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Abstract

Printing of conductive inks is an attractive means of electronic component manufacturing on flexible substrates including textiles. Highly conductive inks are preferably made from metal nanoparticles such as silver. However, such inks are expensive and generally not known to withstand severe washing and wearing to which textiles are normally subjected to during the end use. In the present study, which forms a part of a larger study by the authors, dispersions of conductive grades of carbon black were formulated. These dispersions are converted into finished inks and are found to be very stable after let down with different binders. The final pigment loading in all of the formulated inks was significantly less than the pigment loading that is generally found in commercial conductive inks. In addition, the electrical conductivity after washing and creasing tests of the formulated inks was found to be significantly greater than that of the tested commercial conductive inks.

Keywords

Conductive inks, textile printing, e-textiles, carbon black

1. Introduction

The development of wearable electronics technology has been/and is fuelled by the increasing desire for ubiquitous smart systems^{1, 2}. Many of the claimed prototypes of interactive electronic textiles are based on integrated wiring to which the devices are attached, adding bulk and weight to the garments which make them uncomfortable and impractical for most of the intended uses. In the case of truly wearable e-textiles, it is important to ensure that the hardware integrated into clothing is unobtrusive, washable, and does not impede the garment's ability to conform to body curvatures^{3,4}.

Using conductive wires or filaments in various forms such as yarns, threads, cables or ribbons is the most widely reported method of component fabrication in textile fabrics⁵⁻¹⁰. Some of these techniques are reportedly used in commercial e-textile prototypes. However, there are limitations. Some techniques restrict circuits to specially prepared fabric. Some require the use of specially prepared electrical components and add bulk to the final article. Others cannot be practically automated or mass produced. In addition, virtually all of the techniques based on the use of conductive filaments involve multiple, very complex manufacturing processes, which is arguably the most significant drawback of such techniques.

In contrast, printing allows the one-step formation of an intricate design and thus, offers several obvious advantages when it comes to realization of electronic components on a flexible substrate such as a textile. The availability of an ink that possesses the desired electrical characteristics is a fundamental consideration in the case of printed electronics. It is imperative to mention here that the technology of electrically conductive inks is not new and several producers manufacture electrically functional inks that are frequently employed to develop passive electronic components such as resistors, capacitors, interconnections and so on, as well as the more sophisticated electronic devices such as transistors, displays, etc., on

flexible plastic substrates¹¹⁻¹⁴. However, a number of factors pose great challenges for successful printing of electronic devices on textiles. Firstly, in contrast to the flexible substrates that are commonly employed in the printed electronics industries, for instance polyimide films and polyester films, textile fabrics in general and woven textile fabrics in particular possess a very rough and porous surface. Thus, the lack of ink film integrity becomes a limiting factor in achieving the desired functionality. Secondly, the durability requirements, such as the ability to withstand several washing and wearing cycles, are stringent for inks that are printed onto textile substrates^{15, 16}.

For high conductivity applications, for instance printed interconnections, metallic inks are the obvious choice. Inks containing silver nano-particles or silver flakes can be used for e-textiles fabrication by conventional printing techniques¹⁷⁻¹⁹. To overcome critical problems, such as poor mechanical strength of ink film, poor ink film integrity due to substrate surface roughness, elaborate methods of using one or more layers of polymeric binders can be adopted^{20, 21}. Another approach to produce a conductive pattern is to deposit a metal precursor followed by a second step in which elemental metal is produced *in-situ* on the substrate by a suitable process such as reduction. This method was used to produce electrically conductive patterns of silver on various substrates including a woven cotton fabric by ink-jet printing²².

The recent discoveries of high performing materials such as graphene have triggered the interest of research community in employing these new materials in numerous applications, including the formulation of electrically conductive inks. In one such study, inkjet printable electrically and thermally conductive ink was formulated from nano-graphene platelets²³. Other forms of carbon-based materials, such as CNTs, have also been used in fictionalization

of textiles ^{24, 25}. The use of CNTs in silver nano-particle inks is known to reduce silver concentration drastically, which could be beneficial in terms of ink film flexibility ²⁶.

Electrically conductive polymers are also a strong candidate for use as conductive filler materials in inks/coatings. One of the more widely reported methods of producing electrically conductivity textile yarn/fabrics is the in-situ polymerization of an intrinsically conductive polymer onto a textile material ²⁷. One such method was reported to produce conductive textiles uniformly coated with doped polypyrrole or polyaniline ²⁸. In another study, the generally poor mechanical properties of polyaniline (PANI) were dealt with by adding polyethylene glycol (PEG) during in-situ polymerisation of the conductive polymer onto a polyester fabric ²⁹. However, blending PANI with PEG was found to considerably reduce the conductivity of the conductive fabric.

A number of studies that are summarized in the preceding text reveal the use of conductive inks, comprising of different conductive filler materials, in the formation of conductive prints/coatings. This review shows that the available techniques suffer from one or more of the following limitations.

- a. High loading of metallic fillers usually renders the ink film brittle and impairs the adhesion with flexible substrates such as textile fabrics.
- b. Increasing trend in the cost metals is a major limitation to large scale application of such inks.
- c. In some cases, for instance when metal nano-particles are synthesized *in-situ*, the final conductive layer is produced by depositing several layers of various reagents. Thus, the fabrication process is either too complex or involves several steps.
- d. In case of non-metallic conductive fillers, the electrical conductivity achieved is rather low in many cases.

- e. Furthermore, in a number of cases, performance testing in terms of washing and creasing tests were not carried out and thus the durability of the produced conductive patterns could not be established.

In contrast to other conductive filler materials, carbon black offers the advantages of being relatively low cost and easily processable. In addition, the electrical conductivity of the ink/coating can be adjusted in a broad range with relative ease. In the first phase of this research, dispersions of conductive carbon black pigments were produced, optimized and characterized³⁰. In this part of the study, these dispersions were formulated into finished inks followed by application and performance testing on textile substrates in order to investigate if one or more of the aforementioned limitations can be overcome by using specific grades of carbon black in printing inks.

2. Materials

2.1 Pigment dispersions

A 4-step procedure, as described in a separate study by the authors³⁰, was followed to prepare waterborne dispersions of non-printing ink grades of carbon black pigments. The dispersion formulations containing the pigment, the dispersant and vehicle (water) were subjected to bead milling for an appropriate length of time and under optimised conditions as determined in another study by the authors³¹. The twelve pigment-dispersant combinations, listed in Table I, were used to formulate finished inks which were printed onto textile fabrics and subsequently tested.

Table I: Pigment-dispersant combinations prepared and tested in the 1st phase of study.

Dispersant name (Code*)		
Solsperse 44000	BYK-190	Tego 760W

		(Dispersant1)	(Dispersant2)	(Dispersant3)
Pigment name (Code*)	Vulcan XC605 SA** = 59 m ² /g (Carbon1)	Dispersion 1 (C1D1)	Dispersion 2 (C1D2)	Dispersion 3 (C1D3)
	Ensaco 250G SA** = 62 m ² /g (Carbon2)	Dispersion 4 (C2D1)	Dispersion 5 (C2D2)	Dispersion 6 (C2D3)
	Ensaco 350G SA** = 770 m ² /g (Carbon3)	Dispersion 7 (C3D1)	Dispersion 8 (C3D2)	Dispersion 9 (C3D3)
	Printex XE2B SA** = 1000 m ² /g (Carbon4)	Dispersion 11 (C4D1)	Dispersion 10 (C4D2)	Dispersion 12 (C4D3)
*Code refers to the symbol used in this study for a product instead of its full commercial name ** BET Surface area				

2.2 Binders

In this study, polymeric binders were used for one or more of the following: for the let-down of optimized pigment dispersions; and in the primer coating of textile substrates. Three polymeric binders, which represent the types commonly used in inks/coatings for textiles, were selected. The available details for these binders are provided in Table II. In order to calculate the amount of binder that was required to formulate the finished inks from the optimized pigment dispersions, the solid content of each of the three binders was determined using thermogravimetric analysis.

Table II: Binders used in the study.

Binder (Code)	Supplier	Description	Solids content (wt %)
Impranil DLC-F (Binder1)	BASF	Anionic polycarbonate ester polyurethane dispersion	40
Impranil LP GHG 519 (Binder2)	BASF	Anionic aliphatic polyester- polyurethane dispersion	40
Printofix Binder 83 liquid (Binder3)	Clariant	Aqueous dispersion of a styrene- acrylic copolymer	40

2.3 Commercial conductive inks

The surface resistivity and the durability (resistance to washing and creasing) of the inks that were formulated in this study were compared against two commercially available carbon based inks that were kindly supplied by the respective manufacturers. The surface resistivity values that are shown in Table III relate to an ink layer that has a dry film thickness of 25 microns and deposited on a polyimide film. The solids content of these inks was determined using thermogravimetric analysis.

Table III: Commercial conductive inks used in the study.

Product	Supplier	Surface resistivity (Ω/\square)	Solids content (wt %)
C2030519P4 Carbon/Graphite ink	Gwent Electronic Materials Ltd	13 -20	40
SD 2843 HAL	Peters GmbH	10	73

2.4 Substrates

The substrates on which the printing of conductive patterns was targeted in this study were textile fabrics. There are number of physical and chemical properties of a textile fabric that might influence the resultant print quality and, in turn, influence the performance/behaviour of the printed patterns. These include the surface smoothness, the absorbency, the surface energy, the fabric construction (weave design, warp and weft count in case of woven fabrics), the GSM, the porosity (related to fabric construction and GSM) and the resistance to acidic/alkaline media. These substrate-dependent variables are important. None-the-less this study was focused on achieving high electrical conductivities in ink formulations. Thus, the testing of substrate-related variables was beyond the scope of this study and some of the tests were carried out on calcium carbonate clay coated paperboard substrate of having a weight per unit area of 240 g/m².

For performance testing of inks on textile substrates, 100% cotton and 100% poly(ethylene terephthalate) (elsewhere referred to as polyester) woven fabrics, which are the two more commonly used woven textile fabrics, were selected as the substrates. These fabrics were obtained from Whaley-Bradford Limited, UK. Both the cotton and the polyester fabric had a 1 x 1 plain woven structure and GSM of 80 and 55, respectively.

3. Methods

3.1 Letdown procedure to formulate finished inks

Each of the 12 optimized pigment dispersions was let-down with Binder1 (B1), Binder2 (B2) and Binder3 (B3). The inks were formulated to contain 100% binder solids on the weight of pigment (referred to as %BOWP). For let-down, 10 g of a pigment dispersion was weighed in a 25 mL beaker and the required amount of binder was added drop-wise using a pipette while the mixture was continuously stirred using a magnetic stirrer. The beaker containing the ink was sealed using parafilm to avoid evaporation and mixing was continued for 10 minutes after addition of the required amount of the binder. The ink was then collected in a glass vial that was covered with a PTFE sealed cap to avoid exposure to air and to prevent evaporation of water.

3.2 Sample preparation

For various tests relevant to this study, samples were prepared by depositing the formulated inks onto the paperboard substrate and the selected textile fabrics. Ink deposition was done using a hand coating apparatus from RK PrintCoat Instruments, UK. In the case of textile substrates, primer coated fabrics were also prepared and considered for performance testing of the formulated inks and the commercial inks. To select a binder for primer coating, a separate set of tests was conducted in which each of the three binders that are listed in Table II was

padded onto the fabric substrates using a laboratory padder. Binder3 was found to be better than Binder1 and Binder2 in terms of the washing durability of the coating. Figure 1(a,b) show that actual fabric samples while Figure 1(c,d) show the Binder3 coated fabrics after washing according to BS EN ISO 105-C06 test method. It is evident from Figure 1(c,d) that the primer coating was durable and it also greatly reduced the surface roughness and porosity of both the fabrics. Binder3 primer coating was thus carried out on the cotton and polyester fabrics by padding the fabrics with the binder.

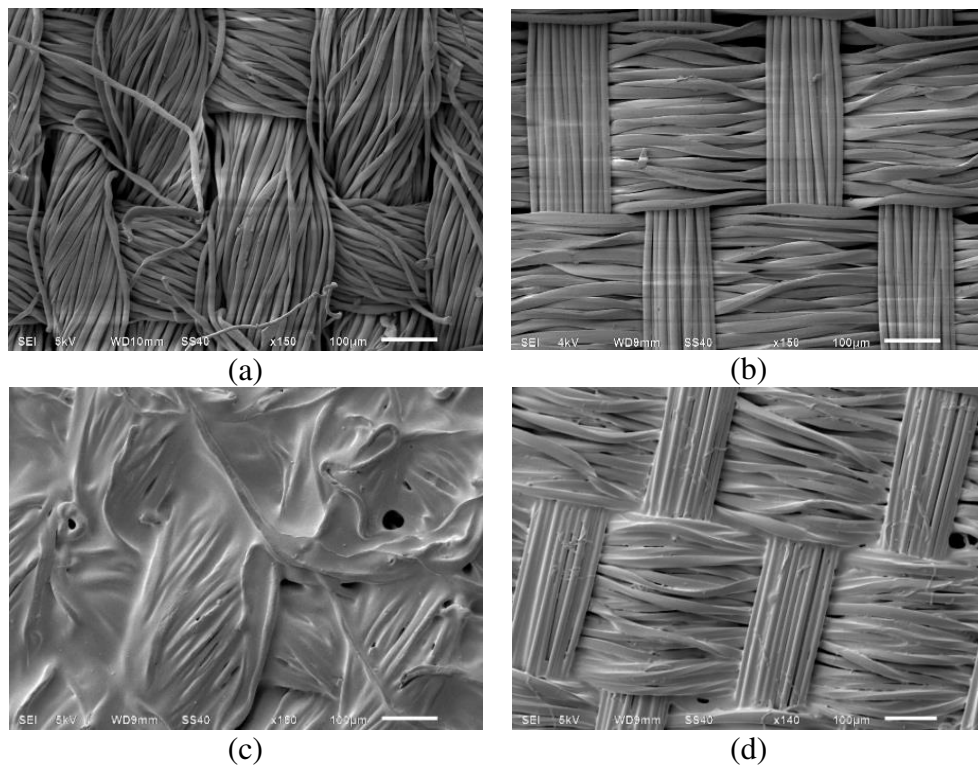


Figure 1: Micrographs of (a) uncoated cotton, (b) uncoated polyester, (c) coated cotton and (d) coated polyester.

3.3 Durability testing of inks

In the research that underpins this paper, the washing and creasing durability of inks that were printed onto textile substrates were evaluated. These two tests are arguably the more important durability tests to be carried out on any product in the domain of e-textiles.

3.3.1 Washing tests

From one of the sets of printed substrates, prepared as described in the preceding section, swatches were cut out for washing tests which were performed according to the BS EN ISO 105-C06:1997 test method. This test method is used to determine the colour fastness of a textile material to domestic washing and commercial laundering. In the context of present study, however, the test was carried out to determine the effects of washing on the surface resistivity of the conductive inks deposited onto textile substrates. The washing tests were carried out on a Mathis Washtester Model WT, which complies with the requirements for a suitable mechanical device, as specified in the standard test method. The AATCC Reference Detergent 1993 (Without Optical Brightener) was used in all the tests.

3.3.2 Creasing tests

Creasing tests were performed according to the ASTM F 2749 – 09 test method, which specifies the procedure, without specifying the precision and bias of the procedure, for the creasing of any part of a membrane switch with conductive circuits. The method defines ‘crease’ as a ridge or groove made by folding and pressing while a ‘crease cycle’ is defined as a 180° crease, followed by straightening of the crease. Five creasing cycles were performed and the surface resistivity of the specimen was recorded between successive cycles.

3.4 Electrical characterization

The surface resistivity of drawdowns of dispersions and the inks was recorded using a Keithley digital multimeter (Model 2100). An electrode, designed to record the surface resistivity of a 30 mm x 30 mm printed area was used. A standard 5 kg weight block was used to press the electrode onto the printed substrate, thus helping to ensure that intimate contact between the electrode and the test surface was achieved.

4. Results and Discussion

4.1 Let down stability of finished inks

As described in Section 3.1, different binders were used to convert the optimized pigment dispersions into finished inks. Addition of a binder can induce instability in a pigment dispersion. In the case of conductive dispersions and inks, such instability could become apparent in the form of significant or complete loss of conductivity due to aggregation of the conductive pigment. Thus, in the present study, the surface resistivity measurements were used as an indirect means of assessing the letdown stability of the pigment dispersions. In the first step, the inks were printed onto paperboard substrate instead of textile fabrics. The use of this substrate facilitated the exclusion of the effects of substrate-dependent variations, such as non-uniform absorption/penetration of ink and a corresponding variation in the thickness of the dried ink layer. K bar 3 was used to prepare drawdowns of each of the finished inks and the surface resistivity was recorded, as described in Section 3.4. The relevant data is provided in Figure 2. It was found that the inks prepared from the dispersions of both the low surface area pigments and the high surface area pigments possessed comparable surface resistivity. In addition, the surface resistivity of finished inks that were prepared by incorporating different binders in a pigment dispersion was also comparable. This could be considered as an indication of the fact that none of the dispersions were destabilized as a result of letdown with the different binders that were considered in this study. This point is particularly significant for the dispersions of high surface area pigments in which large amounts of dispersants, as high as up to 225 % on the weight of pigment, were present.

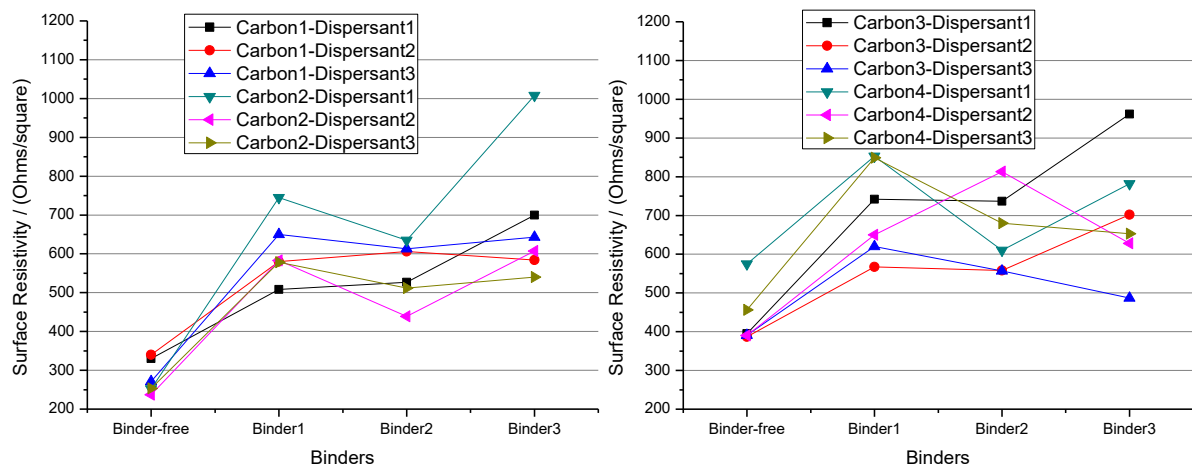


Figure 2: Surface resistivity of inks prepared using different binders.

It was observed in some trial experiments that K bar 9, that gives a wet film deposit of 120 μm , was appropriate for ink deposition on textile substrates. However, when drawdowns were prepared on paperboard substrate using K bar 9, a significant deformation of the paperboard substrate resulted. This made it difficult to obtain a stable measurement of surface resistivity. Thus, the test samples for surface resistivity measurements of multiple layers of ink were produced with three ink layers deposited on top of each other using K-bar 3 (K-bar 3 produces a wet film deposit of 24 μm). The ink layers were air dried at ambient temperature for one hour and the surface resistivity was recorded between successive ink depositions. After deposition of the third ink layer, the printed substrates were oven dried at 60 $^{\circ}\text{C}$ for 30 minutes followed by conditioning at ambient temperature (20-23 $^{\circ}\text{C}$) for one hour before the surface resistivity was recorded. This approach also ensured that potential unwanted effects such as migration of binder to the surface of ink layer during drying were eliminated or minimized. The data thus obtained (Figure 3) provided the electrical characteristics of single layer and multiple layers of the formulated inks.

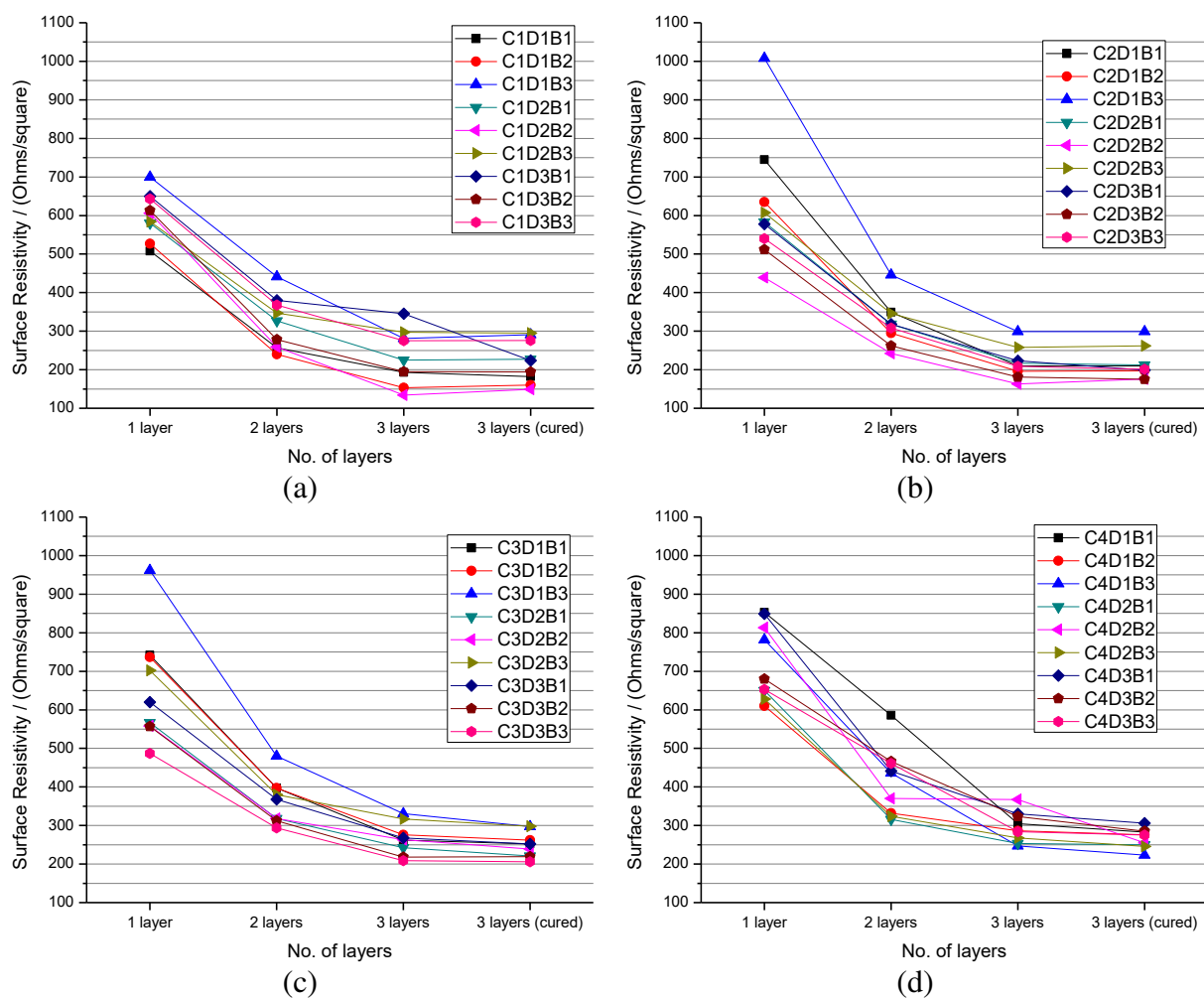


Figure 3: Surface resistivity of single and multiple layers of (a) Carbon1 inks, (b) Carbon2 inks, (c) Carbon3 inks and (d) Carbon4 inks.

4.2 Printing and testing on textile substrates

4.2.1 Washing performance of inks produced using different binders

The presence of hydrophilic entities of the dispersants can result in a significant reduction in the water and chemical resistance of the dry ink film³²⁻³⁴. This effect can be more pronounced if large amount of a dispersant is present in the formulation, as in the case of this study. In order to test the durability of films of inks formulated to contain 100% BOWP of various binders, fabrics were printed and tested. The results of washing tests, tabulated in Table IV and Table V indicate that none of the three binders was significantly inferior to others in terms of furnishing resistance to washing. However, in most cases, the increase in surface resistivity was the least in those inks that were prepared using Binder3. Furthermore, the

surface resistivity of ink films containing Binder3 was generally less (before and after the wash tests) compared to the surface resistivity of ink films containing Binder1 or Binder2.

Table IV: Change in surface resistivity after washing of Carbon1-/Carbon2- containing ink films deposited onto cotton fabric.

Ink composition		Surface resistivity (Ω/\square)		
		Before wash	After wash	% Increase
Carbon1 31 wt%, Dispersant1 15% DOWP	Binder1 100% BOWP	442	536	21.26
	Binder2 100% BOWP	276	365	32.24
	Binder3 100% BOWP	287	311	8.36
Carbon1 31 wt%, Dispersant2 15% DOWP	Binder1 100% BOWP	558	690	23.65
	Binder2 100% BOWP	200	257	28.50
	Binder3 100% BOWP	139	169	21.58
Carbon1 31 wt%, Dispersant3 15% DOWP	Binder1 100% BOWP	725	737	1.65
	Binder2 100% BOWP	225	264	17.33
	Binder3 100% BOWP	99	123	24.24
Carbon2 23 wt%, Dispersant1 17.5% DOWP	Binder1 100% BOWP	220	320	45.45
	Binder2 100% BOWP	155	218	40.64
	Binder3 100% BOWP	127	166	30.71
Carbon2 23 wt%, Dispersant2 17.5% DOWP	Binder1 100% BOWP	270	383	41.85
	Binder2 100% BOWP	144	194	34.72
	Binder3 100% BOWP	140	188	34.28
Carbon2 22 wt%, Dispersant3 17.5% DOWP	Binder1 100% BOWP	307	361	17.59
	Binder2 100% BOWP	166	264	59.04
	Binder3 100% BOWP	151	201	33.11

Table V: Change in surface resistivity after washing of Carbon3-/Carbon4- containing ink films deposited onto cotton fabric.

Ink composition		Surface resistivity (Ω/\square)		
		Before wash	After wash	% Increase
Carbon3 11 wt%, Dispersant1 155% DOWP	Binder1 100% BOWP	568	938	65.14
	Binder2 100% BOWP	441	1050	138.09
	Binder3 100% BOWP	324	573	76.85
Carbon3 11 wt%, Dispersant2 155% DOWP	Binder1 100% BOWP	422	896	112.32
	Binder2 100% BOWP	297	493	65.99

	Binder3 100% BOWP	188	350	86.17
Carbon3 10.5 wt%, Dispersant3 155% DOWP	Binder1 100% BOWP	461	1645	256.83
	Binder2 100% BOWP	216	462	113.88
	Binder3 100% BOWP	112	182	62.50
Carbon4 9.5 wt%, Dispersant1 225% DOWP	Binder1 100% BOWP	1321	2943	122.78
	Binder2 100% BOWP	784	1966	150.76
	Binder3 100% BOWP	376	987	162.50
Carbon4 9.5 wt%, Dispersant2 225% DOWP	Binder1 100% BOWP	647	2955	356.72
	Binder2 100% BOWP	411	1314	219.71
	Binder3 100% BOWP	277	582	110.11
Carbon4 9.3 wt%, Dispersant3 225% DOWP	Binder1 100% BOWP	721	1251	73.51
	Binder2 100% BOWP	241	410	70.12
	Binder3 100% BOWP	206	340	65.05

The results presented in Table IV and Table V also indicate clearly that the increase in surface resistivity was more pronounced in the case of inks containing high surface area pigments. This could be attributed to the considerably large amounts of dispersants present in these formulations. More importantly, it must be noted that in order to maintain a comparable pigment:binder ratio in all of the inks, the amount of binder in a dispersion was calculated on the weight of pigment in the dispersion. Thus, in the case of high surface area pigments, the amount of binder was below the minimum recommended quantities.

4.2.2 Testing of inks on different substrates

It is well-known that substrate surface characteristics can greatly influence the properties and in turn the performance of ink films. Thus, at this stage in the present study, the inks were printed and tested on the uncoated fabric substrates and Binder3-coated fabric substrates that were prepared by following the procedure described in Section 3.2. On the basis of a consideration of the results presented in the preceding section, only the inks containing Binder3 were considered for further testing.

It is evident from the data presented in Table VI and Table VII that in the case of both cotton as well as the polyester fabric substrates, the surface resistivity is generally reduced when an ink is printed onto a binder coated substrate. This is commonly attributed to improved ink film integrity due to a lesser extent of pores and capillaries. Owing to the very low GSM and open structure of the polyester fabric, the drawdowns produced on it were not very uniform and the corresponding surface resistivity values varied within a broad range.

Table VI: Surface resistivity data of the films of inks prepared from the dispersions of Carbon1 and Carbon2 pigments.

Ink composition	Surface resistivity (Ω/\square)			
	Cotton		Polyester	
	Coated	Uncoated	Coated	Uncoated
Carbon1 31 wt%, Dispersant1 15% DOWP, Binder 3 100% BOWP	128	178	130	193
Carbon1 31 wt%, Dispersant2 15% DOWP, Binder 3 100% BOWP	121	101	153	206
Carbon1 31 wt%, Dispersant3 15% DOWP, Binder 3 100% BOWP	83	102	98	137
Carbon2 23 wt%, Dispersant1 17.5% DOWP, Binder 3 100% BOWP	76	149	93	220
Carbon2 23 wt%, Dispersant2 17.5% DOWP, Binder 3 100% BOWP	98	152	91	236
Carbon2 22 wt%, Dispersant3 17.5% DOWP, Binder 3 100% BOWP	101	139	100	252

Table VII: Surface resistivity data of the films of inks prepared from the dispersions of Carbon3 and Carbon4 pigments.

Ink composition	Surface resistivity (Ω/\square)			
	Cotton		Polyester	
	Coated	Uncoated	Coated	Uncoated
Carbon3 11 wt%, Dispersant1 155% DOWP, Binder 3 100% BOWP	216	282	166	274
Carbon3 11 wt%, Dispersant2 155% DOWP, Binder 3 100% BOWP	159	180	133	175
Carbon3 10.5 wt%,	102	127	122	138

Dispersant3 155% DOWP, Binder 3 100% BOWP				
Carbon4 9.5 wt%, Dispersant1 225% DOWP, Binder 3 100% BOWP	187	392	113	407
Carbon4 9.5 wt%, Dispersant2 225% DOWP, Binder 3 100% BOWP	351	301	130	487
Carbon4 9.3 wt%, Dispersant3 225% DOWP, Binder 3 100% BOWP	241	230	114	135

4.2.3 Performance comparison against commercial inks

The washing and creasing performance of the Binder3 containing inks were compared against that of the selected commercial conductive inks. For this purpose, the commercial inks were drawn on the fabric substrates, followed by curing and washing the printed fabrics. From the data provided in Table VIII and Table IX, it is clear that increase in the surface resistivity, when an ink film was deposited on Binder3-coated polyester fabric, was slightly less than that in the surface resistivity of the film of same ink deposited onto uncoated cotton fabric. This difference could probably be due to improved bonding of ink layer onto the binder impregnated substrate.

Prior to washing, the surface resistivity of the films produced from the commercial inks was considerably lower than the surface resistivity of the films produced from the formulated inks. However, the films produced from the commercial inks were less durable, as indicated by a significantly greater increase in the surface resistivity after washing (Table X). Furthermore, the commercial inks were removed from large areas of the fabrics during washing. This showed that the commercial inks tested were not suitable for printing fine lines, a quality that is often required of prints when printing electrical interconnects.

Table VIII: Change in surface resistivity after washing of Carbon1-/Carbon2- containing ink films.

Ink composition	Substrate	Surface resistivity (Ω/\square)		
		Before wash	After wash	% Increase
Carbon1 31 wt%, Dispersant1 15% DOWP, Binder 3 100% BOWP.	Uncoated Cotton	287	311	8.36
	Coated Polyester	112	118	5.36
Carbon1 31 wt%, Dispersant2 15% DOWP, Binder3 100% BOWP	Uncoated Cotton	139	169	21.58
	Coated Polyester	101	99	-1.49
Carbon1 31 wt%, Dispersant3 15% DOWP, Binder3 100% BOWP	Uncoated Cotton	99	123	24.24
	Coated Polyester	129	157	21.58
Carbon2 23 wt%, Dispersant1 17.5% DOWP, Binder3 100% BOWP	Uncoated Cotton	127	166	30.71
	Coated Polyester	139	178	27.87
Carbon2 23 wt%, Dispersant2 17.5% DOWP, Binder3 100% BOWP	Uncoated Cotton	140	188	34.28
	Coated Polyester	153	183	19.63
Carbon2 22 wt%, Dispersant3 17.5% DOWP, Binder3 100% BOWP	Uncoated Cotton	151	201	33.11
	Coated Polyester	223	275	23.15

Table IX: Change in surface resistivity after washing of Carbon3-/Carbon4- containing ink films.

Ink composition	Substrate	Surface resistivity (Ω/\square)		
		Before wash	After wash	% Increase
Carbon3 11 wt%, Dispersant1 155% DOWP, Binder3 100% BOWP	Uncoated Cotton	324	573	76.85
	Coated Polyester	115	129	12.52
Carbon3 11 wt%, Dispersant2 155% DOWP, Binder3 100% BOWP	Uncoated Cotton	188	350	86.17
	Coated Polyester	128	145	12.44
Carbon3 10.5 wt%, Dispersant3 155% DOWP, Binder3 100% BOWP	Uncoated Cotton	112	182	62.50
	Coated Polyester	69	77	11.49
Carbon4 9.5 wt%, Dispersant1 225% DOWP, Binder3 100% BOWP	Uncoated Cotton	376	987	162.50
	Coated Polyester	739	1686	128.14
Carbon4 9.5 wt%, Dispersant2 225% DOWP, Binder3 100% BOWP	Uncoated Cotton	277	582	110.11
	Coated Polyester	116	238	105.17
Carbon4 9.3 wt%, Dispersant3 225% DOWP, Binder3 100% BOWP	Uncoated Cotton	206	340	65.05
	Coated Polyester	102	132	29.41

Table X: Washing tests results of commercial inks

Ink name	Substrate	Surface resistivity (Ω/\square)		
		Before wash	After wash	% Increase
Gwent C2030519P4	Uncoated Cotton	20.83	81.9	293.18
Gwent C2030519P4	Coated Polyester	19.14	77.4	304.39
Peters SD 2843 HAL	Uncoated Cotton	59.96	NR	-
Peters SD 2843 HAL	Coated Polyester	42.5	307	622.35

NR refers to 'no reading', i.e., the surface resistivity was higher than 100 M Ω

The creasing resistance of only the ink films produced on Binder3-coated polyester fabric was tested according to the procedure outlined in Section 3.3.2. Durability of the ink films to withstand up to five creasing cycles was tested and the surface resistivity was recorded after each cycle. The results are tabulated in Table XI. The results of creasing tests indicate that the overall increase in the surface resistivity of the films produced from the formulated inks was considerably lower compared to the increase in surface resistivity of films produced from the commercial inks. Furthermore, it was also noticed that the increase in surface resistivity was more pronounced in case of the inks that were prepared from the dispersions of high surface area pigments. As in the case of washing performance, this can be attributed to the very low binder solids content in the inks formulated from the dispersions of high surface area pigments.

Table XI: Surface resistivity of inks recorded during crease testing of ink films.

Ink composition	Surface resistivity (Ω/\square)						Overall Increase (%)
	Number of crease cycles						
	0	1	2	3	4	5	
Carbon1 31 wt%, Dispersant1 15% DOWP, Binder3 100% BOWP	112	121	125	128	127	128	14.29
Carbon1 31 wt%, Dispersant2 15% DOWP, Binder3 100% BOWP	143	165	170	174	175	181	26.57
Carbon1 31 wt%, Dispersant3 15% DOWP, Binder3 100% BOWP	90	96	99	103	102	104	15.56
Carbon2 23 wt%, Dispersant1 17.5% DOWP, Binder3 100% BOWP	102	106	111	108	113	113	10.78
Carbon2 23 wt%, Dispersant2 17.5% DOWP, Binder3 100% BOWP	92	101	103	103	104	106	15.22
Carbon2 22 wt%, Dispersant3 17.5% DOWP, Binder3 100% BOWP	86	87	87	90	91	91	5.81

Binder3 100% BOWP								
Carbon3 11 wt%, Dispersant1 155% DOWP, Binder3 100% BOWP	134	146	155	157	162	162	20.90	
Carbon3 11 wt%, Dispersant2 155% DOWP, Binder3 100% BOWP	120	139	149	151	156	158	31.67	
Carbon3 10.5 wt%, Dispersant3 155% DOWP, Binder3 100% BOWP	108	113	123	125	134	141	30.56	
Carbon4 9.5 wt%, Dispersant1 225% DOWP, Binder3 100% BOWP	248	289	314	338	364	386	55.65	
Carbon4 9.5 wt%, Dispersant2 225% DOWP, Binder3 100% BOWP	120	147	159	186	207	226	88.33	
Carbon4 9.3 wt%, Dispersant3 225% DOWP, Binder3 100% BOWP	91	123	163	230	408	538	491.21	
Peters SD 2843 HAL carbon ink	28	63	98	157	196	411	311.00	
Gwent C2030519P4 carbon ink	20	71	170	302	432	550	450.00	

4.3 Resistivity of multiple ink layers

Screen printing is arguably the most common technique of textile printing. Since screen printing can be employed to deposit relatively thick layers of inks onto textile substrates, it was attempted to find the surface resistivity of relatively very thick deposits of the formulated inks. For this purpose, Binder3-coated polyester fabric was selected as a substrate on the basis of the results of aforementioned studies, which indicated that this substrate provided a very smooth surface for printing. K bar 9 was used and two layers of an ink to be tested were deposited on top of each other with intermediate air drying for 1 hour between successive depositions. This was followed by air drying for 1 hour and then curing the ink deposit at 110 – 120 °C for 20 minutes. The cured specimens were conditioned at ambient temperature (22 – 25 °C) for 24 hours before measuring the surface resistivity.

In Table XII, the pigment loading in the finished inks that were prepared in this study is provided along with the surface resistivity data for single and double ink layers on the Binder3 coated polyester fabric. The pigment loading in the finished inks was well below 20 wt% and 10 wt% in the case of low surface area pigments and the high surface area pigments,

respectively. Despite the very low pigment loading, most of the formulated inks possessed surface resistivity of less than 100 Ω/\square . The deposition of two inks layers resulted in a considerable decrease in the surface resistivity. This shows that for certain applications allowing thick ink deposits, for instance printing of resistive heating elements, the electrical characteristics of the formulated inks can be adjusted in a broad range with relative ease.

Table XII: Surface resistivity of single and double ink layers on Binder3-coated polyester fabric.

Ink composition	Final pigment loading (wt %)	Surface resistivity (Ω/\square)	
		1 layer	2 layers
Carbon1 31 wt%, Dispersant1 15% DOWP, Binder3 100% BOWP	17.46	89.4	50.72
Carbon1 31 wt%, Dispersant2 15% DOWP, Binder3 100% BOWP	17.46	115	68.65
Carbon1 31 wt%, Dispersant3 15% DOWP, Binder3 100% BOWP	17.46	80	43.68
Carbon2 23 wt%, Dispersant1 17.5% DOWP, Binder3 100% BOWP	14.60	111	53.74
Carbon2 23 wt%, Dispersant2 17.5% DOWP, Binder3 100% BOWP	14.60	106	49.96
Carbon2 22 wt%, Dispersant3 17.5% DOWP, Binder3 100% BOWP	14.19	87.4	46
Carbon3 11 wt%, Dispersant1 155% DOWP, Binder3 100% BOWP	8.63	162	103
Carbon3 11 wt%, Dispersant2 155% DOWP, Binder3 100% BOWP	8.63	140	92
Carbon3 10.5 wt%, Dispersant3 155% DOWP, Binder3 100% BOWP	8.32	152	85
Carbon4 9.5 wt%, Dispersant1 225% DOWP, Binder3 100% BOWP	7.68	530	435
Carbon4 9.5 wt%, Dispersant2 225% DOWP, Binder3 100% BOWP	7.68	180	149
Carbon4 9.3 wt%, Dispersant3 225% DOWP, Binder3 100% BOWP	7.54	157	101

5. Conclusion

Waterborne dispersions of electrically conductive grades of carbon black pigment were converted into finished inks by incorporating various polymeric binders. The electrical properties of the prepared inks clearly indicate that no considerable instability occurred in any

of the dispersions after letdown with various binders. In terms of washing and creasing, the durability of inks that were produced from the dispersions of high surface pigments was inferior but this could be primarily attributed to the considerably low binder quantity in these inks. The conductive ink formulation procedure devised in the present study can be used to produce highly conductive inks from non-printing ink grades of carbon black pigment. The results of this study clearly show that such non-printing ink grades furnish high electrical conductivity at incredibly low pigment loadings. In addition, the electrical properties can be varied within a broad range of values with relative ease. This makes such inks suitable for numerous potential applications in the domain of e-textiles. However, the mechanical properties required of textile printing inks can be further improved by optimizing the binder content.

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7. References

1. Van LL and Hertleer C. Smart clothing: a new life. *International Journal of Clothing Science and Technology* 2004; 16: 63-72.
2. Marculescu D. E-textiles: Toward computational clothing. *Pervasive Computing, IEEE* 2003; 2: 89-95.
3. Troster G, Kirstein T and Lukowicz P. Wearable computing: Packaging in textiles and clothes. *14th European Microelectronics and Packaging Conference & Exhibition*. Friedrichshafen, Germany: Wearable Computing Lab, ETH Zurich, 2003.
4. Mattana G, Cosseddu P, Fraboni B, et al. Organic electronics on natural cotton fibres. *Org Electron* 2011; 12: 2033-2039. DOI: <http://dx.doi.org/10.1016/j.orgel.2011.09.001>.
5. Paradiso R and Rossi DD. Advances in textile technologies for unobtrusive monitoring of vital parameters and movements. In: *Engineering in Medicine and Biology Society, 2006 EMBS '06 28th Annual International Conference of the IEEE* 30/08 - 03/09/2006 2006, pp.392-395.
6. Marculescu D, Marculescu R, Zamora NH, et al. Electronic textiles: A platform for pervasive computing. *Proceedings of the IEEE* 2003; 91: 1995-2018. DOI: 10.1109/jproc.2003.819612.
7. Dhawan A, Seyam AM, Ghosh TK, et al. Woven Fabric-Based Electrical Circuits: Part I: Evaluating Interconnect Methods. *Textile Research Journal* 2004; 74: 913-919. DOI: 10.1177/004051750407401011.

8. Locher I and Troster G. Enabling Technologies for Electrical Circuits on a Woven Monofilament Hybrid Fabric. *Textile Research Journal* 2008; 78: 583 - 594. DOI: 10.1177/0040517507081314.
9. Gimpel S, Mohring U, Muller H, et al. Textile-Based Electronic Substrate Technology. *Journal of Industrial Textiles* 2004; 33: 179-189. DOI: 10.1177/1528083704039828.
10. Post RE, Orth M, Cooper E, et al. *Electrically active textiles and articles made therefrom*. Patent US 6210771, United States, 2001.
11. Bao Z, Feng Y, Dodabalapur A, et al. High-Performance Plastic Transistors Fabricated by Printing Techniques. *Chemistry of Materials* 1997; 9: 1299-1301. doi: 10.1021/cm9701163. DOI: 10.1021/cm9701163.
12. Gray C, Wang J, Duthaler G, et al. Screen printed organic thin film transistors (OTFTs) on a flexible substrate. In: *Organic Field Effect Transistors* San Diego, CA, USA, 2001, pp.89-94. SPIE.
13. Tymecki L, Glab S and Koncki R. Miniaturized, Planar Ion-selective Electrodes Fabricated by Means of Thick-film Technology. *Sensors* 2006; 6: 390-396.
14. Michels JJ, de Winter S and Symonds LHG. Process optimization of gravure printed light-emitting polymer layers by a neural network approach. *Org Electron* 2009; 10: 1495-1504. Article. DOI: 10.1016/j.orgel.2009.08.015.
15. Ghosh TK, Dhawan A and Muth JF. Formation of electrical circuits in textile structures. In: Matilla H (ed) *Intelligent Textiles and Clothing*. Cambridge: Woodhead Publishing, 2006.
16. Merilampi S, Laine-Ma T and Ruuskanen P. The characterization of electrically conductive silver ink patterns on flexible substrates. *Microelectronics Reliability* 2009; 49: 782-790. DOI: 10.1016/j.microrel.2009.04.004.
17. Locher I and Tröster G. Screen-printed Textile Transmission Lines. *Textile Research Journal* 2007; 77: 837-842. DOI: 10.1177/0040517507080679.
18. Kazani I, Hertleer C, De Mey G, et al. Electrical Conductive Textiles Obtained by Screen Printing. *Fibres & Textiles in Eastern Europe* 2012; 20: 57-63.
19. Inoue M, Tada Y, Muta H, et al. Development of highly conductive inks for smart textiles. In: *14th International Conference on Electronic Materials and Packaging (EMAP)* 13-16 Dec. 2012 2012, pp.1-4.
20. Park SM, Cho KS and Chung KH. *Flexible printed conductive fabric and method of fabricating the same*. Patent WO 2008/060101, 2007.
21. Karaguzel B, Merritt CR, Kang T, et al. Flexible, durable printed electrical circuits. *Journal of the Textile Institute* 2009; 100: 1 - 9.
22. Bidoki SM, Lewis DM, Clark M, et al. Ink-jet fabrication of electronic components. *Journal of Micromechanics and Microengineering* 2007; 17: 967-974. DOI: 10.1088/0960-1317/17/5/017.
23. Jang BZ and Zhamu A. *Nano graphene platelet-based conductive inks*. Patent US 20100000441 A1, USA, 2008.
24. Hu L, Pasta M, La Mantia F, et al. Stretchable, Porous, and Conductive Energy Textiles. *Nano Letters* 2010; 10: 708-714. doi: 10.1021/nl903949m. DOI: 10.1021/nl903949m.
25. Krucińska I, Skrzetuska E and Urbaniak-Domagala W. The use of carbon nanotubes in textile printing. *Journal of Applied Polymer Science* 2011; 121: 483-490. DOI: 10.1002/app.33598.
26. Ma R, Suh D, Kim J, et al. A drastic reduction in silver concentration of metallic ink by the use of single-walled carbon nanotubes decorated with silver nanoparticles. *Journal of Materials Chemistry* 2011; 21: 7070-7073. 10.1039/C1JM10850F. DOI: 10.1039/c1jm10850f.

27. Kaynak A and Håkansson E. Generating heat from conducting polypyrrole-coated PET fabrics. *Advances in Polymer Technology* 2005; 24: 194-207. DOI: doi:10.1002/adv.20040.
28. Gregory RV, Kimbrell WC and Kuhn HH. Electrically Conductive Non-Metallic Textile Coatings. *Journal of Industrial Textiles* 1991; 20: 167-175. DOI: 10.1177/152808379102000304.
29. Zhao Y-P, Cai Z-S and Fu X-L. Effect of polymeric additives on the formation and properties of polyaniline films on polyester fabrics. *Journal of the Textile Institute* 2011; 103: 724-732. DOI: 10.1080/00405000.2011.603203.
30. Ali M, Lin L and Cartridge D. High electrical conductivity waterborne dispersions of carbon black pigment. *Progress in Organic Coatings* 2019; 129: 199-208. DOI: <https://doi.org/10.1016/j.porgcoat.2018.12.010>.
31. Ali M and Lin L. Optimisation and analysis of bead milling process for preparation of highly viscous, binder-free dispersions of carbon black pigment. *Progress in Organic Coatings* 2018; 119: 1-7. DOI: <https://doi.org/10.1016/j.porgcoat.2018.02.007>.
32. Hobisch G and Tsang M. New Waterborne Pigment Grinding Resin for High-Performance Coatings. *Paint and Coatings Industry Magazine*. 2009.
33. Baez E, Quazi N, Ivanov I, et al. Stability study of nanopigment dispersions. *Advanced Powder Technology* 2009; 20: 267-272. DOI: <https://doi.org/10.1016/j.appt.2009.02.005>.
34. Ridaoui H, Jada A, Vidal L, et al. Effect of cationic surfactant and block copolymer on carbon black particle surface charge and size. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 2006; 278: 149-159. DOI: <https://doi.org/10.1016/j.colsurfa.2005.12.013>.