The development of devices on textiles such as sensors[1], photodetectors[2], transistors[3], electro-luminescent devices[4], supercapacitors[5] and solar cells[6] is attracting great interest and has led to the emergence of the field of smart textiles. Smart textiles can find an enormous range of applications in several fields, including healthcare, military and fashion[7]. Since the concept of textiles is much wider than clothes and garments, the applications can extend to aviation, automotive and transport, construction, geo-textiles and packaging. While most commercial applications of smart textiles rely on conventional hardware simply mounted onto textiles, the integration of specific functionalities directly on textile fibres promises to revolutionize the field of wearable electronics. With the recent advances in nanotechnology and materials engineering, different functionalities can now be incorporated into textile fibres, such as antibacterial properties, static elimination, and electric conductivity[8].

Conducting fibres are an important component of any e-textile, not only because they can be used as lightweight wiring for simple textile-based electronic components, but also because they can provide a platform for building electronic devices directly on textile fibres. For instance, such fibres can be used as gate electrodes for field-effect transistors, or bottom electrodes for light-emitting diodes and photovoltaic devices. Conductive textile fibres are currently used as a stronger and more flexible weight-saving material in the aviation sector, where the aircraft weight and fuel consumption are reduced by replacing metal wiring with electrically conductive cotton fibres like ARACON®. The most common approach to produce conductive fibres consists in mixing an insulating polymeric matrix with a conductive component, which can be a conducting polymer such as PEDOT:PSS[9], silver nanowires[10], nanocarbon fillers[11], or their hybrids[12–15]. These composite fibres are usually produced by wet-spinning of the polymer with a suspension of the active conducting material, or via electrospinning[16], techniques that require a large consumption of expensive materials and that can cause the loss of flexibility and transparency. A different approach relies on polymer-free fabrication of conducting fibres, but the methods include the use of strong acids and coagulants[17], which greatly limits the potential for scaling up and commercialization. Another strategy is to impregnate fibres with conductive materials after they have been manufactured. This

method has been used for fibres, yarns and fabrics that are highly porous with a complex structure of microfibrils, such as cellulosic fibres. These inks and dyes make use of several types of conductive materials, such as aluminium[18], carbon nanotubes[19], and graphene[20], and have been in the base of demonstrations of wearable and stretchable electronics, including integration onto surfaces of live plants and insects[21, 22]. One of the limitations of this methodology is that such multifilament fibres and fabrics, compared to fibres coated prior or during manufacturing, tend to lose the conductive filling more easily if not completely encapsulated, and can pose end-of-life environmental concerns.

A more practicable emerging strategy to prepare conductive textile fibres is to coat insulating fibres with conductive atomically thin two dimensional layers such as graphene[23]. Graphene, a monoatomic carbon layer, is the strongest known material, the best electrical and thermal conductor which is also mechanically flexible and transparent[24]. Thus, it represents a radical alternative to conventional technologies as it can bend, stretch, compress, twist and deform into complex shapes while maintaining the same levels of performance and reliability[25]. There are already several examples of graphene-based textiles with different functionalities and for different applications[26]. The coating we propose is performed by electrostatic adhesion of graphene at the surface of monofilament fibres and does not involve impregnating an agglomerate of fibres. The adhesion of the graphene coating to the textile fibres is strong and durable, and a straightforward passivation can be achieved by encapsulation with an insulating polymeric layer. This method was developed for tape-shaped polypropylene (PP) and bio-based polylactic acid (PLA) fibres, two polymers with widespread use in the textile industry[27]. This approach depends on the size of the graphene sample, usually in the centimetre range, and although this might not be suitable for electronic wiring, it is appropriate to build electronic devices directly on textile fibres.

In this work, we demonstrate that graphene can be transferred to a large variety of thermoplastic monofilament textile fibres of different types and shapes. To further advance the development of this technique, it is important to understand the various factors that can influence the conductivity achieved by coating textile fibres with graphene. Surface topography and chemical nature seem to be determinant in the conductivity achieved. On the other hand, cracks and tears in the graphene coating will result in a decrease in conductivity, and therefore it

is important to establish their origin. Thus, our present study aims at: (1) establishing how the above-mentioned factors actually influence the quality of the graphene coverage and how it correlates with the resulting conductivity; and (2) explore the suitability of our coating method for different materials, sizes and shapes.Raman spectra of fibres of the three materials (PP2, PE and PLA2), before and after the graphene transfer, are shown in Fig. 1e. For all graphene-coated fibres it was possible to identify the graphene G band at 1585 cm^{-1} . The

2D peak was clearly visible for graphene-coated PP1 and PLA2, at 2685 cm^{-1}

, as well as a small D peak for PE and PLA2. These values match those found for G and 2D bands of the same type of graphene transferred to SiO_2 (see Supplementary Fig. S2; more extended Raman spectra of the graphene-coated fibres is shown in Supplementary Fig. S3, along with details of the G and 2D peaks and corresponding integrations for PP2, PE and PLA2).

To study the factors that lead to the observed differences in sheet resistance, it is important to understand the influence of the topography of the fibres on the effectiveness of the graphene coating. A non-contact optical method was used to determine the macroscopic surface parameters of the untreated fibres. The images obtained are shown in Supplementary Figs S4, S5 and S6, and the parameters are listed in Supplementary Table S1. Figure 2 shows the Atomic Force Microscopy (AFM) amplitude and topography for 5×5μm images of the PP fibres before coating and corresponding height profiles taken at the highlighted lines parallel to the extrusion axis. Compared to PP1, PP2 fibres show considerable differences in terms of AFM topography. PP2 does indeed have a smoother height profile (Fig. 2a, right) with less pronounced height differences than PP1 (Fig. 2a, left). The UVO treatment created a fine roughness throughout the whole surface, which seems to create more points where the graphene sheet can effectively adhere to the surface of the fibre (Fig. 2a, middle). The same conclusions are valid for larger and smaller AFM scanning areas (30×30 μm and 1×1 μm images, see Supplementary Fig. S7). Although in terms of overall thickness and surface features, PP1 and PP2 are very similar (see Supplementary Table S1), in a smaller scale AFM shows that PP2 has areas with less pronounced features than PP1, which is also in accordance with the smaller Kurtosis value in PP2. A similar study was performed for the PLA-based fibres PLA1 and

PLA2 (Fig. 2b and Supplementary Fig. S8), showing that the changes in polymer source grade do not have a substantial impact on the surface morphology in the AFM

scale. However, the difference in roughness is much more significant macroscopically, with PLA2 is rougher than PLA1, showing visible ridges perpendicular to the extrusion lines (Supplementary Fig. S5). On the other hand, we found that the UVO treatment does change the surface of PLA-fibres significantly (Fig. 2b, middle). All three monofilament tape-shaped samples, PP2, PLA2 and PE were also subjected to UVO treatment. Both PLA1 and PLA2 fibres often appeared to be damaged after UVO-treatment, particularly towards the edges, where propagating cracks and microfibrils tearing from the sample were clearly visible to the naked eye (see Supplementary Fig. S8, top middle, where a loose microfibril is clearly noticeable). The damage induced by the UVO treatment is even more severe in PLA2 fibres. The protuberances that are observed on the fibres as a consequence of the UVO treatment are probably a sign of that degradation, and biodegradable polymers like those based on PLA are prone to be damaged even more rapidly. These protuberances can be thermally caused or appear due to the chemical reactions of oxygen radicals at the surface of the polymer. Attempted coating of these UVO-treated PLA2 fibres with graphene was unsuccessful, as all the samples remained insulating after the graphene transfer. The immersion in warm acetone during the PMMA ((poly(methyl methacrylate)) cleaning step seemed to damage the fibres even further, causing them to curl and shrink. It should be noted that the UVO treatment was performed under the same conditions for all fibres, and even though the bio-based PLA1 fibres also showed some signs of quicker degradation when subjected to the UVO treatment, it was still possible to achieve considerably low sheet resistance. The fact that PLA2 fibres are slightly smaller than PLA1, 1.0 mm wide and 0.07 mm thick, compared to 1.2 mm wide and 0.1 mm thick, may also account for the fact that the formers are more fragile and easily degradable.