Melding Vapor-Phase Organic Chemistry and Textile Manufacturing To Produce Wearable Electronics

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CONSPECTUS: Body-mountable electronics and electronically active garments are the future of portable, interactive devices. However, wearable devices and electronic garments are demanding technology platforms because of the large, varied mechanical stresses to which they are routinely subjected, which can easily abrade or damage microelectronic components and electronic interconnects. Furthermore, aesthetics and tactile perception (or feel) can make or break a nascent wearable technology, irrespective of device metrics. The breathability and comfort of commercial fabrics is unmatched. There is strong motivation to use something that is already familiar, such as cotton/silk thread, fabrics, and clothes, and imperceptibly adapt it to a new technological application.24 Especially for smart garments, the intrinsic breathability, comfort, and feel of familiar fabrics cannot be replicated by devices built on metalized synthetic fabrics or cladded, often-heavy designer fibers. We propose that the strongest strategy to create long-lasting and impactful electronic garments is to start with a mass-produced article of clothing, fabric, or thread/yarn and coat it with conjugated polymers to yield various textile circuit components. Commonly available, mass-produced fabrics, yarns/threads, and premade garments can in theory be transformed into a plethora of comfortably wearable electronic devices upon being coated with films of electronically active conjugated polymers. The definitive hurdle is that premade garments, threads, and fabrics have densely textured, three-dimensional surfaces that display roughness over a large range of length scales, from microns to millimeters. Tremendous variation in the surface morphology of conjugated-polymer-coated fibers and fabrics can be observed with different coating or processing conditions. In turn, the morphology of the conjugated polymer active layer determines the electrical performance and, most importantly, the device ruggedness and lifetime. Reactive vapor coating methods allow a conjugated polymer film to be directly formed on the surface of any premade garment, prewoven fabric, or fiber/yarn substrate without the need for specialized processing conditions, surface pretreatments, detergents, or fixing agents. This feature allows electronic coatings to be applied at the end of existing, high-throughput textile and garment manufacturing routines, irrespective of dye content or surface finish of the final textile. Furthermore, reactive vapor coating produces conductive materials without any insulating moieties and yields uniform and conformal films on fiber/fabric surfaces that are notably wash- and wearable and can withstand mechanically demanding textile manufacturing routines. These unique features mean that rugged and practical textile electronic devices can be created using sewing, weaving, or knitting procedures without compromising or otherwise affecting the surface electronic coating. In this Account, we highlight selected electronic fabrics and garments created by melding reactive vapor deposition with traditional textile manipulation processes, including electrically heated gloves that are lightweight, breathable, and sweat-resistant; surface-coated cotton, silk, and bast fiber threads capable of carrying large current densities and acting as sewable circuit interconnects; and surface-coated nylon threads woven together to form triboelectric textiles that can convert surface charge created during small body movements into usable and storable power.

INTRODUCTION

Body-mountable electronics and electronically active garments are the future of portable, interactive devices.1 Recent reports of wearable devices and garments that allow advanced physiological and performance monitoring,2−5 new touch/user interfaces,6−8 portable power generation,9 and energy storage10 represent a few of the sophistications promised by these nascent technologies. However, wearable devices and electronic garments are demanding technology platforms. Wearable devices are subject to large, varied mechanical stresses that can easily abrade or damage microelectronic components, particularly electronic interconnects.11 Consequently, soft electronic materials, particularly conjugated organic polymers, are enabling electronic
components in many body-mounted and garment-based devices because their flexibility, stretchability, low density, and undemanding processing requirements allow them to be practically integrated into nontraditional device platforms.\textsuperscript{12}

Wearable devices composed of soft electronic materials are typically created on strips of thin or ultrathin plastic substrates.\textsuperscript{13} Electronic “textiles” are often made using either metalized synthetic fabrics\textsuperscript{4,15} (e.g., silver- or copper-coated polyesters) or specialty low-throughput-manufactured fibers\textsuperscript{16} with artificially smoothed surfaces, such as diamond-tip-extruded stainless steel thread\textsuperscript{17} or extruded/electrospun conductive composite fibers composed of graphene, carbon nanotubes, or metal nanowires.\textsuperscript{18–21} These designer substrates are coated first with active-layer materials and second with a thin protective polymer layer (cladding), and then these cladded fibers are yarned together to approximate the threads and yarns found in clothing.\textsuperscript{21–23}

Notably, aesthetics and tactile perception (or feel) can make or break a nascent wearable technology, irrespective of device metrics. The breathability, comfort, and feel of commercial fabrics is unmatched. There is strong motivation to use

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**Figure 1.** (a–e) Survey of conjugated-polymer-coated prewoven fabrics created using various deposition methods, including (a) in situ solution polymerization, (b) spin coating (adapted with permission from ref 26; copyright 2017 Royal Society of Chemistry), (c) electrochemical deposition (adapted with permission from ref 25; copyright 2017 Royal Society of Chemistry), (d) dyeing (adapted with permission from ref 25; copyright 2017 Royal Society of Chemistry), and (e) reactive vapor deposition (adapted with permission from ref 26; copyright 2017 Royal Society of Chemistry). (f) Scanning electron micrograph of the polymer-coated fabric swatch in (e). Adapted with permission from ref 26. Copyright 2017 Royal Society of Chemistry. (g) Change in sheet resistance of the polymer-coated fabric swatch from part (e) after exposure to mechanical stresses. Adapted with permission from ref 26. Copyright 2017 Royal Society of Chemistry.

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**Figure 2.** Two different reaction chambers used to vapor-coat mass-produced prewoven fabrics and threads/yarns with soft electronic materials.
something that is already familiar, such as cotton/silk thread, fabrics, and clothes, and imperceptibly adapt it to a new technological application. Especially for smart garments, the intrinsic breathability, comfort, and feel of familiar fabrics cannot be replicated by devices built on metalized synthetic fabrics or cladded, often-heavy designer fibers, no matter how thin or flexible those specialized textiles are.

We propose that the strongest strategy to create long-lasting and impactful electronic garments is to start with a mass-produced garment, fabric, or thread/yarn and coat it with conjugated polymers to yield various textile circuit components. The definitive hurdle is that premade garments, threads, and fabrics have densely textured, three-dimensional surfaces that display roughness over a large range of length scales (microns to millimeters). Therefore, the coating method chosen to create the electronic polymer coating will be uncommonly influential over the device feasibility and performance. To date, dip-coating (i.e., dyeing), in situ solution polymerization, solution-phase surface grafting, and electropolymerization are primarily used to coat familiar textiles and threads/yarns with conjugated polymer films (Figure 1). Although these methods afford suitable samples for academic research, the coatings thus obtained are batch-dependent, nonuniform, and susceptible to mechanically induced degradation and abrasion. Moreover, solution-processed coatings often obfuscate the intrinsic flexibility, breathability, and tactile perception of fabrics.

In this Account, we summarize recent efforts to use vapor-phase chemistry to transform off-the-shelf fabrics and threads into electronic circuit components. Reactive vapor coating methods allow a uniform conjugated polymer film to be directly formed over large areas on the surface of any premade garment, prewoven fabric, or fiber/yarn substrate without the need for...
specialized processing conditions, surface pretreatments, detergents, or fixing agents. This feature means that electronic coatings can be nondisruptively applied at the end of existing high-throughput textile and garment manufacturing routines. Vapor-deposited coatings are conformal and are often thin enough that the original mechanical properties of the substrate (and not those of the coating) are the dominant observables. Furthermore, reactive vapor deposition produces conductive coatings that are notably wash- and wear-stable and can withstand mechanically demanding textile manufacturing routines. Selected textile electronic devices created by melding vapor coating and sewing, weaving, or knitting routines are described.

**DISCUSSION**

1. Apparatus for Vapor Coating of Fabrics and Threads/Yarns

Reactive vapor growth of conjugated polymer films is achieved by translating well-known solution-phase organic chemistry into the vapor phase. For manageable reactor design and systematic mass transport optimization, candidate reactions ideally should involve less than three reactants that are not susceptible to thermal degradation and display minimal side reactions between reactants. Oxidative polymerization, wherein an electron-rich heterocyclic monomer is mixed with a superstoichiometric amount of an oxidant, such as iron(III) chloride or molecular bromine, to produce a conjugated polymer via step-growth mechanism, is the underlying chemical reaction used to create the conducting and semiconducting polymer coatings described herein. This bicomponent reaction is ideal for translation to the vapor phase because the monomers and oxidants are evaporable or sublimable and the operative redox reaction that results in polymer formation is diffusion-limited, meaning that electron transfer between the monomer and oxidant occurs spontaneously when the two reactants are within van der Waals contact.

Two different reaction chambers, denoted as chambers 1 and 2, are used to vapor-coat mass-produced fabrics and threads with conducting or semiconducting conjugated polymer layers (Figure 2). To effect vapor coating, the monomer and oxidant are independently heated and introduced as vapors into
Chamber 2 is specifically designed with globally smaller path lengths to encourage mixing of monomer and oxidant vapor plumes and allow vapor-phase oxidative polymerization of solid, low-volatility monomers. Because of this feature, chamber 2 allows a large range of known solution-phase bicomponent chemical reactions to be translated into the vapor phase.26

Typical polymer growth rates in both chambers 1 and 2 are 10−15 nm/min at a substrate stage temperature of 80 °C. Higher substrate stage temperatures lead to comparatively retarded growth rates due to an increased frequency of monomer desorption from the substrate stage and subsequent transport away from the reaction zone. For example, polymer films grow at a rate of 8−10 nm/min at a stage temperature of 150 °C. Higher-molecular-weight polymer films are obtained when the evaporation rate of the oxidant is higher than the monomer flow rate.26−28 During a typical deposition, the monomer flow rate is maintained at 1 Å/s while that of the oxidant is maintained at 2 Å/s. As a result of the presence of excess oxidant during deposition, the polymer coatings obtained using either chamber 1 or 2 are p-doped, irrespective of monomer identity (Figure 3). A postdeposition rinse (5−60 min) with methanol and/or dilute aqueous sulfuric acid completely removes residual oxidant trapped in the vapor-deposited coatings and yields metal-free conjugated-polymer-coated fabrics (Figure 3c,d). Selected electron-rich monomers, such as 3,4-dialkoxythiophenes, anilines, and pyrroles, remain p-doped even after the postdeposition rinsing step and are optically observed to retain their polaron/bipolaron absorption features in the visible—near-infrared region even after indefinite exposure to ambient conditions. The coatings thus produced are significantly conductive, displaying conductivities higher than 300 S/cm.26,27,29,30 Other monomers, such as alkyl-substituted thiophenes and oligothiophenes, are rapidly dedoped during the postdeposition rinsing step and yield colorful, semiconducting coatings (Figure 3b,d). For example, a polaron absorption band between 600 and 900 nm is observed in films of poly(thienothiophene) (PTT) immediately after deposition (Figure 3e), which arises because of the presence of p-doped (polaronic) species. This absorption feature quickly disappears after rinsing with methanol, indicating dedoping.26

A thoroughly characterized and comparatively mature process termed initiated chemical vapor deposition (iCVD),31 which controllably creates various insulating polymer films (e.g., poly(acrylate)s, among others) via a radical-initiated chain growth polymerization reaction, can be used to vapor-deposit protective insulating coatings on readily available fabrics and fibers. Depending on the choice of monomer, the hydrophobicity of the starting fabric/fiber can be significantly altered.32 Importantly, fibers and fabrics coated with a conducting or semiconducting polymer layer using chamber 1 or 2 can subsequently be subjected to vapor coating in chamber 3 to create a protective insulating layer (cladding) without deteriorating the electrical properties of the underlying electronic polymer layer.33

2. Coating Prewoven or Knit Fabrics

A varied selection of readily available, mass-produced, prewoven or knit fabrics comprising either synthetic or natural threads have been vapor-coated with conjugated polymer films using either chamber 1 or 2 (Figures 4 and 5).26,27 No differences in film morphology, conformality, or uniformity can be observed in polymer coatings created using either chamber 1 or 2. Polymer coatings with thicknesses between 100 nm and 1 μm can be controllably created in either chamber (even on rough fabric
surfaces) by varying the deposition time. To quantify the thickness of the polymer coating on fabric substrates, a silicon “test coupon” is typically coated alongside the target fabric, and the thickness of the polymer coating on this test coupon measured using a profilometer after rinsing; the measured coating thickness on this test coupon is assumed to match the thickness of the conformal polymer coating on the fabrics. Vapor coating (up to 1 μm thick) does not change the feel of any of the fabrics, as determined by touching the fabrics with bare hands before and after coating. The porosity of the fabrics, as determined by measuring air flow through a single 4 in.² swatch of a sample before and after coating, remains unchanged, indicating that the fabrics retain their breathability after vapor coating. Further, the coating does not increase the weight of the fabrics by more than 2%.

Large-area scanning electron microscopy (SEM) images of selected pristine fabrics reveal the interwoven structure of several threads, each of which is composed of fibril bundles as shown in small-area images. Highly uniform and conformal coatings are formed on all of the fabrics investigated to date, irrespective of porosity, knit/weave pattern, and thread composition, without the need for specialized algorithms during deposition. Furthermore, conjugated polymer films are uniformly deposited over the entire surface of the fabric while also conformally wrapping the curved surface of each exposed fibril of the threads constituting the fabric. 26,27 The high conformality of the surface coating is particularly apparent in the SEM images of fuzzy corduroy fabric coated with the semiconducting polymer PTT using chamber 2 (Figure 5c,d). Here the polymer film conforms to all of the exposed surface features of the fabric with high fidelity over multiple length scales and varied surface features. Cross-sectional SEM images confirm that the vapor-deposited polymer film is purely a surface coating and that the bulk of fibrils/threads is not swelled or impregnated with polymer/oligomers (Figure 4b).

The large-area electrical properties of the conjugated polymer coating are influenced by the constituent threads/fibers (linen vs silk vs wool, etc.) of the underlying textile or garment substrate. 27 Ordered and/or single-ply constituent threads, such as nylon, fine silk, and banana or pineapple fiber, lead to fabric electrodes with high large-area surface conductivities (as low as 44 Ω/□) when coated with a 1 μm-thick film of the p-doped conducting polymer poly(3,4-ethylenedioxythiophene) (PEDOT). 33 In contrast, disordered fibers and thick, multi-ply yarns, such as those found in wool and cotton twill, lead to comparatively higher surface resistances.

Any fabric or garment, irrespective of surface chemistry, thread/yarn composition, and weave density, can be vapor-coated without the need for pretreatment steps. To wit, a commercial rain jacket composed of an uncharacterized, proprietary hydrophobic fabric was successfully coated with a 500 nm-thick film of p-doped PEDOT without any extra preparatory steps (Figure 4e). Patterned electronically active polymer coatings can also be created on arbitrary fabrics or premade garment surfaces using straightforward shadow masking. The observed lower limit of patternable features on a premade garment is 1 mm in chamber 2 and 0.1 mm in chamber 1. An untreated, commercial white sports bra vapor-coated with p-doped PEDOT through a plastic shadow mask is shown in Figure 4f.

Process pressures between 100 and 500 mTorr during deposition (in either chamber 1 or 2) translate into mean free paths on the order of millimeters for the reactive oligomers. Since these mean free paths are commensurate with the surface
roughness of woven fabrics, the oligomers should be able to sample multiple sites before finally adhering to a particular surface, yielding conformal coatings. In the case of prewoven fabrics, tuning the chamber pressure between 100 and 500 mTorr during vapor coating allows control over the percent of the total surface area that is polymer-coated (Figure 6). At 100 mTorr, one face of a prewoven fabric swatch can be selectively coated while the other side remains uncoated (effectively, 50% of the exposed surface is coated). Under these conditions, the weave density of the prewoven fabric attenuates the observed surface conductivity of the coated textile, with highly porous, airy fabrics (e.g., linen gauze and pressed silk) yielding the lowest surface resistances and tightly woven fabrics (e.g., twill or cotton) yielding the highest surface resistance values. Increasing the chamber pressure to 500 mTorr lowers the average mean free path of reactive oligomers in the vapor phase and encourages coupling of oligomers that are already adhered to a substrate surface, which effects greater surface coverage of both fabric faces (>90% of the exposed surface is coated). Such selective control over the coating area cannot be matched by solution-based approaches such as dyeing and electropolymerization.

3. Threads and Yarns

Threads and yarns can also be vapor-coated with conjugated polymers in either chamber 1 or 2. A specially designed substrate stage is used to introduce threads/yarns into the reaction chamber of choice and to achieve complete circumferential surface coverage in one deposition. A cartoon of the stage design for threads/yarns is shown in Figure 7. This design was meant to mimic an array of thread spools and can accommodate, on average, 30 ft lengths of thread when carefully wound in vertical layers. Complete circumferential surface coverage is observed for both ordered, single-ply fibers, such as extruded nylon, and disordered, fuzzy, multi-ply yarns, such as a thick cotton yarn typically used to knit winter garments. Figure 7 shows optical micrographs of one such cotton yarn before and after vapor coating. Interestingly, coated wool yarns retain their characteristic terraced surface features even after conjugated polymer coating (Figure 7e). Wool fibers swell when exposed to body heat and perspiration as a result of water penetration into the fibers through these surface terraces. The water-swelled fibers display greater heat retention near the body surface, and thus, wool-based fabrics and garments are optimal for thermal insulation in cold climates. Therefore, the observation that vapor-coated wool yarns retain their scaly surface features indicates that these electronically active wool yarns will likely display the same thermal insulation properties as familiar uncoated wool.

4. Textile Devices

Chambers 1 and 2 are used to coat off-the-shelf plain-woven fabrics, such as linen, silk, and bast fiber fabrics, with conducting polymer films (e.g., persistently p-doped PEDOT), thus transforming these mass-produced textiles into metal-free conducting electrodes. The vapor-deposited polymer coatings...
are insensitive to light bending and creasing and remarkably resistant to harsh mechanical abrasion (Figure 8). The conductivities of the PEDOT-coated fabrics are attenuated by less than 10% after laundering with household laundry detergent (up to eight “normal wash” cycles), dry ironing, and continuous body mounting for 24 h. Notably, such wash stability is not matched by commercial metal-coated fabrics and threads and is a major, explicitly defined requirement to progress commercialization and adoption of electronic textiles and garments.

The large-area resistivities of the textile electrodes are linearly, not exponentially, dependent on length, meaning that they can be feasibly incorporated into garments and other large-area body-mounted devices. Moreover, these fabric electrodes possess the feel, weight, breathability, and pliability of standard fabrics, which is important to enable adoption of wearable devices.

The vapor-deposited conjugated polymer coatings are, furthermore, robust enough to endure conventional textile manufacturing routines, such as cutting, sewing, weaving, and knitting, while maintaining their original optical and electronic properties. For example, vapor-coated cotton yarns can be plain-woven using a table-top loom into a monolithic textile without abrading or deteriorating the polymer layer during the weaving process (Figures 7 and 9). Vapor-coated yarns can also be knitted, complex-woven, or embroidered into different structures and shapes, just like regular yarns, meaning that electronic devices can be created using traditional textile manufacturing routines.

Chamber 1 is used to transform commercial textiles and threads into electric heaters that can be cut/sewn or woven to fashion lightweight fabric heaters for local climate control and personal thermal management (Figure 9). Prewoven fabrics coated with a 1.5 μm-thick film of persistently p-doped PEDOT possess competitively low sheet resistances—44 Ω/□ measured for coated bast fiber textiles and 61 Ω/□ measured for coated cotton textiles (Figure 6)—and act as low-power-consuming Joule heating elements. The electrothermal response of the textile electrodes remains unaffected after cutting and sewing because of the robustness of the vapor-deposited conductive coating. This feature was used to create a lightweight, breathable, electrically heated glove. Coated, conductive cotton yarns can also be plain-woven into a monolithic fabric heater (Figure 9).

Conductive yarns coated with a fluorinated insulating polymer overlayer can also be knitted or woven together with nylon threads to create triboelectric textiles that convert surface charge.
resulting from contact electrification during fabric movement into usable and storable power (approximately 13 μW).34

**CONCLUSIONS AND OUTLOOK**

Having varied approaches for making wearable devices is integral to grow this nascent technology to maturity. We propose that the strongest strategy to create long-lasting and impactful electronic garments is to start with a mass-produced garment, fabric, or thread/yarn and coat it with conjugated polymers to yield various textile circuit components. The definitive hurdle is that premade garments, threads, and fabrics have densely textured, three-dimensional surfaces that display roughness over a large range of length scales (microns to millimeters), which render conventional solution-based printing methods useless for creating reliably uniform, electronically active coatings on these surfaces.

Reactive vapor coating methods24,25,35,36 allow a uniform conjugated polymer film to be directly formed over large areas on the surface of any premade garment, prewoven fabric, or fiber/yarn substrate without the need for specialized processing conditions, surface pretreatments, detergents, or fixing agents. This feature means that electronic coatings can be non-disruptively applied at the end of existing high-throughput textile and garment manufacturing routines. At present, highly conductive prewoven bast fiber fabrics with sheet resistances of 44 Ω/□ can be reliably created using reactive vapor coating.33 Vapor-deposited coatings are conformal and are often thin enough that the original mechanical properties of the substrate (and not those of the coating) are the dominant observables. Furthermore, reactive vapor coating produces conductive coatings that are notably wash- and wear-stable and can withstand mechanically demanding textile manufacturing routines. In summary, reactive vapor coating can transform commonly available, mass-produced fabrics, yarns/threads, and premade garments into a plethora of comfortably wearable electronic devices.

Textile scientists have traditionally shied away from vapor deposition methods to create various textile-based electronics because of the perceived difficulty and high cost of scaling up vapor coating techniques to satisfy the high-volume demand of the textile industry. However, advancements made over the past decade have resulted in the use of vapor deposition methods to stain-guard carpets, lubricate large-area mechanical components, and protect microelectronic devices, demonstrating that vapor coating methods are indeed conducive to large-scale manufacturability.37 At present, the lab-scale reactors depicted in Figure 2 need to be scaled up to demonstrate commodity-scale manufacturability. Most importantly, substrate stages that can produce multiple kilometers of coated threads or yarns or multiple bolts of coated fabrics per day will be needed before vapor coating can be practically added to existing textile manufacturing routines.

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**Notes**

The authors declare no competing financial interest.

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