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Conductive Colorless Films Using Printing Technology Toward Flexible Electronics

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Abstract

The promising and remarkable qualities of graphene have piqued curiosity, making it a viable alternative to many standard materials in a variety of applications including conductive ink which used in construction of flexible electronics. Many methods have been applied to produce graphene-based conductive inks to print flexible substrates. Diverse flexible electronic devices have been reported during the last ten years. The development of graphene-based ink is discussed, focusing on the type of the graphene-like materials used in the conductive ink as well as the composition and properties of these inks. The second objective of this study is clarifying the advantages and disadvantages of graphene-based ink and comparing it with its counterparts from other electrically conductive materials like silver (Ag), gold (Au), and copper (Cu) nanoparticles (NPs), as well as carbon nanotubes and conducting polymers. Additionally, this work discussed graphene hybrid-based inks using various types of metallic nanoparticles to enhance the characteristics of conducting ink.

Keywords: Black mulberry, Fruit leathers, Drying kinetics, Hydrocolloids Application, Scanning electron microscopy (SEM).

1. Introduction

Flexible electronics has attracted tremendous research interest recent years. Graphene has been used to develop optoelectronics due to its remarkable mechanical and physicochemical characteristics as well as high electrical conductivity [1, 2]. graphene has hexagonal structure consist of single layer of carbon atom in two-dimensional honeycomb lattice structure.it included in inks for flexible electronics production by printing processes [3-5]. There are two synthetization techniques for production graphene, (1) top- down method, which begins with graphite flake and (2) bottom-up method, which begins with carbon precursors [6], **Figure 1** represents the different methods for each technique. Each technique represents advantages and drawbacks. **Table 1** represents the advantages and drawbacks for each one [7-13].

Many researches were implemented to produce electrically conductive inks for printable flexible electronic applications, these inks classified into inks based on conductive nanomaterials and inks based on conductive polymers. Conductive inks based on conductive nanomaterials such as silver, gold, and copper nanoparticles. These nanomaterials are high electrically conductive materials; however, both silver and gold nanoparticles have showed high sintering temperature and high cost, limiting their function in flexible electronics. On the other hand, copper nanoparticles are low cost but it oxidized by heat and humidity [14-17]. According to disadvantages for AuNPs, AgNPs and CuNPs that limit their applications [17]. Graphene based inks are excellent to be used in printed electronic applications in the last 10 years and that increases the researches about this type of inks before practical applications, researchers make effort for production high amount with high quality, eco-friendly and low cost and methods to produce this type of inks.

Printing quality depends on ink formulation and properties, so optimization these items is very important to achieve homogeneous printed patterns without coffee ring effect. There are many printing methods such as inkjet, silk screen , 3Dprinting and spray coating which are used in the printing for patterns used in flexible electronic applications [18].

Inkjet printing technique is the most one used according to its simplicity using, time- saving, high repeatability and economy, the properties of ink and its formulation has direct effect on this type of printing because if the ink formulation doesn't suitable inkjet printers that make ink to aggregate in the nozzle. Hoath informed that best ink must have high surface tension and low viscosity to make the passage from the nozzle easier [19].Conductive ink is paste contains conductive materials that has ability for conducting electricity [20]. This paste is thermoplastic and viscous, the ideal conductive ink must be low cost, easily prepared, low viscosity, high printability, highly adhesion to substrate, highly stable and has high electrical conductivity, has no coffee ring effect [14, 21, 22].

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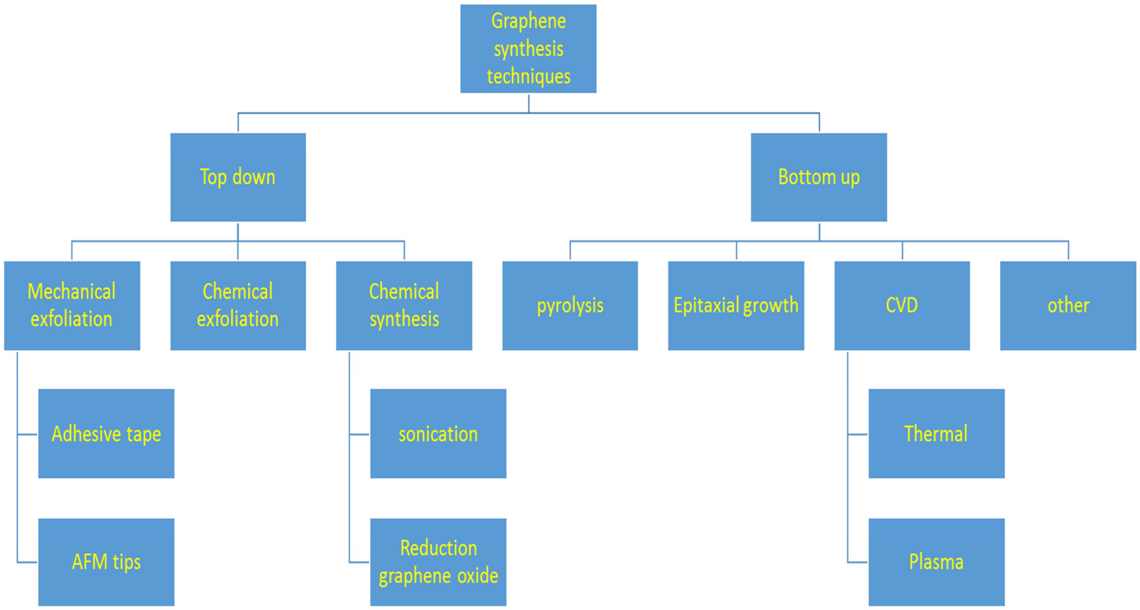

Figure 1: Production techniques of graphene [7]

Table 1: Advantages and drawbacks for top-down and bottom-up techniques

Top –down technique	Bottom- up technique
- more complicated - less graphene output - Higher cost of metal substrate	- high graphene output by using solution-based method - easy to implement (according to using existing bulk form)

2. Graphene-type inks

According to the numerous properties for carbon nanomaterials that make them used in preparation of flexible electronics. Graphene conductive inks used in the printable electrically conductive applications compared to inks based on metallic materials and conductive polymer due to its low biological hazards and low cost [23]. Huang et al. [24] informed that electrical conductivity of few layered GO(FGO) is higher than single layered GO after inkjet printing 25 layers on a polyimide(PI) substrate and that explained due to GO sample has high number of oxygen groups. Pei and LI [25] reported that the dispersion of GO in an equal ratio deionized water, ethylene glycol and ethanol (1:1:1) then by inkjet printing of resulted ink on treated (oxygen- plasma treatment) and untreated poly ethylene terephthalate(PET) substrate after 20 printed layers they found that electrical conductivity of treated substrate is higher than untreated one. Arapov et al. [26] reported the variance in conductivity levels between graphene inks obtained by the two different methods, the first method is by solubilisation of stretched out graphite in the existence of surface active polymeric material and the second one is by the reaction of covalent graphene functionalization then redispersion in a solvent in the absence of surfactant, the conductivity level of first one after 15 printed layers≈1-2 k Ω/sq but for the other one ≈2MΩ/sq. This method is more efficient and simpler so it used in printing large area of conducting films.

Miao et al. [14] reported that by inkjet printing 20 layer for prepared GNps (using electrochemical method in an inorganic salt-based electrolyte) then at 300˚C for 1 h thermal treatment occur and in absence of stabilizer that improve the electrical conductivity for the printed pristine GNPs film. also he reported that by inkjet printing of GNPs which prepared using shear-exfoliation method and assistance of bromine in an aqueous media using stabilizer of water-soluble cellulose making improvement for electrical conductivity after adding treatment by immersing the printed film in a solution of aqueous iodine before drying and annealing in air occur at 300˚C, iodine doping and thermal annealing treatment have positive effect on the improvement the electrical conductivity for printed GNPs films . Miao and derby [14, 27]synthesized a film of reduced graphene oxide (rGO) then printed it on substrates from glass after that dehydrated at 80˚C, the electrical conductivity for rGO films and printed pristine graphene films is similar . The hydrophobic nature for most of GNPs, that make them low soluble in many forms of solvents. To avoid the low solubility for GNPs the addition of stabilizer or surfactant is very important and that enhance the dispersion for conductive ink, the surfactant is used because it has no effect on the electrical conductivity of the resulted printed design and that due to the remaining component in the final product, the removal of surfactant from the printed pattern is important. According to the high amount for oxygen- based functional groups in GO sheets that leads to decreasing its electrical conductivity so the removing this excess of functional groups by reduction method is important, during removing method for these groups high toxic materials are used, affecting on the conductivity. Annealing temperature is another important factor, affecting on the conductivity value. The electrical conductivity for graphene sheet increases by annealing at high temperature and this high temperature not preferred for plastics substrates involved in fabrication flexible electronic applications [14]. The electrical properties for the printed films influenced by the amount of printed layer, degree of annealing temperature, kind of substrate. The flexible electronic applications require plastic substrate which requires low annealing temperature.

3. Other conductive inks

There are many other materials has high conductivity other than graphene. Gold nanoparticles (AuNPs) are metals marked by their stability. They have high conductivity and according to their unique properties, they can be used in many uses such as metal coating, colorants, optics, catalysis and electronics [14, 28, 29]. AuNPs have drawbacks that they need high sintering temperature and long sintering time beside that they have high cost, so these materials not preferred in electronic applications. Cui et al. [14] informed that by using overlapped layers from acrylic resin(AR)and poly N-vinylpyrrolidone(PVP) to make protection for AuNPs ink then printing for 50 printed layers and sintering at 500˚C for 3h , the resulted conductivity is 0.8 x105 S/cm. Silver nanoparticles (AgNPs) is another conductive nanomaterial used in flexible electronics applications, it is the most important nanotechnology other than graphene. Excellent conductivity achieved by using high temperature for sintering the printed film. Flexible polymer substrates (such as PEN, PET) have low glass-transition temperature so by increasing sintering temperature above 200˚C this not compatible for these substrates. Nie et al. [30] reported that curing conditions has effect on the printed Ag film and RFID antenna by inkjet printing, when curing occur at 150˚C ,50 min for silver citrate conductive ink the film extended a low resistivity (17µΩ) but when the curing temperature increased to 230˚C for 50 min the resistivity is 3.1µΩ.

Zhang and Zhu [31] synthesized silver nanoparticles (AgNPs) for inkjet printing of flexible electronics by sintering at room temperature and dipping treatment. This method presented larger controllability, higher stability and lower resistivity. According to high cost, sintering temperature and sintering time for AuNPs and AgNPs that make them not compatible in most of industrial applications. Copper nanoparticles (CuNPs) characterized by their high electrical conductivity and low cost so they used as alternative to AuNps and AgNPs. Kang et al. [17] reported that CuNP ink printed on substrate from flexible glass epoxy and sintering by furnace at 200˚C in nitrogen atmosphere, a grain structure has stability and low resistivity of 36.7 nΩ in a 10-layer printed electrode is obtained. Tsai et al. [14, 32] shows a technique to prepare conductive copper ink antioxidant, this method takes place at calcination in nitrogen atmosphere at temperature 250˚C for 30 min, low resistance film of 47.6mΩ/sq is obtained, this film can be placed in environmental atmosphere with high stability and without oxidation for more than 6 months. Kwon et al. [33] provided new method for sintering Cu complex ion ink using hydrogen plasma which takes place under 150˚C temperature for time 10-20 min that produced ink- jet printed patterns with complete reduction. This pattern has completely microstructure its resistivity 3.2µΩ cm.

Xu et al. [14] considered the performance of copper complex conductive inks which influenced by the dual effects of water with amines as ligands [34]. These effects are balanced by mixing copper format with 2-ethylhexylamine and 2-amino-2- methyl-1-propanol as ligands, the resistivity obtained for printed films decreased from 9.70µΩ cm to 14.42µΩ cm, could be kept for 2 months and verify the requirements of printed electronics. Although CuNPs has high electrical conductivity and their cost is low compared to AuNPs and AgNPs but it has drawbacks as a following: -

a) they can be oxidized under the effect of heat and humidity and that limit their uses.
b) it is hard to yield homogeneous nanoparticles with good dispersion in ink without sedimentation and that due to instability for these materials in most solvents such as water, isopropyl alcohol, and acetone [35].
c) they similar to AuNPs and AgNPs in requirement to high sintering temperatures and sintering times to improve their electrical conductivity.

Carbon nanotubes (CNTs)are graphene sheets rolled-up and the edges for these sheets are joined together to produce smooth cylinders. They are electrically heterogeneous in nature (metallic or semiconducting). CNTs classified according to the number of rolled- up graphene sheets into single, double, triple and multi-walled CNTs (SWCNTs, DWCNTs, TWCNTs, MWCNTs). The number of walls influences on The electronic properties for these materials [36, 37].According to the low electrical resistivity for CNTs (nearly 10 -6 Ω) that make them attractive for many electronic applications [38]. Kordas et al. [14] proved simple method for production electrically conductive CNTs patterns on different substrates surfaces such as paper and plastic. To prepare CNT can be used for inkjet printing, Carboxylation and dispersion in water for MWCNTs which resulted by catalytic chemical vapour deposition (CCVD) takes place. The sheet resistivity for printed MWCNTs is nearly 40 kΩ/sq after several printing. This method can be applied for preparation resistive compounds, electromagnetic interference shielding and gas antennas. CNT beneficial in transparent transistors, however its suspension stability in water is still attract attention according to nanoparticles aggregate easily by their high Van der Waals forces of attraction, it has toxicological issues and it needs careful growth, functionalization and sorting methods for separation to keep its electronic properties.

4. Polymer-type inks

Conductive polymers are another conductive material characterized by its low cost and not required any sintering process specific polyaniline (PANI), polypyrrole (PPy), poly 3,4- ethylenedioxythophene (PEDOT), polystyrenesulfonate (PSS), PEDOT: PSS.

PANI characterized by high electrical conductivity so it is the most favorable conducting polymer, environmental stability and partial solubility in many solvents, its simplicity, its low cost and its chemical and thermal stability [39, 40]. PPy used in multifunctional application, it has moderate environmental stability. The preparation methods for the polymeric films (polymerization electrochemical method, monomer concentration and doping agents) effects on the electrical and physical properties for these films. The polymer in its natural state not conducting and it converts to conducting one by oxidation [14, 41, 42]. PEDOT: PSS is a type of polymers, it is mixing for poly 3,4- ethylenedioxythophene (PEDOT) and plystyrenesulfonate (PSS), its high electrical conductivity, water dispersibility and simple processing makes it most technologically polymers [43-45]. The electrical conductivity for this polymer is lower than 10 S/cm which enhanced by post- treatment with ethylene glycol [14, 46]. Cho et al. [14] informed that formation a conducting polymer on several flexible substrates takes place by using vapour deposition polymerization-mediated inkjet printing. PANI films which thickness of nearly 450 nm has sheet resistance of 3.8×10^3 Ω/sq.

Conductive polymers exhibit electrical conductivity lower than metallic nanoparticles for some electronic applications (**Table 2**). The preparation of conductive ink requires adjusting viscosity and surface tension to avoid the nozzle clogging and the effect of acidic materials which cause mechanical damage during the printing process.

5. Hybrid-type inks

Hybrid inks of graphene and metallic nanoparticles or polymer include GNP/PANI, Ag/rGO, Ag and Au NPs/Go, rGO/carbon black is the recent research for specific electronic applications. The electrical conductivity for some of them is still considered very low, so the improvement electrical conductivity and annealing temperature is very important to make these inks can be used in flexible electronic applications [14, 47-50]. Xu et al. [14] formulated hybrid inks of(GNP/PANI) to form GNP/PANI thin film electrodes, The substrate required is quartz to give conductivities of GNP and GNP/PANI films 3.67 S/cm and 0.29 S/cm for (5 prints, thickness nearly equal 41 µm)and (5 prints, thickness nearly equal 28 µm) respectively, while the conductivity is only 10-9 S/cm for pure PANI [51]. Yang et al. [14, 52]. formulated hybrid ink of Ag/rGo by using silver acetate and ethanolamine with rGo. Their exist inverse relationship between resistivity and annealing temperature which decreased from 0.728 Ω cm to 1.47x 10-5 Ω cm by increasing annealing temperature from 150˚C to 245˚C. Deng et al. prepared ink of graphene and AgNPs for flexible electronic applications and found resistivity decreased 20 Ω/sq by using 400 ˚C annealing temperature for time 30 min. Zhang et al. [14]. reported that by fixing the AgNPs on rGo sheet surface that leads to forming well- dispersed Ag/rGo which is used as conductive ink filler for printable flexible electronics. The best conductivity is obtained 2.0x103 S/cm at 100˚C annealing temperature. The simplicity and low cost for this method make it used for printed flexible electronics in future.

Table 2: Properties of conductive inks [14, 17, 23, 31, 53-55]

Ink material	**Resistivity(Ωcm)/surface resistance(Ω/sq)/conductivity(s/cm)**	**Advantages**	**drawbacks**
AuNPs	0.8 9x105 S/cm	High electrical conductivity	- High cost -high sintering temperature
AgNPs	17µΩ cm (cured at 150˚C For 50 min), 3.1µΩ cm (cured at 230˚C for 50 min) 4.42x106 S/m (annealed at 120˚C) 31.6–26.5 µΩcm (sintered at room temperature 30˚C)	High electrical conductivity	- high cost. -silver particles make clogging for inkjet printer nozzle. - high sintering temperature - long sintering time
CuNPs	36.7 nΩ m (sintered at 200˚C in a furnace under nitrogen atmosphere) 3.6µΩ cm (sintered at 200˚C for 1h in a furnace under nitrogen atmosphere)	High electrical conductivity	- oxidized under the effect of heat and humidity. -high sintering temperature - long sintering time
PEDOT:PSS	1.1 mS/cm	Low electrical conductivity	Does not require sintering process
Carboxyl functional MWCNT and PEDOT:PSS	1.1 9 106 Ω/sq (MWCNT- COOH) and 3.5 9 103 Ω/sq (MWCNT-COOH-PSS)	High electrical conductivity	Toxicological effect

Li et al. [49] formulated hybrid nanomaterial ink from nanotriangle platelets and graphene oxide of Ag, Au (Ag, Au NTPs-Go). The patterns obtained characterized by absence of coffee ring effects and high transparency (nearly equal 98%). The sheet resistance after fabrication and after 100 days is 146.8 Ω/sq,149.5 Ω/sq respectively. This method applied in integrated circuit boards has high stability also in highly transparent devices. Ji et al. [14] formulated hybrid ink from rGo- carbon black, mixture of ethylene glycol, ethyl alcohol, glycerol, deionized water as solvent and binding agent from sodium carboxymethyl cellulose. The formed hybrid ink is applied onto photographic paper using ink jet printing and drying at 100˚C for 30 min. The resulted resistance value decreased from 1.43MΩ to 0.09MΩ after 1-5 printed layers. Zhang et al. [50]. prepared PEDOT: rGO/AuNps (PrGANPs), using inkjet printing multilayer films(10-60-layers) were printed on PET substrate. The resulted sheet resistance decreased by increasing number of printed layers (500 kΩ /sq after 50 printed layers).

5.1. Compositions of conductive inks

Conductive ink formulation depends on its application and the type of printing processes. Yang and Wang reported that the conductive materials should not aggregate or precipitate because aggregation and precipitation effects on ink performance [56]. The agglomeration of conductive material- based ink is due to increasing ink viscosity and that change the properties for ink and that during printing process cause nozzles clogging of the printer [57]. To avoid this agglomeration some additives as surfactants, stabilizing agents, dispersants, adhesion promoters are used to ink. Conductive material-based inks are suspension from materials has conductivity in medium includes additives, both medium and additives not naturally conductive. Minimum volume from Conductive materials on the substrate surface is required to achieve interparticle connectivity also removing the remaining nonconductive materials from the printed patterns leads to enhancement the connection between conductive materials and increase conductivity. Inks are mixture of pigments or dyes, water, humectant, additives such as rheological, modifiers for surface tension, deformers and binders [58, 59]. The importance of additives is enhancement the performance of conductive ink [35]. Conductive ink classified to aqueous- based, solvent- based, oil- based, hot-melt and UV-curable inks.

5.2. Aqueous-immobilized inks

Aqueous- based ink was the primary remains used till now in ink jet printing. Its primary solvent is water; it contains pigment or dye or both of them as colorant. Pigments may be organic materials (alcohols or glycols) or inorganic materials. This type of inks can be applied on absorbent papers for décor printing also can be applied in corrugated packaging and in flexible plastic packaging, ceramic tile market. [60]. There are some difficulties by using aqueous- based ink that is volatile in open system as inkjet printing and drying on substrates especially non- absorbent substrates. To avoid these difficulties addition for non- volatile solvents is required also humectants, surfactants, special additives all of that increase the performance of ink during and after printing processes. Using functional materials during fabrication of ink such as emulsion polymers as colorants that make ink can be used in specific applications. The surface tension properties of aqueous- based inks affected by Migration speed for the surface- active compositions such surfactants, solvent and polymers, also surfactants play an essential function in wetting characteristics for ink in the printhead and on substrate surface. Tai and Yang informed a method by direct writing to make high efficiency conductive patterns for flexible electronics applications, they used aqueous conductive ink from Ag solution [61].

Another method to prepare ink by using aqueous conductive Cu nanosheet for flexible electronics was informed by Dang et al. [62]. The conductive performance for Cu nanosheet is excellent. the preparation method for GNP ink was informed by Majee et al. In this method, exfoliated GNPs in water was mixed with cellulose stabilizer (hydroxyl propyl methyl cellulose) [63].

5.2.1. Solvent-immobilized inks

Solvent based ink characterized by its high conductivity and that make it is widely used in industry. This type of ink consists of solvent, pigmented colorant, resin and glossing agent [64]. There are two types of solvent-based inks (1) inks contains more aggressive solvent (2-butoxyethyl acetate and cyclohexanone), this type characterized by high substrate compatibility, and (2) inks contain less aggressive solvents, eco-solvent inks, (dipropylene glycol monomethyl ether) – but they are harmful due to their dangerous chemicals. Solvent based inks have high stability and fast drying but it releasing the volatile organic compounds so during preparation for this type of ink manufacturing controls are required to increase health and safety profiles (ex: ecological and toxicological profiles) also reducing volatility. Arapov et al. [26]. reported the constituent of solvent- based conductive inks used for inkjet printing, this type of inks produced by combination between expanded graphite with non- toxic solvents. Kell et al. [65] fabricated an Ag- based conductive ink for flexible electronic applications by using aliphatic solvent and polymer. This kind of inks characterized by its low resistance and high hold to substrate.

5.2.2. Oil-immobilized inks

Oil-based inks consist of very slow- drying carrier fluid comes from mineral oil. This type of ink used in ceramic tiles printing and corrugated boxes coding, according to the low surface tension for purified petroleum distillates so it intended to use mixture from their oils to control viscosity, rate of absorption wettability on the surface without surfactants. Oil- based inks have low drying after printing so using maintainable alternatives oil as vegetable- oil derivatives for preparing this ink can overcome this low drying [59]. - The compatibility of Oil- based inks with nanoparticles and stabilizing agents since it is good dispersing medium for nanoparticles that make this type of ink useful for the dispersion and stabilization of suspension and can be used for printing with high resolution [60]. Zhang et al. [66] informed that CuNP oil- based ink can be kept under ambient conditions for 30 days also it can be printed with low electrical resistivity using silk-screen printing process.

5.2.3. Melt-type inks

Hot- melt inks (phase- change inks) are gel-like at room temperature. These type of inks characterized by fast-drying, ecofriendly and has high print quality according to its rapid solidification but there exist drawbacks that they have poor stability and abrasion resistance [59]. The commonly method used to prepare it is combination for wax and resin materials has different melting points with dye or pigment, the additives as plasticizers are used for controlling the surface tension, affecting on the final properties. These inks can be applied in barcodes printing on non-permeable substrate [60].

5.2.4. Ultraviolet-curable inks

UV- curable inks are type of inks that cured by using UV light. It used in inkjet printing and can be applied in wide- format printing of rigid substrates. This type of inks characterized by immediate drying once it leaves the printer and no solvents penetrate the substrate and it has safety and health issues [60] but Its cost is high. Hwang et al. [67] fabricated Cu nano- ink(highly conductive) by ultra- high speed photonic sintering method, this method comprises flash white light(FWL) combined with near infrared (NIR) and deep ultraviolet (UV) light irradiation. The resistivity for Cu nano- ink films produced by using UV- assisted FWL sintering method is the lowest one 7.62 μΩ cm, and is higher than Cu bulk film 1.68 μΩ cm [67].

5.3. Properties of conductive inks

The printing processes are affected by the characteristics of conductive inks such as high adhesion with the substrate, high resolution, low printer maintenance and longer shelf life [68].There are many types for printing techniques, for examples: inkjet, flexography, screen printing, gravure. Each type from these techniques depends on the conductive ink properties such as viscosity, surface tension, droplet size and impact factor of droplets on dry surface consequently that effects on the printed conductive patterns. The surface tension and viscosity are the two main factors that control the quality of the printed patterns [19]. For example: in inkjet printing, it requires printable conductive ink has low viscosity and high surface tension to flow easily from the printer nozzle without leakage, drying out, coagulation.

5.3.1. Rheological properties

Viscosity is the important property for liquids, it defined as the resistance of liquid to flow. The liquids resistance due to cohesive intermolecular forces which leads to friction between neighboring layers of the liquid in relative motion. Both temperature and pressure influence the viscosity; (i.e. the viscosity decreased by increasing temperature) [19, 69]. Conductive ink viscosity differs for printing methods **(Table 3)**. inkjet printing needs low viscosity compared to other printing methods to prevent clogging of the printer nozzle [70]. Both rheological behavior and dispersion state of the printable conductive ink in inkjet printing should be controlled according to their significance influence on the printed patterns [71].

Table 3: Printing methods of conductive inks

Printing methods	Ink viscosity (cP)	Line width (μm)	Line thickness (μm)	Speed (m/min)
Inkjet	10–20	30–50	Approx. 1	Slow (rotary screen: 10 m/s)
Offset	100–10000	10	10	Mid-fast (approx. 1000)
Gravure	100–1000	10–50	Approx. 1	Fast (approx. 1000)
Flexography	50–500	45–100	< 1	Fast (approx. 500)
Screen	500–5000	30–50	5–100	Mid (approx. 70)

5.3.2. Surface tension

Surface tension is the second important property, affecting on the printing quality. Due to cohesive intermolecular force in the liquid, there is an asymmetric attractive force between molecules at the interface and between liquid and air and that leads to tension at the surface **(Figure 2a)** [59].

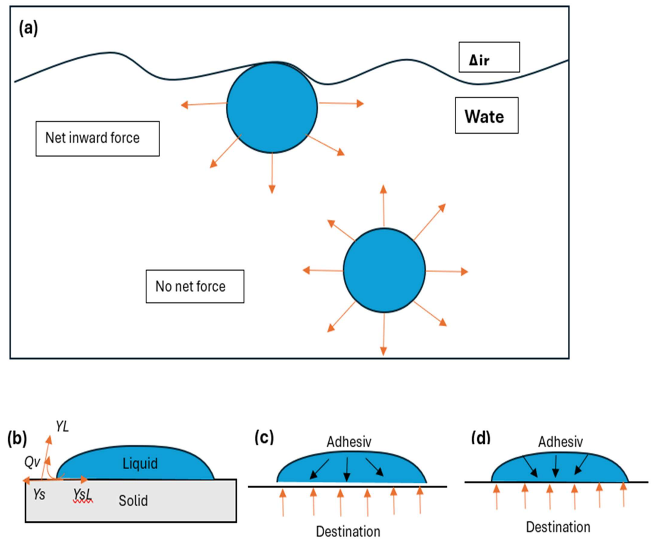

Figure 2: (a) Intermolecular force-driven surface tension, (b) droplet on a surface, and droplets on a surface with (c) high surface energy and (d) low surface energy.

Temperature leads to direct effect on the surface tension, by increasing temperature the surface tension decreased. There are two forces in liquids adhesion force and cohesion force, the adhesion force enables the liquid to adhere on the surface of substrate [59]. By deposition the droplets of ink on the substrate surface, the shape of these droplets relies on strength for these molecular forces. If adhesion force is greater than cohesion force that leads to spreading the ink droplet on the substrate surface and causing hydrophilicity, in difference if cohesion force greater than adhesion force that leads to beading up the ink droplet on the substrate surface (hydrophobicity).The ideal state for spreading the droplet onto the surface of substrate is only when balance constant angle is obtained **(Figure 2b)**, the liquid surface tension makes the force to act perpendicularly to the substrate surface and that can be measured in terms of surface energy. The surface energy between liquid, solid and gas interfaces can be identified by the following young's equation $\gamma S = \gamma SL + \gamma L \cos\theta$; γS: is solid surface energy, γSL is solid/ liquid interface surface energy and γL is the liquid surface energy [72]. **Figure 2c-d** represents the two different states of ink droplet on the substrate surface. **Figure 3** represents the droplet wetting behavior on the surface of substrate. To produce a stream of droplets for inkjet printing The surface tension recommended values should be 25–50 mN m^{-1} [14, 59].

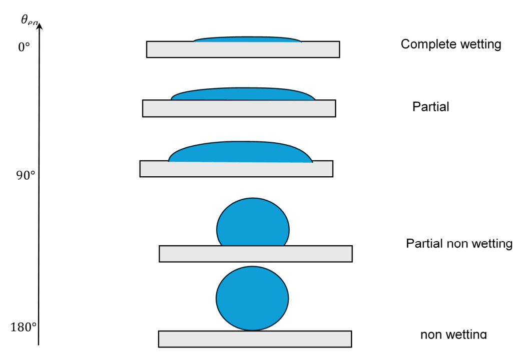

Figure 3: Droplet wetting of a surface.

6. Conclusion and future remarks

We evaluated recent research on the varieties of graphene materials utilized in conductive ink, as well as the compositions and key features of conductive inks, in this study. Inks containing other popular conductive nanomaterials were examined to see how well graphene conductive ink performed. Clearly, graphene-based inks have a number of benefits over other conductive nanomaterials, including strong electrical conductivity. However, further research is needed to examine environmentally acceptable solvents, ink stability, graphene material varieties, and types of substrates. As a result, considerable problems keep on developing large-scale, best-quality, low-cost, and graphene-based ink processes and handling. Graphene hybrid-based ink is another option for improving conductive ink's electrical characteristics; however, this process has raw ingredients with a high cost also optimization for ink preparation is still required. We believe that this review will be useful resource for scholars who are concerned with graphene- and other conductive nanomaterial-based inks. this review as a stepping stone to the future of flexible electronics, which will be the next technology.

7. Declaration of competing interests

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.

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