by proper spreading of ink onto substrate while poor wetting behaviour will result to droplet form of ink where the ink is not spread (Zeng and Zhang, 2019). The smaller the contact angle, the lower the surface tension of the substrate, the better the adhesion between the ink deposited towards the substrate. Generally, most of polymer substrates are hydrophobic, where it repels the adhesion forces upon deposited ink

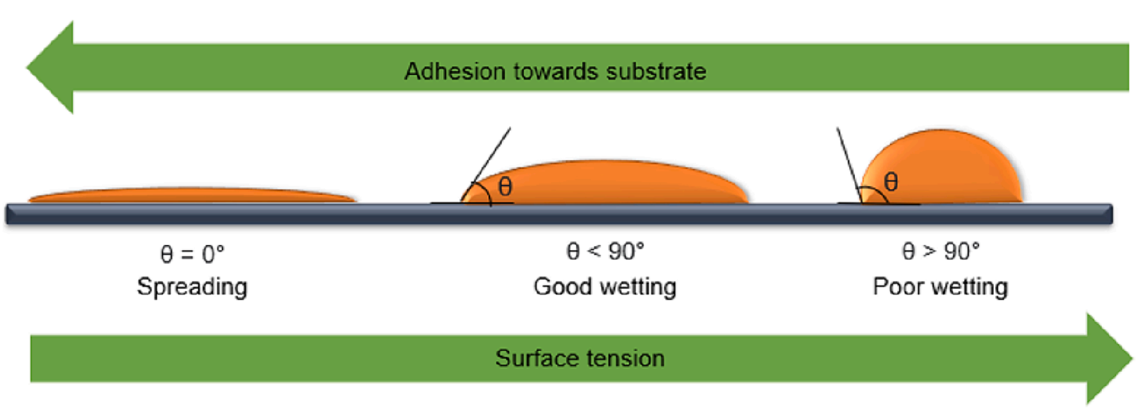

Fig. 14. Wetting behavior of ink droplet on substrate.

Table 3
Carbon-based conductive ink synthesis using eco-friendly materials.

Polymer binder	Conductive filler	Solvent	Additives	Synthesis Process	Filler weight composition/ filler:binder ratio	Conductivity (S/m)	Percolation threshold	Ref
Chitin nanocrystal	MWCNT	Deionized water (DI water)	Chitin itself act as surfactant	Hydrochloric acid hydrolysis method to obtain chitin nanocrystal (ChNCs) suspension. Simple mixing and ultrasonication process for MCWNT-ChNCs composite ink	0.9, 4.5, 6.3, 7.2 wt%	1150	At 0.9 wt% CNT	(He et al., 2022)
Chitosan	Graphite	DI water	Glycerol	Simple mixing and centrifuged	2, 44, 55 % (w/w)	–	–	(Camargo et al., 2022)
Polylactic acid (PLA)	Graphene nanoplatelets	Anisole	Tween-65, Ultra-pure Mili-Q water and sodium carboxymethyl cellulose	Mixed, stirred and ultrasonicated	1:1.5, 1:1, 1:0.5	34.5	At 1:1 filler-binder ratio	(Najafi et al., 2022)
Shellac	Graphite and carbon black	Ethanol or Pentanol	Polyethylene glycol (PEG)	Mixed and ball-milled	20, 30, 40, 50, 60, 70 wt%	1000	At 40 wt% MWCNT	(Poulin et al., 2021)
Nanocellulose	CB	Water and propylene glycol	Glycerol	Vacuum pump to exchange the solvent from water to propylene glycol. All material then mixed using speed mixer	6..08, 6.24, 10.08 wt%	400	–	(Brooke, et al., 2021)
Cellulose	Graphene	Water	Ethanol	Simple mixing and ultrasonicated	0.58—14 wt %	55	At 1.9 wt% graphene	(Franco et al., 2020)
(TEMPO)-oxidized cellulose nanofibrils (TOCNs)	SWCNT	Distilled water	–	Mixed, ultrasonicated and centrifuged	0.02, 0.04, 0.06, 0.08, 0.10 and 0.12 vol%	1000	At 0.4 vol% SWCNT	(Koga et al., 2013)
PDMS	MWCNT	Pentane	–	Stirred, blended and ultrasonicated. Require several steps to remove pentane from composite	5, 7, 9 wt%	69.3 *m*	At 5 wt% MWCNT	(Fu et al., 2019)
TPU	MWCNT	Dimethylformamide (DMF)	–	Ultrasonicated and mechanically stirred	1.5 and 3.0 wt %	0.57 *m*	At 1.98 wt% of MWCNT	(Xiang, 2019)

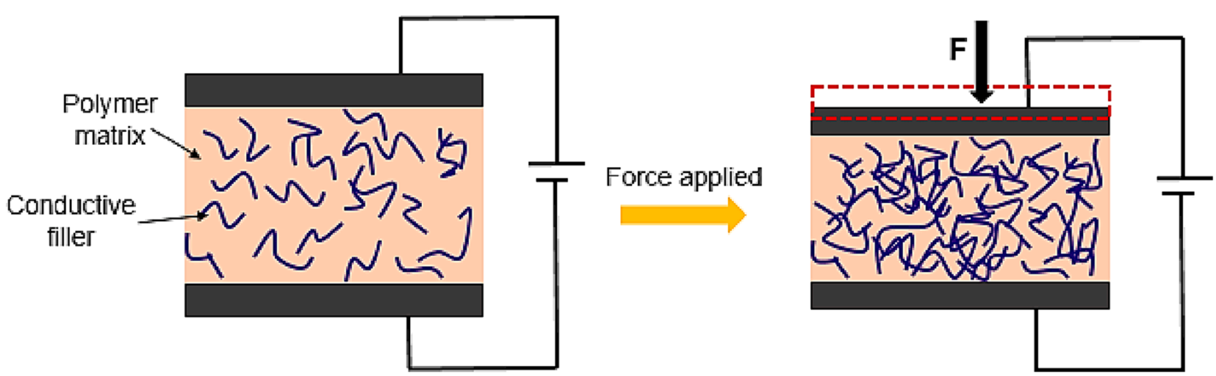

Fig. 15. Schematic illustration on piezoresistive effect mechanism.

(Zeng and Zhang, 2019); (Senthil Kumar et al., 2019). Hence, surface modification is likely required to modify the wetting behaviour of the substrate. There are several techniques of surface modification such as polymer coating or plasma etching, or also known as plasma treatment (Karlapudi et al., 2023); (Sappati et al., 2021). Trantidou et al. (Trantidou et al., 2016) reported successful surface modification on PDMS substrate using two-step method of the deposition of polyvinyl alcohol (PVA) after plasma treatment. The PDMS substrate was firstly exposed to oxygen gases and PVA solution was instantly poured onto the treated PDMS. The plasma treated PDMS substrate achieved hydrophilic surface having contact angle of deposited ink at 24.9 ± 0.4°, indicating good wetting behaviour (Trantidou et al., 2016). Hence, the material selection of substrate and the amount of polymer binder used within the ink synthesis are crucial to be considered as it will affect the ink viscosity and the wetting behaviour of ink towards substrate.

3.4. Summary of previous work on the synthesis of carbon-based conductive ink using eco-friendly materials

The emergence of greener alternatives using eco-friendly conductive carbon-based materials started to be implemented in printed electronics application. Based on previous discussion, selection of material and

Table 4
Substrate selection for flexible printed strain sensors (Li et al., 2022).

Classification of Substrate	Substrate name	Advantages/Limitation
Thermoplastic polymer	Polyethylene terephthalate (PET)	Transparency, smooth surface
	Polyimide (PI)	High mechanical and chemical strength for flexible PCB
	Thermoplastic polyurethane (TPU)	stretchable,
Cellulose fibers	Paper	Eco-friendly, available in abundance, low cost but high surface roughness (more than 1)
Silicone Elastomer	Polydimethylsiloxane (PDMS)	Biodegradable, high stretchability

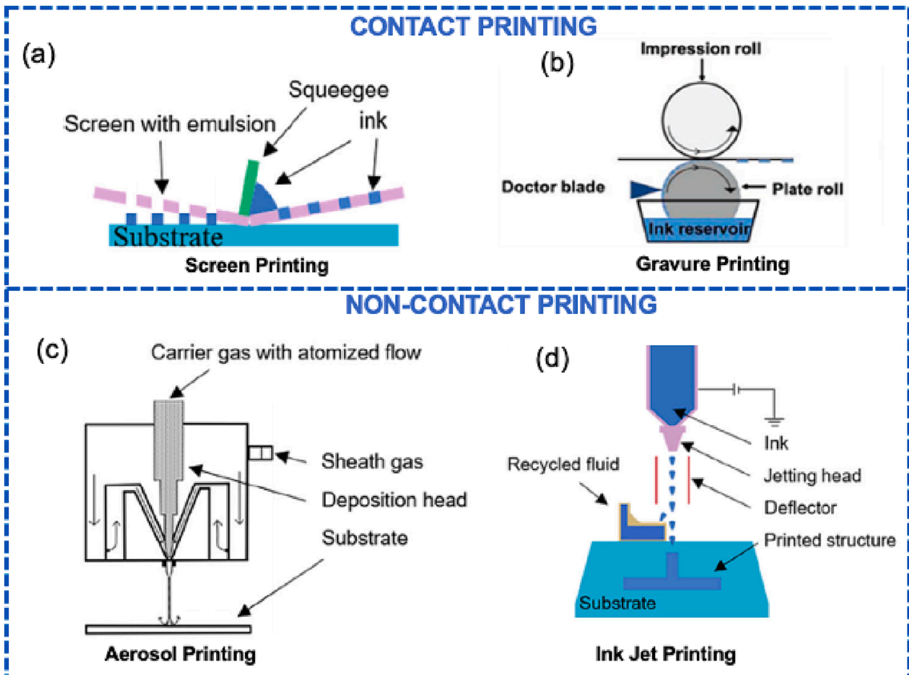

Fig. 16. Printing technology for printed strain sensors using sustainable carbon-based conductive ink (Li et al., 2022).

filler/binder ratio are crucial as it will affect the electrical properties of the synthesized ink. A study by He et al. (He et al., 2022) proved that integration of chitin nanocrystal (ChNCs) as biopolymer binder can eliminate the needs of surfactant in their water-based ink synthesis process as ChNCs itself can also act as surfactant. This is because the amphiphilic properties of ChNCs can reduce the surface energy in aqueous phase, thus enabling them to also act as surfactant in the ink synthesis process (He et al., 2022). This work reported to have excellent conductivity recorded at 1150 S/m with percolation threshold obtained at 0.9 wt% of MWCNT. Several research works have been compiled in Table 3 which demonstrated various eco-friendly material selection comprises of whole ink composition (filler, polymer binder, solvent, additives), ink synthesis process and filler/binder ratio. This shows that material selection will impact the conductivity and percolation threshold obtained for each ink fabrication.

4. Piezoresistive theory of printed strain sensor

Piezoresistivity was rooted from two different linguistic which is Greek and Latin where the Greek word of 'piezo' called 'piezein' and Latin word of 'resistive' called 'resistere' hold meaning of to compress and to stop, respectively (Yazdani et al., 2016). Generally, piezoresistivity was based on the piezoresistive effect. It experiences a change in electrical resistance by transducing the mechanical variations into detectable resistive signal when mechanical stress is applied (Thiyagarajan et al., 2019); (Sanchez-Duenas, et al., 2023). This method was widely used because of its robustness, highly sensitive, low cost, simple structure (Sanchez-Duenas, et al., 2023) and also consume less energy (Lian, 2022). The occurrence of piezoresistive effects exist in three different materials which are metal conductor, semiconductor and polymer composite. The changes in resistance occurred can be influenced by three factors which are the geometrical effect, structural effect and the disconnection mechanism (Thiyagarajan et al., 2019). Geometrical effect refers to the geometrical change of the material used based on Poisson's ratio (υ) fundamental, which explains that materials tend to contract in diagonal direction of stretching when strain was applied (Thiyagarajan et al., 2019). Next, the changes in resistance that occurred especially in semiconductor material were caused by structural deformation. When the stretching of the conductor increases, the cross-sectional area will be reduced resulting in increase of the resistance. In addition, sensitivity which is represented by gauge factor (GF) correlate with the variation of resistance in the case of strain sensor as it depends on the geometry and resistivity of the sensing material. For metal conductor, the changes of initial resistance, denoted by R_0 into new resistance value after applied strain (ε), denoted as R, are crucial indicators in determining the presence of piezoresistive effect. Eq. (2) presents the gauge factor of metal conductor where l is the relative change in length, ρ is the changes of resistivity (Fiorillo et al., 2018).

$$GF = \frac{dR/R}{dl/l} = 1 + 2v + \frac{1}{\varepsilon}\frac{d\rho}{\rho} \tag{2}$$

In contrast with piezoresistive effect in metal conductor, gauge factor (GF) in semiconductor was strongly influenced by the changes of resistivity (ρ) or conductivity (σ). Based on Eq. (3), the change in number of free electrons are related to the conductivity where n is the electron concentrations (Fiorillo et al., 2018).

Table 5
Printing technology for printed strain sensors using sustainable carbon-based conductive ink (Maddipatla et al., 2020; Altay et al., 2020).

	Printing process	Viscosity (Pa.s)	Shear rate (s^{-1})	Description	Pros and Cons
Contact	Screen printing	0.5–60	1–1000	Carbon conductive Paste is transferred by forcing the ink through the screen using squeegee either by semi or fully automated system	*Pros:* High aspect ratio and high viscosity *Cons:* Patterned stencils are needed
	Gravure	0.01–1.1	10–1000	The ink from the reservoir will flood the entire gravure cylinder and the ink is transferred to the substrate with high pressure	*Pros:* High speed of printing *Cons:* Low viscosity and limited range are needed
Non-contact	Inkjet	0.001–0.1	10–50000	Use digital image by dropping the ink onto the substrate by drop on demand technique. Ink needs to be in low viscosities. The droplet is formed by thermal or piezoelectric technique. Maskless fabrication, high print resolution	*Pros:* High speed of printing and maskless *Cons:* Nozzle clogging
	Aerosol	0.001–1	-	Noncontact deposition based on the atomization of the conductive ink to form aerosol that is deposited on the substrate.	*Pros:* Printability on complex nonplanar surfaces Can print wide range of conductive ink, rapid and affordable *Cons:* Contamination

$$GF = 1 + 2v - \frac{1}{\varepsilon}\frac{d(\sigma)}{\sigma} = 1 + 2v - \frac{1}{\varepsilon}\frac{d(n\mu)}{n\mu} \quad (3)$$

Fig. 15 shows the illustration of piezoresistive effect in polymer composite given by Eq. (4) which shows the influence of geometrical changes where R denote the resistance, υ is the Poisson's ratio, ρ is resistivity and ε is the strain applied (Yang et al., 2017)

$$\frac{\Delta R}{R} = (1 + 2\upsilon)\varepsilon + \frac{\Delta\rho}{\rho} \quad (4)$$

When the calculated GF is constant, the dependency of resistance on strain is linear but if it is vice versa, the GF is said to be highly correspond with the tested deformation range (Yang et al., 2017). The difference between GF of strain and pressure sensor is that strain sensor measures the changes of resistance proportion to ε which is the applied strain while changes of resistance in pressure sensor is proportion to P representing the intensity of pressure.

5. Printing technology for printed strain sensors using eco-friendly carbon-based conductive ink

Flexible and printed sensors have been used extensively in many different applications. Printing technology is an additive process where functional conductive ink is deposited selectively on the chosen flexible substrate either in contact or noncontact printing. Printed sensors have the advantage of producing less waste because they do not require the masking and etching that traditional photolithography methods require (Maddipatla et al., 2020). The selection of material for the substrates, the conductive ink, and the printing technique are crucial factors in the fabrication of printed sensors. Smoothness, stretchability, and flexibility are some of the most important factors for the substrates. The most common substrate used are thermoplastic polyurethane (TPU), poly (ethylene terephthalate) (PET) and polydimethylsiloxane (PDMS) and papers as shown in Table 4 (Maddipatla et al., 2020). For contact printing, the ink is transferred by physical contact to the chosen substrate. Some examples of contact printing are screen printing technique (Poulin et al., 2021), gravure (Zhang, 2020) and flexography printing (Higuchi, 2013) as shown in Fig. 16. Meanwhile, for non-contact printing, the ink is transferred via nozzles to the substrate with no physical contact. Aerosol (Li et al., 2014) and inkjet printing (Htwe and Mariatti, 2021) are some examples of noncontact printing as illustrated in Fig. 16. Investigating the rheological features of the conductive ink, such as its viscosity and wetting qualities, is crucial as they have a direct impact on the printing quality. For contact printing, screen printing and flexography are some examples where the ink is transferred to the substrate due to physical contact. Ink jet and aerosol printing method does not have physical contact with the substrate where the ink is transferred via nozzles or openings (Maddipatla et al., 2020). Each printing technique has different requirements for the rheological properties of the ink itself such as viscosity as shown in Table 5.

As mentioned above, different printing techniques require different ink properties. Viscosity is one of the crucial ink parameters to be focused on as it will impact the printed result and performance of strain sensor. However, limited reports on eco-friendly based ink for strain sensor fabrication reported on ink viscosity for their synthesized ink based on the printing technique used. Yi et al. (Yi et al., 2021) obtained carbon-based ink with 0.022 Pa.s for piezoresistive strain sensor using drop casted technique. Meanwhile, screen printed ink synthesized by Poulin et al. (Poulin et al., 2021) exhibited 10 Pa.s ink viscosity. These two studies showed that different printing techniques require different ink viscosity even having same filler type which is carbon black. Performance of carbon-based strain sensor in terms of strain detection and sensitivity for several research works have been compiled in Table 6. All these previous works used eco-friendly material for their strain sensor ink fabrication which covers the polymer binder, solvent and additives.

Table 6
Summary on printed carbon/polymer composite piezoresistive strain sensor performance.

Polymer binder	Conductive filler	Solvent	Additives	Substrate	Sensor fabrication/ Printing method	Strain (%)	Gauge factor (GF)	Ref
Ecoflex	MWCNT	Isopropyl alcohol (IPA)	Silicone oil	Ecoflex	Stencil printing	Up to 955	3–12287	(Hwangbo et al., 2023)
Ethylene glycol	Carbon black	–	–	PDMS	Drop-casted	5	25.2 ± 0.9	(Yi et al., 2021)
Shellac	CB and graphite	Ethanol or Pentanol	Polyethylene glycol (PEG)	Paper	Screen-printing	4 to 6	Not mentioned	(Poulin et al., 2021)
Polyure-thane	Carbon black (CB)	IPA	–	Parafilm	Direct deposition	< 25	4.9 for 0.8 vol%, 2.0 for 1.0 vol% CB	(Sousa et al., 2020)
Chitosan	Graphite and MWCNT	Water	–	Latex	Direct deposition	Up to 150	352	(Tang, 2019)
PDMS	MWCNT	Pentane	Slygard	–	Mould and sandwich method	Up to 40	5 to 9	(Fu et al., 2019)
TPU	MWCNT	–	–	–	3D printing	100	176	(Christ et al., 2017)
–	Graphene	DI water	PVP	PET	Inkjet printing	50	Not tested	(Htwe and Mariatti, 2021)

6. Conclusion and future direction

In this review, comprehensive information on carbon-based conductive ink components for printed strain sensor using eco-friendly materials is presented. The conductive carbon-based ink component compiled in this paper highlight on the eco-friendly materials which includes carbon-based filler, polymer binder, solvent and additives. Greener alternatives are chosen in conductive carbon-based ink synthesis for fabrication of printed strain sensor as incorporation of toxic chemicals in the ink formulation such as organic solvent and additives emits harmful VOCs that can harm both humans and environment. At present, numerous literatures reported on fabrication of printed strain sensor. However, limited studies on implementation on eco-friendly material especially the solvent and additives within ink fabrication. Incorporation of toxic chemicals was usually required to enhance the synthesized ink properties and the performance of printed strain sensor. Thus, greener alternatives towards toxic solvent and additives are still in challenge in achieving good synthesized conductive carbon-based ink for high performance printed strain sensor. The synthesized conductive carbon-based ink must possess good rheological behavior and wetting properties in order to produce excellent printed strain sensor performance. Studies on developing eco-friendly conductive carbon-based ink are still being conducted and started to expand as industries are moving towards environmental-friendly option. Numerous possibilities lie ahead for eco-friendly carbon-based conductive ink for printed strain sensor development in future which is as follows:

- Researching the method to recycle the eco-friendly carbon-based conductive ink which covers the recyclability of the carbon as conductive filler for multiple ink synthesis and recyclability of the end of life of the printed strain sensors. This can greatly enhance the efforts of achieving eco-friendly ink and can reduce costs.
- Ensuring the carbon material to be used as conductive filler produced using green synthesis method and green materials. This can ensure effective carbon-based ink synthesis using full green materials including carbon as conductive filler.
- Investigating renewable and sustainable materials to improve the performance of eco-friendly carbon-based conductive ink for printed strain sensors which can be applied to various printing methods. This can avoid shortage of materials for industrial scale production and reduce costs.

Nevertheless, integrating eco-friendly carbon-based conductive ink using green materials which covers binder, solvent and additives can significantly decrease the environmental impact and provide promising applications for printed strain sensors industry.

CRediT authorship contribution statement

Nur Iffah Irdina Maizal Hairi: Writing – review & editing, Writing – original draft, Methodology, Investigation. **Aliza Aini Md Ralib:** Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization. **Anis Nurashikin Nordin:** Writing – review & editing. **Muhammad Farhan Affendi Mohamad Yunos:** Writing – review & editing. **Lim Lai Ming:** Writing – review & editing, Resources. **Lun Hao Tung:** Writing – review & editing. **Zambri Samsudin:** Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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References

Akindoyo, J.O., Ismail, N.H., Mariatti, M., 2021. Development of environmentally friendly inkjet printable carbon nanotube-based conductive ink for flexible sensors: effects of concentration and functionalization. J. Mater. Sci. Mater. Electron. 32 (9), 12648–12660. https://doi.org/10.1007/s10854-021-05900-y.

Alshammari, A.S., Shkunov, M., Silva, S.R.P., 2014. Inkjet printed PEDOT: PSS/MWCNT nano-composites with aligned carbon nanotubes and enhanced conductivity. Phys. Status Solidi - Rapid Res. Lett. 8 (2), 150–153. https://doi.org/10.1002/pssr.201308231.

M. Amjadi and I. Park, "Carbon nanotubes-ecoflex nanocomposite for strain sensing with ultra-high stretchability," Proc. IEEE Int. Conf. Micro Electro Mech. Syst., vol. 2015-February, no. February, pp. 744–747, 2015, doi: 10.1109/MEMSYS.2015.7051065.

Altay, B.N., Bolduc, M., Cloutier, S.G., 2020. Sustainable advanced manufacturing of printed electronics: An environmental consideration. Green Energy Environ. 1.

Asnawi, M., Azhari, S., Hamidon, M.N., Ismail, I., Helina, I., 2018. Synthesis of Carbon Nanomaterials from Rice Husk via Microwave Oven. J. Nanomater. 2018 https://doi.org/10.1155/2018/2898326.

Azizi, S., David, É., Fréchette, M.F., Ouellet-Plamondon, C.M., Dec. 2019. Numerical simulation of the effective permittivity of low-density polyethylene composite filled by carbon black. IET Nanodielectrics 2 (4), 129–134. https://doi.org/10.1049/iet-nde.2019.0017.

Balandin, A.A., 2011. Thermal properties of graphene and nanostructured carbon materials. Nat. Mater. 10 (8), 569–581. https://doi.org/10.1038/nmat3064.

Bellani, S., et al., 2019. Scalable Production of Graphene Inks via Wet-Jet Milling Exfoliation for Screen-Printed Micro-Supercapacitors. Adv. Funct. Mater. 29 (14), 1–14. https://doi.org/10.1002/adfm.201807659.
Bernd, M.G.S., Bragança, S.R., Heck, N., Filho, L.C.P.D.S., 2017. Synthesis of carbon nanostructures by the pyrolysis of wood sawdust in a tubular reactor. J. Mater. Res. Technol. 6 (2), 171–177. https://doi.org/10.1016/j.jmrt.2016.11.003.
Bhagavatheswaran, E.S., et al., 2015. Construction of an Interconnected Nanostructured Carbon Black Network: Development of Highly Stretchable and Robust Elastomeric Conductors. J. Phys. Chem. C 119 (37), 21723–21731. https://doi.org/10.1021/acs.jpcc.5b06629.
D. Boikanyo, M. L. Masheane, L. N. Nthunya, S. B. Mishra, and S. D. Mhlanga, Carbon-supported photocatalysts for organic dye photodegradation, no. i. Elsevier Inc., 2018.
M. N. Bougot, T. M. D. Dang, N. N. Le, and M. C. Dang, “Realization of stable and homogenous carbon nanotubes dispersion as ink for radio frequency identification applications,” Adv. Nat. Sci. Nanosci. Nanotechnol., vol. 4, no. 2, 2013, doi: 10.1088/2043-6262/4/2/025008.
Brandley, A., Hollfelder, R., Nesaei, S., Vanwie, B., Abu-Lail, N., Gozen, B.A., 2018. Direct-Ink-Writing of Degradable Carboxymethylcellulose. Procedia Manuf. 26, 993–1002. https://doi.org/10.1016/j.promfg.2018.07.130.
R. Brooke et al., “Nanocellulose based carbon ink and its application in electrochromic displays and supercapacitors,” Flex. Print. Electron., vol. 6, no. 4, 2021, doi: 10.1088/2058-8585/ac3ddb.
Camargo, J.R., Silva, T.A., Rivas, G.A., Janegitz, B.C., 2022. Novel eco-friendly water-based conductive ink for the preparation of disposable screen-printed electrodes for sensing and biosensing applications. Electrochim. Acta 409. https://doi.org/10.1016/j.electacta.2022.139968.
Choi, H.J., Kim, M.S., Ahn, D., Yeo, S.Y., Lee, S., 2019. Electrical percolation threshold of carbon black in a polymer matrix and its application to antistatic fibre. Sci. Rep. 9 (1), 1–12. https://doi.org/10.1038/s41598-019-42495-1.
Christ, J.F., Aliheidari, N., Ameli, A., Pötschke, P., 2017. 3D printed highly elastic strain sensors of multiwalled carbon nanotube/thermoplastic polyurethane nanocomposites. Mater. Des. 131, 394–401. https://doi.org/10.1016/j.matdes.2017.06.011.
Conti, S., et al., 2023. Hybrid Flexible NFC Sensor on Paper. IEEE J. Flex. Electron. 2 (1), 4–10. https://doi.org/10.1109/jflex.2023.3238682.
L. Dai and J. Sun, “Mechanical properties of carbon fiber.pdf,” 2016.
Deng, L., Eichhorn, S.J., Kao, C.C., Young, R.J., 2011. The effective young’s modulus of carbon nanotubes in composites. ACS Appl. Mater. Interfaces 3 (2), 433–440. https://doi.org/10.1021/am1010145.
Du, J., et al., 2020. Optimized CNT-PDMS Flexible Composite for Attachable Health-Care Device. Sensors 20 (16), 4523. https://doi.org/10.3390/s20164523.
Dybowska-Sarapuk, L., et al., 2018. Efficient inkjet printing of graphene-based elements: Influence of dispersing agent on ink viscosity. Nanomaterials 8 (8), 9–14. https://doi.org/10.3390/nano8080602.
Eckelman, M.J., Mauter, M.S., Isaacs, J.A., Elimelech, M., 2012. New perspectives on nanomaterial aquatic ecotoxicity: Production impacts exceed direct exposure impacts for carbon nanotoubes. Environ. Sci. Technol. 46 (5), 2902–2910. https://doi.org/10.1021/es203409a.
Fiorillo, A.S., Critello, C.D., Pullano, A.S., 2018. Theory, technology and applications of piezoresistive sensors: A review. Sens. Actuators, A: Phys. 281, 156–175.
Franco, M., Alves, R., Perinka, N., Tubio, C., Costa, P., Lanceros-Mendéz, S., 2020. Water-Based Graphene Inks for All-Printed Temperature and Deformation Sensors. ACS Appl. Electron. Mater. 2 (9), 2857–2867. https://doi.org/10.1021/acsaelm.0c00508.
Fu, X., Ramos, M., Al-Jumaily, A.M., Meshkinzar, A., Huang, X., 2019. Stretchable strain sensor facilely fabricated based on multi-wall carbon nanotube composites with excellent performance. J. Mater. Sci. 54 (3), 2170–2180. https://doi.org/10.1007/s10853-018-2954-4.
Hamid, Z.A., Azim, A.A., Mouez, F.A., Rehim, S.S.A., 2017. Challenges on Synthesis of Carbon Nanotubes from Environmentally Friendly Green Oil Using Pyrolysis Technique, vol. 126. Elsevier B.V.
Hao, F., Maimaitiyiming, X., 2022. Fast 3D Printing with Chitosan/Polyvinyl alcohol/Graphene Oxide Multifunctional Hydrogel Ink that has UltraStretch Property. ChemistrySelect 7 (10), pp. https://doi.org/10.1002/slct.202200201.
He, P., et al., 2019. Screen-Printing of a Highly Conductive Graphene Ink for Flexible Printed Electronics. ACS Appl. Mater. Interfaces 11 (35), 32225–32234. https://doi.org/10.1021/acsami.9b04589.
He, Y., Lin, X., Feng, Y., Luo, B., Liu, M., 2022. Carbon Nanotube Ink Dispersed by Chitin Nanocrystals for Thermoelectric Converter for Self-Powering Multifunctional Wearable Electronics. Adv. Sci. 9 (33), 1–13. https://doi.org/10.1002/advs.202204675.
Higuchi, K., et al., 2013. High-mobility, flexible carbon nanotube thin-film transistors fabricated by transfer and high-speed flexographic printing techniques. Appl. Phys. Express 6 (8), 2–6. https://doi.org/10.7567/APEX.6.085101.
Htwe, Y.Z.N., Mariatti, M., 2021. Surfactant-assisted water-based graphene conductive inks for flexible electronic applications. J. Taiwan Inst. Chem. Eng. 125, 402–412. https://doi.org/10.1016/j.jtice.2021.06.022.
Htwe, Y.Z.N., Mariatti, M., 2022. Printed graphene and hybrid conductive inks for flexible, stretchable, and wearable electronics: Progress, opportunities, and challenges. J. Sci. Adv. Mater. Devices 7 (2), 100435. https://doi.org/10.1016/j.jsamd.2022.100435.
Hu, N., et al., 2010. Investigation on sensitivity of a polymer/carbon nanotube composite strain sensor. Carbon n. y. 48 (3), 680–687. https://doi.org/10.1016/j.carbon.2009.10.012.
Hu, M., et al., 2021. High-performance strain sensors based on bilayer carbon black/PDMS hybrids. Adv. Compos. Hybrid Mater. 4 (3), 514–520. https://doi.org/10.1007/s42114-021-00226-z.
Hu, X., et al., 2022. A Super-Stretchable and Highly Sensitive Carbon Nanotube Capacitive Strain Sensor for Wearable Applications and Soft Robotics. Adv. Mater. Technol. 7 (3), 1–9. https://doi.org/10.1002/admt.202100769.
Hu, S., Wei, Z., Chang, Q., Trinchi, A., Yang, J., 2016. A facile and green method towards coal-based fluorescent carbon dots with photocatalytic activity. Appl. Surf. Sci. 378, 402–407. https://doi.org/10.1016/j.apsusc.2016.04.038.
Hwangbo, Y., Nam, H.J., Choa, S.H., 2023. Highly Stretchable Strain Sensor with a High and Broad Sensitivity Composed of Carbon Nanotube and Ecoflex Composite. J. Korean Inst. Met. Mater. 61 (7), 500–508. https://doi.org/10.3365/KJMM.2023.61.7.500.
Jacob, M.V., et al., 2015. Catalyst-Free Plasma Enhanced Growth of Graphene from Sustainable Sources. Nano Lett. 15 (9), 5702–5708. https://doi.org/10.1021/acs.nanolett.5b01363.
J. Y. Je and S. K. Kim, Chitosan as Potential Marine Nutraceutical, 1st ed., vol. 65. Elsevier Inc., 2012.
Jeong, S., et al., 2008. Controlling the thickness of the surface oxide layer on Cu nanoparticles for the fabrication of conductive structures by ink-jet printing. Adv. Funct. Mater. 18 (5), 679–686. https://doi.org/10.1002/adfm.200700902.
Joo, D., Kang, M.-S., Park, S.J., Yu, S.A., Park, W.-T., 2022. Fabrication Method of Flexible Strain Sensors with Cnts and Solvents. SSRN Electron. J. 1–25. https://doi.org/10.2139/ssrn.4034699.
Kanoun, O., Bouhamed, A., Ramalingame, R., Bautista-Quijano, J.R., Rajendran, D., Al-Hamry, A., 2021. Review on Conductive Polymer/CNTs Nanocomposites Based Flexible and Stretchable Strain and Pressure Sensors. Sensors 21 (2), 1–29. https://doi.org/10.3390/s21020341.
Kantarak, E., et al., 2020. Fabrication, Design and Application of Stretchable Strain Sensors for Tremor Detection in Parkinson Patient. Appl. Compos. Mater. 27 (6), 955–968. https://doi.org/10.1007/s10443-020-09834-2.
M. C. Karlapudi, M. Vahdani, S. M. Bandari, S. Peng, and S. Wu, “A Comparative Study on the Effects of Spray Coating Methods and Substrates on Polyurethane/Carbon Nanofiber Sensors,” Sensors, vol. 23, no. 6, 2023, doi: 10.3390/s23063245.
S. Khan, S. Ali, and A. Bermak, “Recent developments in printing flexible and wearable sensing electronics for healthcare applications,” *Sensors (Switzerland)*, vol. 19, no. 5, 2019, doi: 10.3390/s19051230.
Khan, T., Irfan, M.S., Ali, M., Dong, Y., Ramakrishna, S., Umer, R., 2021. Insights to low electrical percolation thresholds of carbon-based polypropylene nanocomposites. Carbon n. y. 176, 602–631. https://doi.org/10.1016/j.carbon.2021.01.158.
Kim, H., et al., Dec. 2017. Tensile properties of millimeter-long multi-walled carbon nanotubes. Sci. Rep. 7 (1), 9512. https://doi.org/10.1038/s41598-017-10279-0.
Klemens, P.G., 2000. Theory of the a-plane thermal conductivity of graphite. J. Wide Bandgap Mater. 7 (4), 332–339. https://doi.org/10.1106/7FP2-QBLN-TJPA-NC66.
Koga, H., Saito, T., Kitaoka, T., Nogi, M., Suganuma, K., Isogai, A., 2013. Transparent, conductive, and printable composites consisting of TEMPO-oxidized nanocellulose and carbon nanotube. Biomacromolecules 14 (4), 1160–1165. https://doi.org/10.1021/bm400075f.
Kumar, R., Tiwari, R.S., Srivastava, O.N., 2011. Scalable synthesis of aligned carbon nanotubes bundles using green natural precursor: Neem oil. Nanoscale Res. Lett. 6 (1), 2–7. https://doi.org/10.1186/1556-276X-6-92.
Li, L., et al., 2018. Design of a wearable and shape-memory fibriform sensor for the detection of multimodal deformation. Nanoscale 10 (1), 118–123. https://doi.org/10.1039/c7nr06219b.
D. Li, K. Yao, Z. Gao, Y. Liu, and X. Yu, “Recent progress of skin-integrated electronics for intelligent sensing,” Light Adv. Manuf., vol. 2, no. 1, 2021, doi: 10.37188/lam.2021.004.
Li, W., Guo, J., Fan, D., 2017. 3D Graphite-Polymer Flexible Strain Sensors with Ultrasensitivity and Durability for Real-Time Human Vital Sign Monitoring and Musical Instrument Education. Adv. Mater. Technol. 2 (6), 1–9. https://doi.org/10.1002/admt.201700070.
Li, Y., Liu, Y., Bhuiyan, S.R.A., Zhu, Y., Yao, S., 2022. Printed Strain Sensors for On-Skin Electronics. Small Struct. 3 (2), 1–23. https://doi.org/10.1002/sstr.202100131.
Li, S., Park, J.G., Wang, S., Liang, R., Zhang, C., Wang, B., 2014. Working mechanisms of strain sensors utilizing aligned carbon nanotube network and aerosol jet printed electrodes. Carbon n. y. 73, 303–309. https://doi.org/10.1016/j.carbon.2014.02.068.
Lian, H., et al., 2022. Three-Dimensional Printed Carbon Black/PDMS Composite Flexible Strain Sensor for Human Motion Monitoring. Micromachines 13 (8), 1–10. https://doi.org/10.3390/mi13081247.
Lin, W., Rotenberg, Y., Ward, K.P., Fekrmandi, H., Levy, C., 2017. Polyaniline/multi-walled carbon nanotube composites for structural vibration damping and strain sensing. J. Mater. Res. 32 (1), 73–83. https://doi.org/10.1557/jmr.2016.361.
L. Lin et al., “Wearable and stretchable conductive polymer composites for strain sensors: How to design a superior one?,” Nano Mater. Sci., no. xxxx, 2022, doi: 10.1016/j.nanoms.2022.08.003.
Liu, C., et al., 2018. 3D printing technologies for flexible tactile sensors toward wearable electronics and electronic skin. Polymers (basel) 10 (6), 1–31. https://doi.org/10.3390/polym10060629.
C. Liu, S. Han, Z. Du, Y. Liu, and C. Liu, “Highly sensitive wearable strain sensors using copper nanowires and elastomers,” 2018 Int. Conf. Electron. Packag. iMAPS All Asia Conf. ICEP-IAAC 2018, pp. 333–338, 2018, doi: 10.23919/ICEP.2018.8374317.
Liu, Y., Zheng, H., Liu, M., 2018. High performance strain sensors based on chitosan/carbon black composite sponges. Mater. Des. 141, 276–285. https://doi.org/10.1016/j.matdes.2017.12.046.

Lo Presti, D., et al., 2021. Plant wearable sensors based on FBG technology for growth and microclimate monitoring. Sensors 21 (19), 1–16. https://doi.org/10.3390/s21196327.

N. L. Lukman Hekiem, A. A. Md Ralib, M. A. M. Hatta, F. B. Ahmad, R. A. Rahim, and N. F. Za'bah, "Performance Analysis of VOCs Detection using Polyisobutylene and Chitosan Overlayed on QCM Sensor," Proc. - 2021 IEEE Reg. Symp. Micro Nanoelectron. RSM 2021, pp. 157–160, 2021, doi: 10.1109/RSM52397.2021.9511614.

Ma, J., Wang, P., Chen, H., Bao, S., Chen, W., Lu, H., 2019. Highly Sensitive and Large-Range Strain Sensor with a Self-Compensated Two-Order Structure for Human Motion Detection. ACS Appl. Mater. Interfaces 11 (8), 8527–8536. https://doi.org/10.1021/acsami.8b20902.

D. Maddipatla, B. B. Narakathu, and M. Atashbar, "Recent Progress in Manufacturing Techniques of Printed and Flexible Sensors: A Review," Biosensors, vol. 10, no. 12, 2020, doi: 10.3390/BIOS10120199.

Maizal Hairi, N.I.I., Md Ralib, A.A., Ahmad, F.B., bt Mat Hattar, M.A., 2022. Synthesis and characterization of different graphite/chitosan ink ratio composition towards flexible strain sensor performance. J. Mater. Sci. Mater. Electron. 33 (19), 15574–15585. https://doi.org/10.1007/s10854-022-08463-8.

Menon, H., Aiswarya, R., Surendran, K.P., 2017. Screen printable MWCNT inks for printed electronics. RSC Adv. 7 (70), 44076–44081. https://doi.org/10.1039/c7ra06260e.

Mohammed Ali, M., et al., 2018. Printed strain sensor based on silver nanowire/silver flake composite on flexible and stretchable TPU substrate. Sensors Actuators, A Phys. 274 (2010), 109–115. https://doi.org/10.1016/j.sna.2018.03.003.

Mubarak, N.M., Abdullah, E.C., Jayakumar, N.S., Sahu, J.N., 2014. An overview on methods for the production of carbon nanotubes. J. Ind. Eng. Chem. 20 (4), 1186–1197. https://doi.org/10.1016/j.jiec.2013.09.001.

Nag, A., Mitra, A., Mukhopadhyay, S.C., 2018. Graphene and its sensor-based applications: A review. Sensors Actuators, A Phys. 270, 177–194. https://doi.org/10.1016/j.sna.2017.12.028.

Najafi, M., Zahid, M., Ceseracciu, L., Safarpour, M., Athanassiou, A., Bayer, I.S., 2022. Polylactic acid-graphene emulsion ink based conductive cotton fabrics. J. Mater. Res. Technol. 18, 5197–5211. https://doi.org/10.1016/j.jmrt.2022.04.119.

Oliva-Avilés, A.I., Avilés, F., Sosa, V., 2011. Electrical and piezoresistive properties of multi-walled carbon nanotube/polymer composite films aligned by an electric field. Carbon n. y. 49 (9), 2989–2997. https://doi.org/10.1016/j.carbon.2011.03.017.

K. Pan et al., "Sustainable production of highly conductive multilayer graphene ink for wireless connectivity and IoT applications," Nat. Commun., vol. 9, no. 1, 2018, doi: 10.1038/s41467-018-07632-w.

Park, J.J., Hyun, W.J., Mun, S.C., Park, Y.T., Park, O.O., 2015. Highly stretchable and wearable graphene strain sensors with controllable sensitivity for human motion monitoring. ACS Appl. Mater. Interfaces 7 (11), 6317–6324. https://doi.org/10.1021/acsami.5b00695.

Park, J., You, I., Shin, S., Jeong, U., 2015. Material approaches to stretchable strain sensors. ChemPhysChem 16 (6), 1155–1163. https://doi.org/10.1002/cphc.201402810.

Parsons, S., Murphy, R.J., Lee, J., Sims, G., 2015. Uncertainty communication in the environmental life cycle assessment of carbon nanotubes. Int. J. Nanotechnol. 12 (8–9), 620–630. https://doi.org/10.1504/ijnt.2015.068883.

Paul, S., Samdarshi, S.K., 2011. A green precursor for carbon nanotube synthesis. Xinxing Tan Cailiao/new Carbon Mater. 26 (2), 85–88. https://doi.org/10.1016/S1872-5805(11)60068-1.

Pellis, A., Byrne, F.P., Sherwood, J., Vastano, M., Comerford, J.W., Farmer, T.J., 2019. Safer bio-based solvents to replace toluene and tetrahydrofuran for the biocatalyzed synthesis of polyesters. Green Chem. 21 (7), 1686–1694. https://doi.org/10.1039/c8gc03567a.

Poulin, A., Aeby, X., Siqueira, G., Nyström, G., 2021. Versatile carbon-loaded shellac ink for disposable printed electronics. Sci. Rep. 11 (1), 1–9. https://doi.org/10.1038/s41598-021-03075-4.

M. Qasim, A. N. Clarkson, and S. F. R. Hinkley, "Green Synthesis of Carbon Nanoparticles (CNPs) from Biomass for Biomedical Applications," Int. J. Mol. Sci., vol. 24, no. 2, 2023, doi: 10.3390/ijms24021023.

Qin, Y., Ouyang, X., Lv, Y., Liu, W., Liu, Q., Wang, S., 2023. A Review of Carbon-Based Conductive Inks and Their Printing Technologies for Integrated Circuits. Coatings 13 (10), pp. https://doi.org/10.3390/coatings13101769.

Rahman, G., et al., 2019. An Overview of the Recent Progress in the Synthesis and Applications of Carbon Nanotubes. C 5 (1), 3. https://doi.org/10.3390/c5010003.

R. Rizvi, "Piezoresistance in Polymer Nanocomposite," p. 108, 2014, [Online]. Available: http://hdl.handle.net/1807/65733.

Roshni, V., Ottoor, D., 2015. Synthesis of carbon nanoparticles using one step green approach and their application as mercuric ion sensor. J. Lumin. 161, 117–122. https://doi.org/10.1016/j.jlumin.2014.12.048.

Saidina, D.S., Eawwiboonthanakit, N., Mariatti, M., Fontana, S., Hérold, C., 2019. Recent Development of Graphene-Based Ink and Other Conductive Material-Based Inks for Flexible Electronics. J. Electron. Mater. 48 (6), 3428–3450. https://doi.org/10.1007/s11664-019-07183-w.

L. Sanchez-Duenas et al., "A Review on Sustainable Inks for Printed Electronics: Materials for Conductive, Dielectric and Piezoelectric Sustainable Inks," Materials (Basel)., vol. 16, no. 11, 2023, doi: 10.3390/ma16113940.

Sappati, K.K., Rout, B., Girard-Lauriault, P.L., Bhadra, S., 2021. Plasma Treatment of Composite Piezoelectric Thin Films for Good Adhesion of Printed Conductive Ink. ACS Appl. Polym. Mater. 3 (1), 319–328. https://doi.org/10.1021/acsapm.0c01117.

D. D. Saputri, A. M. Jan'ah, and T. E. Saraswati, "Synthesis of Carbon Nanotubes (CNT) by Chemical Vapor Deposition (CVD) using a biogas-based carbon precursor: A review," IOP Conf. Ser. Mater. Sci. Eng., vol. 959, no. 1, 2020, doi: 10.1088/1757-899X/959/1/012019.

Secor, E.B., et al., 2017. Enhanced Conductivity, Adhesion, and Environmental Stability of Printed Graphene Inks with Nitrocellulose. Chem. Mater. 29 (5), 2332–2340. https://doi.org/10.1021/acs.chemmater.7b00029.

Secor, E.B., Prabhumirashi, P.L., Puntambekar, K., Geier, M.L., Hersam, M.C., 2013. Inkjet printing of high conductivity, flexible graphene patterns. J. Phys. Chem. Lett. 4 (8), 1347–1351. https://doi.org/10.1021/jz400644c.

K. Senthil Kumar, P.-Y. Chen, and H. Ren, "A Review of Printable Flexible and Stretchable Tactile Sensors," Research, vol. 2019, pp. 1–32, 2019, doi: 10.34133/2019/3018568.

Seo, D.H., et al., 2017. Single-step ambient-air synthesis of graphene from renewable precursors as electrochemical genosensor. Nat. Commun. 8, 1–9. https://doi.org/10.1038/ncomms14217.

Singla, T., Pal Singh, A., Kumar, S., Singh, G., Kumar, N., 2021. Characterization of MWCNTs-polystyrene nanocomposite based strain sensor. Proc. Inst. Mech. Eng. Part E J. Process Mech. Eng. 235 (2), 463–469. https://doi.org/10.1177/0954408920966301.

Sousa, E.A., Lima, T.H.C., Arlindo, E.P.S., Sanches, A.O., Sakamoto, W.K., Fuzari-Junior, G. de C., 2020. Multicomponent polyurethane–carbon black composite as piezoresistive sensor. Polym. Bull. 77 (6), 3017–3031. https://doi.org/10.1007/s00289-019-02888-8.

Szabó, A., Perri, C., Csató, A., Giordano, G., Vuono, D., Nagy, J.B., 2010. Synthesis methods of carbon nanotubes and related materials. Materials (basel) 3 (5), 3092–3140. https://doi.org/10.3390/ma3053092.

Tang, W., et al., 2019. Rapid fabrication of wearable carbon nanotube/graphite strain sensor for real-time monitoring of plant growth. Carbon n. y. 147, 295–302. https://doi.org/10.1016/j.carbon.2019.03.002.

Temizel-Sekeryan, S., Wu, F., Hicks, A.L., 2021. Global scale life cycle environmental impacts of single- and multi-walled carbon nanotube synthesis processes. Int. J. Life Cycle Assess. 26 (4), 656–672. https://doi.org/10.1007/s11367-020-01862-1.

K. Thiyagarajan, G. K. Rajini, and D. Maji, "Paper-based screen printed MWCNT-PDMS composite strain sensor for human motion monitoring," 2019 IEEE 16th India Counc. Int. Conf. INDICON 2019 - Symp. Proc., pp. 14–17, 2019, doi: 10.1109/INDICON47234.2019.9030347.

Tong, L., et al., 2018. Electrically Conductive TPU Nanofibrous Composite with High Stretchability for Flexible Strain Sensor. Nanoscale Res. Lett. 13 https://doi.org/10.1186/s11671-018-2499-0.

Tran, T.S., Dutta, N.K., Choudhury, N.R., 2018. Graphene inks for printed flexible electronics: Graphene dispersions, ink formulations, printing techniques and applications. Adv. Colloid Interface Sci. 261, 41–61. https://doi.org/10.1016/j.cis.2018.09.003.

Trantidou, T., Elani, Y., Parsons, E., Ces, O., 2016. Hydrophilic surface modification of pdms for droplet microfluidics using a simple, quick, and robust method via pva deposition. Microsystems Nanoeng. 3 (November), 2017. https://doi.org/10.1038/micronano.2016.91.

Wang, W., Asci, C., Zeng, W., Sonkusale, S., 2023. Zero-power screen printed flexible RFID sensors for Smart Home. J. Ambient Intell. Humaniz. Comput. 14 (4), 3995–4004. https://doi.org/10.1007/s12652-022-04466-9.

Wang, R., Xu, W., Shen, W., Shi, X., Huang, J., Song, W., 2019. A highly stretchable and transparent silver nanowire/thermoplastic polyurethane film strain sensor for human motion monitoring. Inorg. Chem. Front. 6 (11), 3119–3124. https://doi.org/10.1039/c9qi00989b.

Williams, N.X., Bullard, G., Brooke, N., Therien, M.J., Franklin, A.D., 2021. Printable and recyclable carbon electronics using crystalline nanocellulose dielectrics. Nat. Electron. 4 (4), 261–268. https://doi.org/10.1038/s41928-021-00574-0.

Xiang, D., et al., 2019. Enhanced performance of 3D printed highly elastic strain sensors of carbon nanotube/thermoplastic polyurethane nanocomposites via non-covalent interactions. Compos. Part B Eng. 176, no. July, 107250 https://doi.org/10.1016/j.compositesb.2019.107250.

Yang, T., Xie, D., Li, Z., Zhu, H., 2017. Recent advances in wearable tactile sensors: Materials, sensing mechanisms, and device performance. Mater. Sci. Eng. R: Rep. 115, 1–37.

Yazdani, H., Smith, B.E., Hatami, K., 2016. Electrical conductivity and mechanical performance of multiwalled CNT-filled polyvinyl chloride composites subjected to tensile load. J. Appl. Polym. Sci. 133 (29), 1–12. https://doi.org/10.1002/app.43665.

Yi, Y., Samara, A., Wang, B., 2021. A new approach for an ultra-thin piezoresistive sensor based on solidified carbon ink film. J. Mater. Sci. 56 (1), 607–614. https://doi.org/10.1007/s10853-020-05309-8.

Yi, Y., Wang, B., Liu, X., Li, C., 2022. Flexible piezoresistive strain sensor based on CNTs–polymer composites: a brief review. Carbon Lett. 32 (3), 713–726. https://doi.org/10.1007/s42823-022-00320-2.

Zeng, M., Zhang, Y., 2019. Colloidal nanoparticle inks for printing functional devices: Emerging trends and future prospects. J. Mater. Chem. A 7 (41), 23301–23336. https://doi.org/10.1039/c9ta07552f.

Zhang, X., et al., 2020. Printed carbon nanotubes-based flexible resistive humidity sensor. IEEE Sens. J. 20 (21), 12592–12601. https://doi.org/10.1109/JSEN.2020.3002951.

Zhang, F., et al., 2022. Structural engineering of graphite network for ultra-sensitive and durable strain sensors and strain-controlled switches. Chem. Eng. J. 452 (September), 2023. https://doi.org/10.1016/j.cej.2022.139664.

Zhang, J., Lei, Z., Luo, S., Jin, Y., Qiu, L., Zhang, W., 2020. Malleable and Recyclable Conductive MWCNT-Vitrimer Composite for Flexible Electronics. ACS Appl. Nano Mater. 3 (5), 4845–4850. https://doi.org/10.1021/acsanm.0c00902.