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FLEXIBLE AND STRETCHABLE PRINTABLE CONDUCTIVE INKS FOR WEARABLE TEXTILE APPLICATIONS

GİYİLEBILIR TEKSTİL UYGULAMALARI İÇİN ESNEK VE GERİLEBİLEN BASILABİLİR İLETKEN MÜREKKEPLER

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Derleme Makalesi / Review Article

FLEXIBLE AND STRETCHABLE PRINTABLE CONDUCTIVE INKS FOR WEARABLE TEXTILE APPLICATIONS

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ABSTRACT: As wearable electronic devices become increasingly integrated into our daily routines, there is a growing demand for soft, flexible, and comfortable devices that can seamlessly deliver electronic functionalities. Electronic textiles (e-textiles) combine the electronic capabilities of devices such as sensors, actuators, energy storage, and communication tools with the comfort and flexibility inherent in traditional textiles. The rising interest in E-textile and sensor applications has thrust the field of printed electronics (PE) into the spotlight. Printed electronics is a rapidly expanding technology that allows the construction of electronic devices on affordable, flexible substrates, including paper and textiles. This is achieved through printing techniques, such as screen printing, 3D printing, gravure printing, offset printing, flexography, and inkjet printing, which are traditionally used in various industries like graphic arts, textiles, and polymers. This paper provides a comprehensive overview of printable conductive inks, with a focus on their role in designing textile-based wearable conductive devices for E-textile applications. Within this scope, it was examined the properties of conductive inks, presented the various printing methods used to fabricate wearable textile materials for potential use in wearable textile devices, and analyzed their performance characteristics. Lastly, it was addressed the key challenges faced in this field and identify future research directions. The aim of this paper is to contribute to the advancement of cost-effective functional conductive inks and formulations, promoting their integration into E-textile applications.

***Keywords*:** Electronic textiles, printed electronics, conductive inks, wearable devices

GİYİLEBILIR TEKSTİL UYGULAMALARI İÇİN ESNEK VE GERİLEBİLEN BASILABİLİR İLETKEN MÜREKKEPLER

ÖZ: Giyilebilir elektronik cihazlar günlük rutinlerimize giderek daha fazla entegre hale geldikçe, elektronik işlevleri kusursuz bir şekilde sunabilen yumuşak, esnek ve konforlu cihazlara olan talep de artmaktadır. Elektronik tekstiller (e-tekstil), sensörler, aktüatörler, enerji depolama ve iletişim araçları gibi cihazların elektronik yeteneklerini geleneksel tekstillerin doğasında bulunan konfor ve esneklikle birleştirir. E-tekstil ve sensör uygulamalarına artan ilgi, baskılı elektronik alanını ilgi odağı haline getirdi. Baskılı elektronik, elektronik cihazların kâğıt ve tekstil dahil uygun fiyatlı, esnek yüzeyler üzerinde oluşturulmasına olanak tanıyan, hızla genişleyen bir teknolojidir. Bu, grafik sanatlar, tekstil ve polimerler gibi çeşitli endüstrilerde geleneksel olarak kullanılan serigrafi, 3D baskı, gravür baskı, ofset baskı, fleksografi ve inkjet baskı gibi baskı teknikleri ile elde edilir. Bu makale, E-tekstil uygulamaları için tekstil bazlı giyilebilir iletken cihazların tasarlanmasındaki rollerine odaklanarak, yazdırılabilir iletken mürekkeplere kapsamlı bir genel bakış sunmaktadır. Bu kapsamda iletken mürekkeplerin özellikleri incelenmiş, giyilebilir tekstil cihazlarında potansiyel kullanım için giyilebilir tekstil malzemelerinin üretilmesinde kullanılan çeşitli baskı yöntemleri sunulmuş ve performans özellikleri analiz edilmiştir. Son olarak bu alanda karşılaşılan temel zorluklar ele alındı ve gelecekteki araştırma yönleri belirlendi. Bu makalenin amacı, uygun maliyetli fonksiyonel iletken mürekkeplerin ve formülasyonların geliştirilmesine katkıda bulunarak bunların E-tekstil uygulamalarına entegrasyonunu teşvik etmektir.

Anahtar Kelimeler: Elektronik tekstiller, baskılı elektronikler, iletken mürekkepler, giyilebilir cihazlar

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1. INTRODUCTION

In recent years, garments have evolved to provide not only warmth and comfort but also electronic features with various functions. This transition from traditional textiles to electronic or e-textiles has made them suitable tools for wearable electronics. Many researchers have conducted studies in which they incorporated solar cells, thermoelectric devices, or piezoelectric devices into textiles to generate electricity. Furthermore, textile products capable of storing electricity have been designed by integrating lithium-ion batteries and supercapacitors with textile surfaces [1, 2]. These innovative textiles are particularly attractive due to their seamless adaptation to different sizes and shapes of the human body. E-textiles have evolved not only to capture physical, chemical, and electrophysiological signals from the skin under various conditions but also to visualize this data and provide immediate information in daily life. Several e-textile products have entered commercial markets, especially in healthcare and sports. These products include smart socks, shirts, sleeves, and gloves designed for wearable sensing applications, monitoring parameters such as walking speed, blood pressure, skin temperature, respiration rate, cardiac rhythm, blood oxygen levels, and daily activity levels. Additionally, e-textiles incorporating photometric or colorimetric units are being used in wearable displays, serving not only as fashionable illuminated apparel but also as communication tools, visual aids, safety precautions, and health monitoring devices [3]. E-textiles are made by weaving, knitting, or embroidering conductive wires into fabrics. Yet, they encounter challenges like production methods, fabric constraints, and possible harm to natural fabric structures [3-7]. Textile surface flexibility allows seamless integration of wearable electronics on the skin, with promise for future applications, like strain sensors, flexible displays, and antennas [8]. Electroconductive textiles can be produced through various methods, such as integrating conductive fibers or yarns, applying conductive coatings, or using conductive inks [9]. A schematic representation of the functionalization of textile sensors was given in Figure 1. In recent years, printing methods such as inkjet printing, soft lithography, and screen printing have been employed to produce electronic circuits [4, 5]. This increased interest is primarily driven by the rapid advancements in wearable electronic technology, which aims to produce environmentally friendly smart products using cost-effective printing methods such as screen printing and inkjet printing [4, 10, 11].

Printed electronics are very popular in electronic applications due to their lightness and flexibility. They are accepted as a new technology, unlike traditional methods used to produce electronic circuits. The primary advantage of this technology is that it does not require costly and time-consuming production steps such as coating and masking, which are used in traditional methods like photolithography. Therefore, given the demand for fast and inexpensive wearable electronic devices produced with low-cost and high-quality materials, it is considered a necessity in today's conditions. [13, 14]. As evidenced by the rapid increase in research and the publication of scientific papers on stretchable electronics over the last ten years, printed electronics have a promising future, particularly given their superior properties such as mechanical strength and portability. According to research on printed flexible electronics, its market share is estimated to reach nearly $73 billion in 2027, up from approximately $14 billion in 2017, with an increase of around 13.6%. Stretchable electronic devices are expected to possess certain properties, including electrical conductivity, strength, biocompatibility, lightness, and environmental friendliness. For this reason, attention should be paid to the following important points during the printing phase [15]:

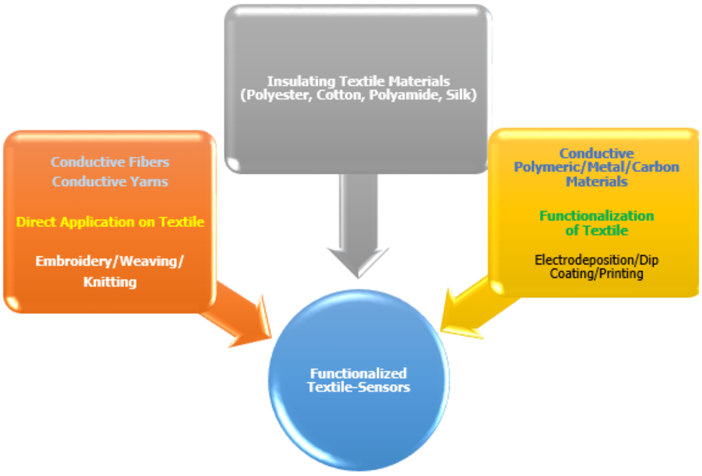

Figure 1. A visual representation of the functionalization of textile sensors using a range of techniques, incorporating both organic materials (cotton and silk), as well as synthetic materials (polyester and polyamide) [12].

(i) Using films with superior flexibility for the easy connection of electronic devices to the body.

(ii) Employing environmentally friendly, easily soluble, and chemically stable inks for sustainable printing applications.

(iii) Creating clear, high-resolution shapes for lightweight devices with good electrical properties.

(iv) Preventing breakage and slippage through the design of special device structures to enhance durability.

Electronic equipment in printing is necessary for achieving perfect electrical contact in environments where various components coexist. Therefore, devices such as electrochemical cells, antennas, photodetectors, and electrochemical sensors, which require flexibility and stretchability, can be fabricated using special inks containing conductive fillers to provide functionality [10, 15, 16]. Electrically conductive materials are receiving significant attention due to their wide range of applications in the field of wearable devices [4-6]. To develop conductive E-textiles various types of conductive materials such as polymer-based, carbon-based, and metal based are being investigated to prepare conductive inks or formulations for printing/coating applications on textile substrates. Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and polypyrrole (PPy) are significant conductive polymer materials investigated for E-textile applications, owing to their distinct electrical, optical, and mechanical properties [2, 17, 18]. Also known as carbon-based materials, carbon nanotubes (CNTs), carbon fibers, and graphene are being explored for conductive E-textile applications because of their outstanding properties, such as high mechanical strength and superior electrical conductivity. Metallic materials are known for their high conductivity properties compared to other conductive materials, making them preferred materials for wearable electronics [2]. The growing interest in printed wearable electronics has driven the need for cost-effective materials and simple manufacturing methods and conductive inks are crucial in the printed electronics market. However, challenges persist, such as expensive toxic solvents and complex manufacturing processes [15, 18, 19]. Research has explored using materials like CNTs, graphene, copper (Cu), and silver (Ag) for ink production. While carbon materials offer advantages such as lightweight properties and corrosion resistance, achieving high electrical conductivity with carbon-based inks remains a challenge due to significant contact resistance. Among various metallic materials, silver-based conductive ink stands out as the most promising option. This is due to its excellent electrical conductivity, chemical inertness, and cost-effectiveness. Considerable efforts have been dedicated to the advancement of Ag-based conductive inks [20]. Successfully produced graphene-based inks have been used in printed sensors, and among other applications [21]. Graphene, an allotrope of carbon, consists of a two-dimensional sp2 bonded carbon lattice [22-24]. The geometry of graphene sheets in printable applications is of interest due to the low size of the particles: can be less than 1 nm of thickness, 0.77 μm of width, and show a high surface area (2630 g/m^2) [21]. With its high mechanical, thermal and electrical properties, it is a suitable material for printed flexible electronics, as well as attracting great attention for a wide variety of applications [25-30]. Graphene has also been used for the development of composite materials and piezoresistive and thermoresistive sensors in the field of stimuli sensitive materials [29, 31, 32].

Conductive inks rely on polymers as stabilizing agents to obtain conductive materials. These polymers not only prevent the aggregation of conductive fillers but also serve as protective capping agents against oxidation. Additionally, the polymer component provides essential binding properties that ensure the adhesion of printed patterns to the substrate. This binding function can be attributed to either the drying/evaporation of solvents present in ink formulations or the initiation of a polymerization reaction during ink curing [33]. By the using multilayer hydrophobic film-like materials such as binders like PU, Poly(diallyl dimethylammonium chloride) PDDA, polydimethylsiloxane (PDMS) on the textile surface, a protective layer can be improved which also provides the wash durability for wearable e-textiles [17, 18]. In recent years, polyurethanes (PUs) have been used in a variety of industrial applications such as foams, coatings, and textiles, in terms of their low cost and versatility. In addition to the traditional applications of PUs, their potential application fields such as soft electronics, and biosensors has been developed [2]. Flexible sensors based on PUs offer significant working strain capabilities thanks to their exceptional tensile characteristics [34]. Water-based PUs (WPUs) are also environmentally friendly by using water instead of volatile organic compounds for sustainable production in the process of adaptation to climate change [35, 36]. Polyurethanes are block-copolymers consisting of a hard segment and a soft segment. Polyols give flexibility to the polymer, while hard segments that provide rigidity and strength are formed by an isocyanate and a chain extender [37, 38]. WPU is a colloidal structure that disperses in aqueous media, as ionic hydrophilic segments are included in their structure. Functional composite mixtures can be formed with other colloid particles and inorganic nanoparticles (NPs) to tune the mechanical or thermal properties of the PU polymer [2, 39-41]. Recent years studies related to WPU applications increased in the fields such as synthesis, chemistry, coatings of WPU nanocomposites, conductive films, printable inks and pastes [2, 35-37, 42-44].

This paper provides an overview of printable conductive inks in the context of designing textile-based wearable conductive devices for E-textile applications. This paper is organized into five parts. First, it provides an overview of textile-based wearable conductive devices for E-textile applications. In the second part, it examines and discusses conductive materials for E-textiles and conductive inks. The third part delves into common printing techniques for flexible and stretchable electronics. The fourth part focuses on the design of flexible and stretchable electronics. Lastly, the fifth part presents studies related to hybrid conductive inks that can be used in flexible and stretchable E-textile applications. In conclusion, the article offers remarks and perspectives.

2. CONDUCTIVE MATERIALS FOR E-TEXTILES

It is crucial to define the functional reliability of wearable textile electronics for applications in daily life. Different textile surfaces are used for various applications in healthcare, electronics, robotics, sports, space exploration, fitness, and lifestyles. These distinctions are critical when designing functional and wearable smart clothing. Two-dimensional (2D) textile surfaces with large surface areas, such as woven, knitted, and nonwoven fabrics, can be manufactured at a high scale and are suitable for electrical functionality. On the other hand, fibers (1D) are well-suited for wearable electronics due to their high aspect ratio and flexibility on the film surface. These textile structures are made from both natural fibers and synthetic polymers and serve as substrates for new and popular applications, including energy storage, sensing, and transistors. The production process of fiber and fabric-based electronics can be challenging to perform using pattern processing techniques such as lithography. This difficulty arises from the curved geometries and rough surface structures of the textile surfaces used. The primary factors influencing the electrical behavior of conductive textile structures are the thickness of the conductive layers, the structural geometry, and the material type.

Conductive materials used in textronic construction can be classified into three groups: conductive polymers, metallic wires/nanoparticles, and carbon nanoscale materials [45].

2.1 Conductive Polymers for E-Textiles

Conductive polymers with excellent electrical conductivity are an excellent choice for wearable electronics. Conducting polymers offer numerous advantages, including high adhesion, low density, biocompatibility, low processing temperatures, cost-effectiveness, and controllable electrochemical properties. As a result, they find wide-ranging applications in transistors, batteries, sensors, solar cells, functional coatings, light-emitting diodes, and more. Frequently used conductive polymers, such as polythiophene (PT), polyaniline (PANI), and PPy, exhibit excellent printability, solubility, and conductivity. Another notable conductive polymer is Poly(3-hexylthiophene) (P3HT), which is commonly employed in solar cells and other power conversion applications [45].

2.2 Conductive Metals for E-Textiles

Materials such as silver, copper, gold, and stainless steel, which are among the traditional metallic nanoparticles, possess both electrical and thermal conductivity. Furthermore, these materials can address the issue of air aging, especially prevalent in non-metallic particles used in the construction of wearable conductive circuits. However, the difference in mechanical performance between textiles and rigid metallic components has limited the utilization of metallic materials in flexible conductive devices. This limitation arises from the fabrics containing these materials having inherent defects, such as hardness, susceptibility to rusting, poor air permeability, and high weight. To overcome such challenges, low-dimensional metallic nanoparticles, nanowires, and nanosheets have been developed for use in the fabrication of flexible and wearable electronics [45].

2.3 Carbon Materials for E-Textiles

In recent studies, carbon nanomaterials have garnered significant attention due to their large surface area, lightweight nature, mechanical strength, superior electrical conductivity, and thermal conductivity properties. Carbonaceous fillers such as carbon black (CB), MXene, graphene, and CNTs can be incorporated into textile structures using three different approaches. The first approach involves transferring carbon fibers and yarns to the textile layer through weaving, knitting, or processing. The second approach entails achieving high-temperature carbonization of the substrate under an inert atmosphere. Finally, the most effective approach is the direct coating of carbonaceous materials onto the fabric surface. Among these methods, the last approach is advantageous because the coating process can maintain the mechanical stability and flexibility of the wearable electrode while preventing brittleness that can result from carbonization [45].

3. CONDUCTIVE INKS

Conductive inks are complex formulations that consist of a blend of conductive materials, solvents, and various additives like binders, surfactants, and polymers. These inks are expected to meet several critical criteria, including high electrical conductivity, cost-effectiveness, low viscosity, stability, printability, and excellent adhesion to a wide range of substrates. Furthermore, they must maintain their electrical conductivity and chemical inertness after the printing process, ensuring a consistent, coherent conductive film on the substrate. As the fields of wearable electronics and bioelectronics continue to expand, biocompatibility has emerged as a particularly desirable characteristic for conductive inks. Typically, conductive inks comprise three primary components: the conductive material, a binder that facilitates efficient particle aggregation, and a solvent that regulates the ink's viscosity for the chosen printing method. Commonly used conductive materials encompass metallic nanoparticles and various carbon materials, such as graphite, graphene, and carbon nanotubes. While metal-based conductive inks offer superior electrical conductivity, they are often more expensive and less biocompatible when compared to their carbon-based counterparts. Additionally, metal-based inks can be less stable due to the susceptibility of metallic nanoparticles to oxidation and aggregation. Conductive inks can be broadly categorized into two groups: solvent-based and water-based inks. Solvent-based inks offer distinct advantages, including ease of application, low viscosity, and rapid drying, making them more manageable for various manufacturing processes. However, the high cost and potential toxicity of organic solvents pose significant challenges, particularly in large-scale production. Conversely, water-based inks, aside from being environmentally friendly, offer cost-effectiveness and non-flammability. These appealing attributes have spurred ongoing efforts to develop more efficient methods for producing water-based conductive inks [30].

Wearable and flexible screen printed electronics can be manufactured using functional conductive inks, including metal nanoparticles, CNTs, graphene, and conductive polymers [4]. With recent developments in the field of printed electronics, nanomaterial-based conductive inks with electrical conductivity and glossy surface properties have emerged that can be used in the production of stretchable and flexible electronic wearable devices. Material selection can be made according to the physical properties of the device to be produced. These physical properties include flexural and buckling resistance, visual clarity, and durability of the printed design, which are critical to electronic application performance. It has been observed that carbon-based nanomaterials have a high potential in the development of printed and flexible electronics compared to other nanomaterials [15, 46]. The physical properties of the inks, including the adhesion of the pattern to be printed on the substrate and compatibility with the printing technique, are crucial factors in the selection of the conductive material. Specifically, the rheology and surface energy of the ink have a significant impact on the properties and behavior of the ink. The rheology of the ink should be suitable for the applied printing equipment and process, ambient conditions, production speed, and parameters. Figure 2 provides an overview of the required properties of conductive inks [15]. Graphene, used as a filler in the preparation of graphene/polymer composites, has garnered increasing attention due to its excellent mechanical properties and thermal stability and it has found extensive use in some researches [29, 31, 32]. The interfacial interaction between the dispersion surface of graphene and the polymer matrix is a determining factor in the properties of graphene/polymer composites. Furthermore, in most polymers, pure graphene exhibits poor dispersibility. To address this issue, methods such as covalent and non-covalent bonding functionalization have been employed to enhance the dispersion of graphene and the interfacial interaction between graphene and the polymer matrix while altering the surface properties of graphene. Graphene modified through covalent functionalization provides increased stability, and strong interfacial bonding is necessary for efficient charge transfer from the polymer matrix to graphene. The surface of the graphene oxide (GO) layer contains numerous reactive groups, including hydroxyl, carboxyl, epoxide, and carbonyl [32]. Based on the type of conductive filler used, conductive inks can be categorized into four groups: inorganic materials, conductive materials, organic materials, and composite conductive inks [47-49]. Resins can be employed to offer appropriate adhesiveness and mechanical properties, as well as to aid in the dispersion of conductive particles. During the formulation preparation stage, solvents are selected based on the printing surface and application to disperse the resin and control the rheological properties of the ink. Subsequently, additives are introduced into the formulation to ensure functionality [15]. The key strategy in formulation development is to prevent deformation of the polymer matrix by creating a three-dimensional interconnected conductive network within flexible and conductive polymer composites [50].

Figure 2. Summary of required properties of conductive inks [15].

4. DESIGN OF FLEXIBLE AND STRETCHABLE ELECTRONICS

Flexible and stretchable electronics are produced by printing conductive devices on flexible substrates or by mounting electronic circuits on these flexible substrates. Efficiency, cost, availability, and compatibility with organic materials are important key factors in the production of flexible wearable electronic devices. It is of great importance to determine the appropriate flexible substrates, conductive ink materials, and the most suitable printing method to be used in the application for the fabrication of flexible printed electronic devices with the desired properties and performance. Essentially, the fabrication of stretchable and flexible circuits is quite complex and challenging as it involves connecting circuits and sensors to provide power transmission and data, as well as creating conductive lines on the flexible surface according to the specified design [2]. Printing techniques offer a straightforward, efficient, and scalable method for applying conductive coatings to fabric. This approach finds utility in various industries due to its suitability for large-scale manufacturing. A diverse range of conductive inks can be employed for fabric printing, including those composed of metal nanoparticles, conductive polymers, and carbon materials. For effective printing, the inks must possess a suitable viscosity and adhere securely to the fabric's surface. They should not deteriorate or lose conductivity when the fabric is subjected to stretching, washing, or reuse [51].

Recent literature highlights the incorporation of inorganic materials into polymeric substrates to achieve specific sensor properties, such as electrical and thermal conductivity and antimicrobial features. Composite materials, formed by blending different materials, combine their unique characteristics to enhance sensor performance. These composites typically involve a reinforcement phase within a matrix, fortifying the materials and transferring loads effectively. Commonly used matrices include polymers like styrene-ethylene/butylene-styrene (SEBS), PU, polyethylene, and various thermoplastic elastomers (TPEs). TPE-SEBS polymers, characterized by hard styrene segments and a flexible cis-polybutadiene core, provide resilience, excellent tensile strength, and durability. However, they are generally non-conductive. To address this limitation, conductive nanoparticles like graphene, carbon black, carbon nanotubes, silver nanoparticles, or silver nanowires can be added to create composite materials with electrical conductivity. Silver nanoparticles are preferred for their outstanding electrical and thermal conductivity, elasticity, tensile properties, and stability during processing [52].

The electrical conductivity properties of composites are determined by the dependence of the composite on the conductive filler content (Fig. 3). As the concentration of the conductive filler in the composite increases, a conductive path is formed within the structure, allowing for the movement of free electrons and resulting in a higher level of electrical conductivity (Fig. 4). In Fig. 4, the composite conductivity-volume percent curve is displayed in three sections. In the first section (Part I) with low filler concentrations, the conductive particles tend to move apart from each other, and the electrical conductivity of the composite approaches that of the matrix material. Part II represents the phase where the conductivity of the composite shifts as the fillers begin to form a conductive network, with the starting point of the conductive path marked as "A". At this critical volume, known as the percolation threshold, there is enough filler present to establish interparticle interactions and a percolating network. In a range of filler concentrations not far above this threshold, electrical conductivity increases significantly. With higher filler concentrations, the electrical conductivity becomes several times higher than that of the pure polymer. This phenomenon can be attributed to the formation of multiple conductive paths created by the fillers, and the mixing rule is no longer applicable in this section [53]. The formation of this conductive network in composite formulations can be explained by the principles of percolation theory [54]. The electron transfer mechanism in conductive composites is explained by percolation theory. Percolation occurs when there is a continuous bonding between fillers, and this threshold value is reached at a specified filler concentration. As filler is added to the matrix, the resistivity of the conductive composite continues to decrease until it reaches the percolation threshold. Until this threshold is reached, the coupling between the fillers remains low, but after reaching the threshold, the resistance drops drastically [2]. The conductivity of composite threshold (σ) can be calculated by the following:

$$\sigma = \sigma_0 \, (V_f - V_C)^s \tag{1}$$

The conductivity of the composite is denoted as σ, and σ_0 represents a scaling factor associated with the conductivity of the filler. The filler volume fraction, percolation threshold, and the effectiveness exponent are represented as V_f, V_c, and s, respectively [2]. As the amount of conductive filler increases, the composite material reaches the percolation threshold. Once this threshold value is surpassed, the electrical conductivity of the composite material increases with the growing amount of conductive filler integrated into the material's structure. Polymer resins containing various graphene fillers lead to an enhancement in the electrical conductivity of the composites (Table 1). The rise in electrical conductivity is attributed to the conductive filler forming a conductive path within the dielectric matrix material. This path is established once the conductive filler content exceeds the percolation threshold [54].

By incorporating conductive fillers into polymers, materials suitable for various applications can be produced and designed. The electrical conductivity values of conductive polymers typically range between 10^{-14} and 10^{-17} S/cm. In contrast, materials like carbon black, carbon fiber, and graphite exhibit electrical conductivity values of 10^2, 10^4, and 10^5 S/cm, respectively. In conductive applications, composite materials should ideally possess electrical conductivity values ranging from 10^{-12} to 10^{-2} S/cm. It is well-known that the type of conductive filler significantly impacts the electrical conductivity capacity of the composites. Furthermore, the type, shape, size, and distribution of the filler within the composite play a crucial role in determining its electrical conductivity. Some of the materials that can serve as conductive fillers include CNTs, carbon black (CB), graphite, and graphene. However, since each filler material has different electrical conductivity values, the composite's electrical conductivity depends on the specific filler employed. The inclusion of conductive fillers can also alter the percolation threshold, thus affecting the overall conductivity of the composites. If the filler shape is spherical or the aspect ratio is less than 1, the percolation threshold is reduced. The distribution of the filler within the matrix materials and the surface properties of each component also influence the electrical conductivity and percolation threshold of the composites. Additionally, a high level of electrical conductivity can be achieved if there is a small surface energy difference between the matrix and the filler [55].

Table 1. The percolation threshold of graphene reinforced different polymer resins [54].

Material	Percolation Threshold
Functionalized graphene filled epoxy composites	0.1
Neat graphene/epoxy nanocomposite	0.53
Graphene/polyethylene composite	0.07
The graphene/polyethylene terephthalate (PET) nanocomposite	0.47
TRGO (Thermally reduced graphene oxide)	<0.5
Graphite	>2.7

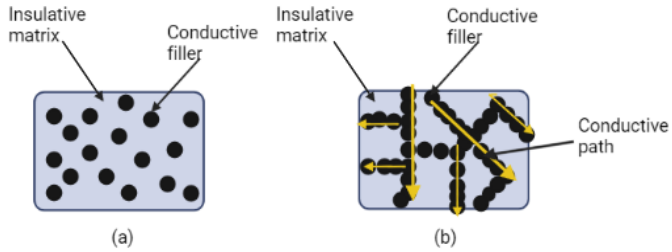

Figure 3. Schematical representation of filler distribution in polymer matrix, a) at low content, b) at high content [54].

When mechanical stress is applied to stretchable electronic materials, conductive fillers become active within the matrix, altering the conductivity of the composites. This phenomenon is known as the piezoresistive effect. In contrast, a stretchable electronic material integrates the manipulation of this effect with percolation theory to ensure that conductivity is maintained even in the presence of stress [2]. The electrical resistance of the material is given in the following equation [18]:

$$R = \frac{\rho l}{A} \tag{2}$$

In equation (2) ρ is the electrical resistivity; l is the length of a sample, and A is the cross-sectional area of the resistor.

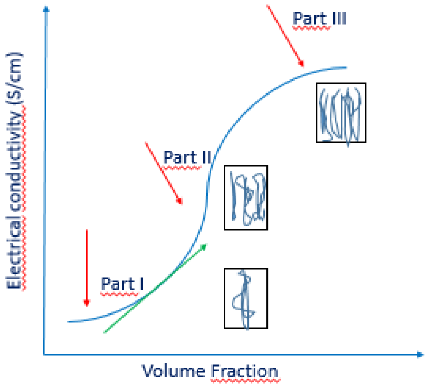

Figure 4. The dependence of electrical conductivity on the filler volume fraction [53]

5. COMMON PRINTING TECHNIQUES FOR FLEXIBLE AND STRETCHABLE ELECTRONICS

Printing technology has been employed to create electrical patterns on diverse surfaces, such as paper, textiles, and polymers, to aid in the development of wearable devices. Wearable smart devices are typically manufactured by applying conductive ink onto a substrate using various printing techniques, with flexography, inkjet printing, screen printing, and stencil printing being the most common methods. Each of these techniques has its unique advantages and limitations, but they were all developed to provide a straightforward, quick, efficient, and cost-effective means of creating conductive traces on flexible substrates. These printing techniques are appealing as a manufacturing process because they employ additive fabrication methods, offering simplicity, affordability, scalability, and minimal waste generation. Therefore, the integration of innovative conductive inks with these efficient printing techniques is likely to play a crucial role in constructing the next generation of electronic devices [30, 47]. Methods such as screen and inkjet printing enable cost-effective, scalable production of eco-friendly, flexible electronic devices in various sizes [21]. Inkjet printing, screen printing, and 3D printing are common methods for manufacturing and processing water-based PU composite ink structures. Printing inks are typically prepared with micro- or nano-sized fillers or by mixing the polyelectrolyte in water-based PU through mechanical or sonication processes, and surfactants are added to improve dispersion [4]. Conductive materials lack adhesion to textiles, causing cured inks to wipe off surfaces and lose conductivity after washing. Polymer binders are essential to improve adhesion of conductive ink to textiles in E-textile applications [2]. By using multilayer hydrophobic film-like materials, such as binders like PU, Poly(diallyl dimethylammonium chloride) (PDDA), and polydimethylsiloxane (PDMS) on the textile surface, a protective layer can be improved, which also enhances the wash durability of wearable e-textiles [45, 56]. Adding a polymer binder to printing ink can reduce conductivity and stretchability in E-textiles due to the textile's porous structure. Cracks may develop in the printed film when curing on the textile's surface. To prevent this, low-viscosity ink formulations can be employed, but they may sacrifice conductivity due to low solid content. Conversely, high-viscosity inks adhere well to the textile surface but may not penetrate the porous structure effectively, resulting in ink film cracks and a loss of flexibility and conductivity during stretching [2]. Recent studies have replaced traditional inks, which use toxic solvents, with eco-friendly alternatives. Although curing water-based inks requires high temperature and time, it offers benefits like reduced volatile organic compounds and improved reactivity. Optimizing the filler-to-binder ratio is crucial due to polymer binders being insulating materials. Consequently, the properties of printed films depend significantly on the formulation's viscosity and content [21]. Given the diverse array of printing technologies available, materials such as inks or pastes and substrates must satisfy specific criteria tailored to the type of printing technology employed and the intended application. The coating material, typically in the form of solutions with specific rheological properties (e.g., viscosity and surface tension), needs to be adjusted to meet the printing technology's requirements. This might involve making it thinner (as ink) or thicker (as paste) as necessary. Furthermore, these materials should facilitate easy printing on a wide variety of substrates. In certain cases, it may be feasible to modify the surface of substrates to enhance ink wetting and ensure strong adhesion of the resultant ink [57]. The most commonly used printing methods are explained below:

Screen printing

Screen printing, a stencil-based method, transfers a design to a flat surface using a mesh screen, typically made from silk or nylon. This technique offers advantages like cost-effectiveness and scalability, although it generates relatively high waste and involves a time-consuming curing step as its primary limitations. Screen printing has low requirements for the physical and chemical properties of conductive ink, making it compatible with a wide range of formulated inks. Additionally, it boasts high versatility, as it is compatible with numerous substrates and design possibilities [30]. A typical screen printer has a screen made of nylon, polyester, silicone, or stainless steel. By applying pressure, the movable squeegee presses the screen into contact with the substrate, facilitating the transfer of the printing paste onto the surface [47]. Screen printing offers cost-effective and environmentally friendly production procedures, allowing for pattern design and large-scale production. It can also be easily applied to a variety of substrates, including plastics, paper, polymers, and textiles [4], [58]. The screen printing technique, known for efficiently transferring materials to predefined surfaces, is widely used in printed electronics [21]. Screen printing is ideal for creating conductive pathways on a surface and is widely used in textile manufacturing. It can create denser networks than conductive yarn, allowing tracks to intersect without electrical connections. It also simplifies integration with planar electronics compared to conductive yarn systems [59]. Screen-printed conductive inks for electronic printing typically consist of three components: conductive particles, binders, and solvents [21].

Stencil Printing

Stencil printing is a simpler and more cost-effective variation of screen printing that eliminates the need for screens and the associated equipment. In stencil printing, ink is applied directly to the substrate through an open mask made from plastic adhesives. While this method is highly cost-effective for immobilizing conductive inks on flexible substrates, it comes with limitations, including high waste generation and a lack of reproducibility. Stencil printing necessitates the use of more viscous conductive inks compared to screen printing to ensure even substrate coverage and prevent ink adhesion to the mask [30].

Inkjet printing

Inkjet printing garners more attention than other printing techniques due to its straightforward printing process, high repeatability, cost-effectiveness, and time-saving features. In inkjet printing, conductive ink replaces traditional ink in the printer cartridge, offering several appealing features such as automation, versatility, speed, minimal waste generation, and reproducibility. Nonetheless, there are notable drawbacks to this approach, primarily stemming from the expensive nature of high-resolution printers and challenges associated with nozzle clogging. Inkjet printing places stringent demands on the physicochemical characteristics of the conductive ink. To be suitable for inkjet printing, the conductive ink must possess specific attributes, including low viscosity within the range of 10 to 20 mPas and high surface tension. These properties facilitate the creation of well-dispersed ink droplets, typically ranging in diameter from 10 to 150 µm, which align with the optimal requirements of the inkjet printing process [30, 47, 60]. In e-textile fabrication, inkjet printing offers advantages over traditional methods like weaving conductive yarn. It allows for precise material deposition, reducing waste and water usage. However, the challenge in inkjet printing for e-textiles is to create continuous, highly conductive tracks on rough, porous fabric. Textiles have inherent property variations due to fiber orientation and changing fiber morphology when exposed to water. This dynamic nature makes achieving uniform, continuous conductive paths with low-viscosity inkjet inks very challenging [61].

Pen Drawing

Pen-drawing offers a simple way to apply conductive ink without generating waste, unlike screen and stencil printing. It typically involves modifying ballpoint pens to dispense conductive ink directly onto the substrate, but it's a manual process with lower reproducibility. Computer-controlled plotters can improve repeatability. This method has minimal requirements for the conductive ink's properties, with drying time being the key consideration. An intriguing variation involves using a paintbrush to spread conductive ink on a larger paper substrate, which is later cut to create the final electrode. This is necessary because the precision of spreading with a paintbrush is considerably lower than that of a ballpoint pen [30].

3D printing

3D printing technology, also referred to as additive manufacturing (AM), enables the creation of intricate topographical structures through layer-by-layer fabrication. It has found applications in various industries, including biomedical, food electronics, and soft robotics. Unlike other printing methods, 3D printing excels in producing complex geometries and multi-compositions [2].

6. PRINTABLE CONDUCTIVE INKS AND THEIR POTENTIAL APPLICATIONS IN FLEXIBLE AND STRETCHABLE E-TEXTILES

Thanks to advancements in nanotechnology and electroactive materials, traditional textile materials have evolved into a versatile wearable electronic platform. This transformation has had a positive impact on the development of flexible electronics, which has been a prominent research focus in recent years. The integration of conductive nanoparticles, such as polymers, metals, or nanocarbons, into textile structures with various properties, including exceptional stretchability and functionality, has enabled the creation of smart textile devices. These devices offer a portable and personalized means for interaction, communication, and sensing within a straightforward framework [55]. The principles described in section 4 about the strategies used for design can be applied to produce a conductive composite ink. The concentration of fillers in conductive colloidal dispersions is a crucial factor influencing the conductivity and mechanical properties of the conductive inks. To achieve a stable dispersion, negatively charged colloids are utilized. For instance, negative charges have been introduced to CNTs using sodium dodecyl sulphate (SDS) to create a water-based PU/CNT colloidal dispersion and produce a conductive composite. Conversely charged polymers are added to negatively charged WPU dispersions through layer-by-layer bonding rather than direct mixing [2]. An overview of conductive inks and their potential applications in flexible and stretchable E-textiles has been given in Table 2. There are limited studies related to water-based conductive inks were conducted [20, 21, 62-64]. In this section, first of all, studies on the characterization of conductive thin films and coatings in which water-based polyurethane is used as matrix material, and then studies on the application of these conductive formulations to textile surfaces and their potential applications in the field of E-textiles are summarized. In some studies, in the literature, PU-based thin films and coatings were produced by using conductive fillers and performance tests of films and coatings were carried out for potential application areas by characterizing them [20, 40, 44, 65-68]. In one study, Ying et al. designed and synthesized a PU (BS-PU-3) with excellent and well-balanced properties, including flexibility, durability, self-healing, and waterproofing, for use in e-skin applications. They subsequently created a demonstration e-skin using PU as the flexible matrix and a liquid metal gallium-indium-tin alloy (galinstan, GaInSn) as the conductive filler. The researchers suggested that the production strategy and resulting material could have wide-ranging applications in the field of e-skin and beyond [67]. Cunha and Pavia (2019) utilized graphite materials to create multilayer graphene (FLG). They examined the distribution of FLG, water vapor permeability, electrical resistance, and mechanical properties of the films produced through solvent casting and spray coating methods [40]. Lie et al. conducted research on the mechanical hysteresis of PU-containing textile substrates and provided recommendations for reducing the electromechanical hysteresis of coated strain sensors. The findings of this study can serve as a valuable resource for researchers aiming to comprehend the structural and mechanical properties, as well as their interplay, in these electronic sensors. This knowledge is essential for the integration of PU-containing textile materials into textile-based electronics designed for wearable applications [68]. Water-based polyurethane/polydopamine (PDA) reduced graphene oxide (WPU/PDRGO) nanocomposites were prepared by Zhang et al. through in-situ emulsification. An analysis of the PDA layer was conducted using Fourier Transform Infrared Spectroscopy (FTIR), X-ray diffraction (XRD), Raman spectroscopy, and thermogravimetric analysis (TGA). It was determined that the interface PDA layers enable the dispersion of PDRGO layers in the WPU matrix, enhancing the mechanical properties of the WPU matrix. The nanocomposites resulting from these water-based polyurethane/graphene nanocomposite dispersions, which have been thoroughly studied, have shown promising results for potential applications in anti-corrosion, antistatic, conductive, and electromagnetic interference protective coatings [66].

Larraza et al. prepared various nanocomposites with different reinforcement types and contents using water-based polyurethane as the matrix and graphene or graphene oxide as the reinforcement. To analyze potential changes and effects on matrix properties, the composites produced in various structures were characterized and evaluated. This included the characterization of nano-entities and an assessment of their impact on composite preparation, as well as the effect of the reinforcement content utilized [65].

Jia et al. reported the preparation of an environmentally friendly conductive ink comprising silver flakes, water-based polyurethane, and a fluorocarbon surfactant, with deionized water as the solvent. This conductive ink can be applied to polyethylene terephthalate film to create a protective coating with an ultra-high electromagnetic shielding effectiveness (EMI SE) of 74.5 dB at just 10 μm thickness. It has been determined that this protective coating exhibits high mechanical durability and chemical resistance to various organic solvents, even under ultrasonic treatment. These remarkable properties indicate that the obtained conductive ink is well-suited for achieving effective electromagnetic shielding in highly integrated equipment and devices [20].

Wang et al. utilized thermoplastic polyurethane (TPU) as the matrix and modified nanocrystalline cellulose (NCC) with chemically reduced RGO as conductive fillers to fabricate flexible conductive films. The study analyzed the relationships between the electrical and thermal properties, tensile strength, and electrothermal response performance of the composite film concerning the mass content of RGO and the initial TPU concentration [44].

In another study, an electrochemical textile-based lactate biosensor was developed using a Graphite-PU paste to enhance the electron transfer rate and chemically modified RGO, coupled via an intermediary material. A conductive silver thread hanging electrode was embroidered onto the fabric substrate to establish a connection between the tailpieces and the device. This smart textile, designed for wearable electrochemical lactate analysis from sweat, demonstrates a relative standard deviation (RSD) of 3.06%. This approach allows for reliable determination of the limits of detection (LOD) and limits of quantification (LOQ) at 0.4 mM and 1.3 mM, respectively [62]. Jia et al. reported the development of a water-based conductive ink for electromagnetic interference (EMI) shielding coating with high mechanical durability and resistance to organic solvents. The coating formulation was prepared using deionized water as a solvent, along with silver flakes, water-based polyurethane, and a fluorocarbon surfactant, resulting in an environmentally friendly conductive ink. The properties of this obtained conductive ink have demonstrated its effectiveness for electromagnetic shielding in highly integrated equipment and devices [20].

Formulations in which graphene nanoparticles (GNPs) are exfoliated in situ in a matrix of water-based polyurethane have been prepared by Hu et al. They created WPU/GNP compositions with varying graphene contents, which were then applied as coatings to flexible cotton fabrics. It has been determined that the fabric coated with a high GNP content can convert heat energy into far-infrared radiation. The applied coating increased far-infrared (FIR) emission to 0.911 within the wavelength range of 4-18 μm. A fabric coated with 0.8% by weight of graphene exhibited a UV protection value 60 times higher than that of pure cotton fabric. This improvement is attributed to the well-balanced and thorough dispersion of nanoparticles within the polyurethane matrix in the coating. Due to the low graphene contents in the coatings applied to the cotton fabric, the UV transmission spectra showed a significant reduction due to the blocking of UV rays. Thus, the electrical, UV blocking, and far-infrared radiation emission properties of functional fabrics obtained with polyurethane/graphene coatings have proven to be suitable for various applications [63].

Table 2. Conductive ink formulations and their application areas.

Material	Method	Potential Application Areas	References
PU/gallium-indium-tin alloy (GaInSn)	Conductive ink printed on PU film with homemade direct ink writing	E-skin and other areas	[67]
WPU/polydopamine (PDA) reduced graphene oxide (PDRGO)	Coating on PET film	Anticorrosive, antistatic, and conductive coatings	[66]
WPU/Ag	Drop Coating on PET Film	High-performance EMI shielding coating	[20]
TPU/Nanocrystalline cellulose (NCC)/RGO	Tape casting	Wearable devices, stretchable antennas, energy devices etc.	[44]
TPU/Ag NWs	Scraper coating on textile and 3D printing	Washable e-textiles	[7]
WPU/Graphene ink	Screen printing on cotton fabrics	Flexible wearable heaters and strain sensors	[69]
PU/Ag NPs	Printing on cotton substrate using a syringe	Wearable electronics	[70]
WPU/MWCNTs/Ag	Screen printing on WPU substrate	Resistive strain sensor for wearable electronics	[71]
TPU/MWCNTs	Screen printing on polyamide woven fabric	Wearable E-textiles	[72]
TPU/GNPs	Screen printing	Skin-compatible wearables e.g., motion sensors, heart rate monitors etc.	[73]
Poly(vinyl butyral-co-vinyl alcohol-co-vinyl acetate)(PVBVA)/Graphene	3D printing square mesh pattern	Smart wearable electronics for healthcare and soft robotics	[74]
Silver -cladded copper NPs/2D Graphene/PU ink	Steel plate stencil printing	Smart wearable devices for EMG and ECG signals	[52]
Di alcohol cellulose/glycerol/PEDOT: PSS bio-ink	3D printing	Wearable supercapacitors and biopotential monitoring devices (ECG and EMG)	[75]
AgCu/epoxy resin paste	Screen printing on cotton, PET and PES/spandex fabrics	Solderable conductive paste for E-textiles	[76]
PU/Ag micro-particles	Direct ink wring system on TPU foil for textile substrates (cotton twill, elastic twill, and swimwear)	Stretchable electronics	[77]
Ag and C inks	Inkjet printing on polyimide (PI), poly (vinyl butyral-co-vinyl alcohol-co-vinyl acetate) (PVB) and polystyrene (PS) membranes	Flexible and smart textiles	[78]
Solution based Cu complex ink	Screen printing on knitted fabrics	Fabricating e-textiles such as wearable gloves	[79]
Starch/$AgNO_3$ conductive ink	Inkjet printing on PU coated cotton fabrics	Wearable electronics applications	[80]
MWCNTs/WPU	Screen printing on WPU substrate	Wearable electronics for medical health and human-computer interaction	[34]
RGO electroconductive layers	Inkjet printing on PAN, PET, and PP textile substrates	Supercapacitors	[81]

Yan et al. developed hybrid membranes made of polypyrrole/zirconium carbide/polyurethane (PPy/ZrC/PU), which exhibit photothermal and electrothermal conversion properties. Based on the results obtained in this study, it is indicated that PPy/ZrC/PU hybrid membranes have the potential to advance the development of thermal comfort textiles, evaporators, sensors, and similar application areas [64].

In another study, Franco et al. prepared electrically conductive graphene inks using carboxymethyl cellulose (CMC) as a water-based binder for the development of screen-printable water-based inks. This study showcases the potential of composites and the printing technique for multifunctional composites. Additionally, the inks have been examined and optimized not only for the production of conductive films through printing but also to enhance their piezoresistive and thermoresistive responses, thereby evaluating their potential for the development of printable sensors [21].

7. CONCLUSION

In this review, comprehensive information on conductive materials and inks, their associated printing techniques, and current research findings in the literature aiming to develop flexible printed electronic devices for potential applications in wearable textiles are provided.

So far, there has been significant research interest in developing a wide range of conductive inks using carbon and metal nanomaterials as well as conductive polymers. These inks have proven to be viable options for creating flexible, stretchable, and wearable printed electronic devices. It's worth noting that once these formulated conductive inks are applied to textiles, they may lose their electrical conductivity. Therefore, additional processes are required to restore and maintain conductivity after deposition. When it comes to printing these inks, it's evident that the current research efforts are still in their early stages, primarily focusing on demonstrating the basic printability and conductivity of the inks. Printed textiles are preferred for printed electronics due to their flexibility and washability. An essential step in this process involves studying the porosity, wettability, and surface roughness of the fabric. These factors determine whether additional support is needed. In general, it is observed that due to the porous nature of textile supports, pretreatment is necessary to facilitate the continuous formation of an electronic path when printing with inkjet technology on the fabric. E-textile materials and devices can be developed using a variety of metals, conductive polymers, carbon allotropes, and 2D materials, offering numerous advantages but also presenting unique challenges. Carbon allotropes and metals, in particular, exhibit high conductivity, but their stiffness makes them unsuitable for flexible textiles. However, when used in combination with natural materials such as synthetic polymers, silk, or cellulose, it is possible to create flexible and stretchable materials with high wash and abrasion resistance, making them ideal for textile production. Presently, ink-to-textile printing technologies primarily rely on inkjet printing, but they come with limitations regarding ink compatibility. While certain printing technologies can handle inks with specific rheological properties, ensuring high-quality prints, especially at high speeds, remains a common challenge. Three-dimensional printing (3D) technology, on the other hand, presents itself as an additive technique that brings economic efficiency, sustainability, waste reduction, and innovative product design, offering versatility in material selection and introducing novel concepts to the printing landscape. Experts argue that 3D printing theoretically enhances rapid prototyping, concept development, reliability, and cost reduction when compared to traditional printing methods. In the textile sector, 3D printing technology emerges as an innovative fashion avenue, enabling the projection of diverse emotions and aesthetics onto substrates. In terms of ink formulation, 3D printing covers a wide spectrum of processes for creating three-dimensional models by incrementally adding materials and liquids to solids, layer by layer. Due to its relevance in the modern textile industry, current research is increasingly drawn to 3D printing technology as a promising prospect for fashion-related industries. As previously mentioned, it is evident that the utilization of conductive inks in conjunction with various printing methods and substrate materials offers a straightforward and cost-effective means to create and advance virtually any electronic device. Consequently, the potential impact of conductive inks on the future of the electronics industry is anticipated to undergo significant and swift expansion. Future research should focus on designing hybrid materials, incorporating metals, polymers, and 2D materials with suitable electrical, electronic, and electrochemical properties that can be easily separated into their respective components. To facilitate the development of sustainable e-textile materials, it would be advantageous to work with a limited number of materials. Sustainability should be a primary consideration throughout the entire lifecycle of e-textiles, encompassing production, use, waste collection, and recycling. Furthermore, the use of solvent-based compounds and solvents in product development should be minimized to safeguard the health of employees and e-textile users potentially exposed during production and recycling phases.

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