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Electrical conductivity of the graphene nanoplatelets coated natural and synthetic fibres using electrophoretic deposition technique

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ABSTRACT

Herein, electrically conductive natural and synthetic yarns through electrophoretic deposition (EPD) technique were fabricated. A parametric study on the conductivity enhancement of the yarns is carried out by Taguchi method. Using this method, the desirable conditions are determined by studying the effects of important parameters on the electrical conductivity of the yarns in the EPD coating process. Based on the L18 design of experiments table, the preferred combination of factors to obtain the highest electrical conductivity of the yarns is found by Taguchi analysis. In addition, the Pareto ANOVA analysis is conducted to identify the major contributing factors on the electrical conductivity of the yarns. Characterisation techniques, such as scanning electron microscopy (SEM), Fourier transformed infrared spectroscopy (FTIR) in attenuated total reflectance (ATR) mode, and thermogravimetric analysis (TGA) are utilised for better understanding the microstructure and physical properties. When powered by only 3 V, the maximum temperature of a Joule heated conductive sample based on natural fibre yarns reached 102°C in less than 25 s.

1. Introduction

Over the past few decades, the use of continuous fibres, such as carbon fibre (CF), glass fibre (GF), and natural fibre (NF) as reinforcing materials for polymeric composites have delivered a wide range of properties [1,2]. Their lightweight, high mechanical properties and resistance to harsh environments have attracted them as outstanding materials in a number of applications, including aerospace, automotive, marine, buildings, wind turbine blades, and sports goods, as alternatives to metal-based components [1,2].

The NF based composites have seen increased popularity in the past few years with a dramatic increase in the composite market due to their environment friendly attributes [3]. Considering the fact that the disposal and recycling process of synthetic fibres such as GF and CF reinforced polymer composites could result in environmental concerns, NF could offer solutions for these issues, thanks to their...
recyclability, biodegradability and renewability [3–10]. In addition, they are abundant and cost-effective reinforcing materials with attractive properties, such as low density, high specific strength to stiffness ratio, high toughness, low energy utilisation during the fabrication process, low abrasiveness, and neutrality to CO$_2$ [3–10]. Consequently, researchers have stepped forward to manufacturing eco-friendly NFs based composites as alternatives to those fabricated with synthetic fibres [3–10]. NF based composites have been used in diverse applications, such as packaging, furniture, automotive industries, disposable accessories, building, insulation materials, and fire retardant materials [3–10]. However, their electrical insulating nature limits their applications in energy storage materials, batteries, sensors, heating elements, electromagnetic interference (EMI) shielding materials, and actuators, where high electrical conductivity is necessary. With recent advances in nanomaterials, many researchers have investigated the possibility of using conductive micro/nanomaterials or polymer coatings on NFs to enhance their electrical conductivity. For instance, flax fibres spray coated with carbon nanotubes (CNTs) reinforced epoxy composites were utilised as quantum resistive sensors for structural health monitoring applications [11]. In another study, novel and cost-effective conductive cotton/carbon black composites were fabricated utilising the knife-over-roll technique to apply structured carbon black. The lowest resistivity of the composites was reported to be 60 ± 5.4 Ohm/cm$^2$ and their EMI shielding properties were characterised [12]. Dwivedi et al. fabricated CNT dip-coated sisal fibres within epoxy composites and investigated their AC electrical conductivity with regard to the length of composites and dielectric constant [13]. Qi et al. developed composite fibres composed of CNT and cellulose with volume resistivity in the range of 230 to 1 Ohm.cm for 2 to 8 wt.% of CNT loading and then evaluated their humidity and temperature sensing ability [14]. Zhang et al. coated functionalised CNTs on flax fibres through soaking or spray-drying processes. Afterwards, they characterised mechanical properties of composites made by these fibres and an epoxy polymer [15].

Most of these multifunctional yarns or fabrics were mainly fabricated by dip coating of CNT as a highly conductive nano-material. There exists a lack of studies related to the use of graphene derivatives as the promising electrically conductive nanomaterials (with electrical conductivity of up to 6000 S/cm) to coat on NFs for the fabrication of multifunctional materials [16]. In addition, there have been limited studies in the literature regarding the use of other coating techniques to make natural or synthetic fibres electrically conductive. In particular, one could clearly notice the research gap on a parametric study on the enhancement of electrical conductivity of the natural and synthetic yarns by electrophoretic deposition (EPD) coating technique in the literature. The EPD technique offers several advantages including cost-effectiveness, simplicity of the setup, scalability of the size, controllable deposition rate and thickness of deposits, and possibility of using graphene based dispersions; nevertheless, the need for electrically conductive electrodes and the possibility of the creation of side electrochemical reactions are considered as its limitations [17]. A number of parameters are involved in the EPD technique that can effectively alter the coating quality and thus the electrical conductivity of natural fibre yarns, such as applied voltage between the electrodes, deposition time, concentration and pH of the dispersion, and functionalisation of particles. Understanding the effects of all of these parameters on the electrical conductivity of the coated yarns would be arduous and demand a great numbers of
experiments. Design-of-Experiment (DoE) is a statistical method that allows researchers to evaluate the individual and interactive effects of manufacturing factors on the performance of the processes or products [18]. In contrast to a full factorial method, the Taguchi method as an effective and powerful DoE has been widely exploited to assist experimentalists in optimising the number of experiments and decrease the time and cost [18].

Various graphene derivatives such as graphene oxide (GO), graphene nanoplatelets (GNPs), and reduced GO (rGO) have been widely utilised to be dispersed in various solvents such as water, N-methyl-2-pyrrolidone (NMP), and N,N-dimethylformamide (DMF), and other solvents through chemical modifications or by additives [17]. Surfactants are considered as one of the most effective additives that are used for dispersing carbon particles in a solvent like water. The use of sodium dodecyl benzenesulfonate (SDBS), sodium dodecyl sulfate (SDS), and 4-(1,1,3,3-tetramethylbutyl) phenyl-polyethylene glycol (Triton X-100) were found to be the most effective surfactants for dispersing graphene particles in water and their electrical conductivity [19,20]. Dispersion of graphene derivatives with SDBS in water resulted in a comparably high electrical conductivity relative to the other two surfactants [19].

Here, we report a simple, rapid, environment friendly, and cost-effective coating process using the EPD technique to fabricate conductive natural and synthetic yarns by SDBS functionalised-graphene nanoplatelets (f-GNPs) dispersed in de-ionised (DI) water. We chose flax and ultra-high molecular weight polyethylene (UHMWPE) as natural and synthetic yarns, as these materials possess adequate mechanical properties, low density, biocompatibility, and flexibility [4–7,21]. The important parameters in the EPD coating process included the type of yarn, type of electrode, distance between electrodes, applied voltage, amount of surfactant in the dispersion and deposition time. The EPD coating setup was changed for each set of experiments in accordance with the Taguchi L₁₈ table. The coated yarns were characterised to understand their electrical and thermal behaviour. The electrothermal behaviour of a conductive flax yarn was evaluated in the range of 1 to 3 V. The results suggest the capability of utilising conductive natural fibre yarns as electroresistive heating wires.

2. Experimental details

2.1. Materials and samples preparation

In this study, flax yarns, as a natural material, in the forms of bleached (Figure S1(a)) and non-bleached (Figure S1 (b)) were supplied from Jayshree Inc., India with the approximate average diameter of 400 microns. As a comparison, UHMWPE braids were provided by Spectra Extreme Braids, Japan with the approximate average diameter of 450 µm (Figure S1(c)). GNPs with a size of 5 µm in width and 5 nm in thickness were purchased from EMFUTUR, Spain. To disperse hydrophobic GNPs in DI water, an anionic surfactant SDBS from Sigma Aldrich was utilised. Two different pairs of electrodes made of either carbon sheets or stainless steel were employed during the EPD process. The thickness, height, and width of the electrodes were 1, 50, and 100 mm, respectively.

GNPs were dispersed in DI water following the process described: first, the desired amount of GNPs (3g) with the addition of specified amount of SDBS (1, 1.5, and 2 g) were added to DI water (1l) under bath sonication (60 Hz) for 30 min. Next, the dispersions were stirred for four hours at 60 ºC. Finally, they were bath sonicated for 2 hours to obtain more homogenous dispersions (Figure 1).
A schematic view of the EPD process is presented in Figure S2. The goal is to deposit the conductive carbon materials on the surface of the natural and synthetic yarns. The pristine yarns were attached on the surface of an electrically conductive electrode. Here, either a pair of carbon sheets or stainless steel was employed. The pH of the dispersions before the EPD process was measured to be in the range of 11.2 to 11.8. Considering the anionic nature of SDBS in DI water, the yarns were attached to one side of anode (positively charged) (Figure S2) [17,19]. By generating an electric field between the two electrodes, we observed that the negatively charged well-suspended f-GNPs particles in DI water migrated towards the anode. Hence, the nanoparticles would be deposited on the surface of the yarns attached to the anode. After half of the coating time, the yarns were reversed and then the coating process was resumed under the same conditions. This procedure resulted in a more homogeneous coating of the particles on the surfaces of the yarns [17]. Finally, the samples were dried in an oven at 60 ºC for 12 h.

Following the Taguchi L$_{18}$ table, the magnitude of applied voltage, distance between electrodes, time of deposition, the amount of SDBS, the type of electrode, and the type of yarns were altered to fabricate samples for each set. The readers are referred to Section 2.3 for detailed discussion on Taguchi analysis.

### 2.2. Testing methods

The morphology of f-GNPs coatings was investigated using a scanning electron microscope (SEM) (FEI (Philips) XL30 S-FEG). Samples were platinum sputtered prior to SEM observations. Fourier transform infrared spectroscopy (FTIR) measurements in attenuated total reflectance (ATR) mode were conducted by a Nicolet FTIR iS 50 instrument. The measurements were performed between 600 and 3800 cm$^{-1}$ wavenumbers. Thermogravimetric analysis (TGA) was conducted with a TA instrument (Q5000 model) in a nitrogen atmosphere with a temperature increase rate of 10°C min$^{-1}$ up to 800°C.

The electrical resistance of samples was measured by a four-point probe setup utilizing a digital multimeter (Keithley DMM 2100) and a current source (Keithley 220) in accordance with the ASTM-D4496 standard (Figure S3). The gap between the probes was 18 mm. The specific electrical conductivity of the coated yarns with regard to the average value of diameter for each sample was calculated by $\sigma = (4L/\pi d^2R)$ as suggested in the previous studies [22]. In this equation, $\sigma$ denotes the electrical conductivity, $L$ is the distance between the electrodes, $R$ represents the resistance of the samples, and $d$ is the diameter of the yarns.
the average diameter of the yarns. A high-resolution microscope at six different locations quantified the diameter of each yarn and the average value of these measurements was used as the representative diameter of each yarn. Five samples for each set of experiments with regard to the Taguchi L\textsubscript{18} table (Figure S4) were prepared and the average of the measured electrical conductivity of the samples was used for the Taguchi analysis.

The time-dependent electrothermal behaviour of a conductive flax yarn was characterised. A thermometer was attached to the surface of the sample in the middle of the electrodes to monitor the surface temperature while different voltages were applied to the sample. The distance between the electrodes on the sample was 15 mm.

### 2.3. Taguchi design of experiment method

DoE is a statistical method that allows researchers to understand individual and interactive effects of manufacturing factors on the performance of the processes or products [18]. A typical DoE (e.g., a full factorial method) can assess all of the effective parameters on the output; however, it demands a great number of experimental trials and high cost and time. The Taguchi method as a highly effective and powerful DoE technique has been widely used to optimise the number of experiments and lower the cost and time using the specifically designed orthogonal arrays [18]. In this research, the Taguchi analysis was utilised to systematically assess the effect of each parameter in the experimental procedure, identify the most significant ones, and obtain the highest electrical conductivity. The desirable manufacturing conditions to achieve the highest electrical conductivity were then explored [23].

In Taguchi analysis, one of the major steps is to choose the suitable manufacturing factors and their sub-levels [18]. Consequently, factors including the amount of SDBS, the magnitude of applied voltage, the distance between electrodes, type of electrode, type of yarn, and the deposition time were chosen among all of parameters to be studied in accordance with our preliminary experiments and the previous studies [17]. This design was compatible with the L\textsubscript{18} Taguchi DoE layout with five factors with three levels and one factor with two levels (Table 1). Table 2 presents the selection of each parameter for each experimental setup in accordance with the L\textsubscript{18} Taguchi DoE layout.

According to Table 1, the two types of electrodes were carbon sheets and stainless steel; and the three types of yarns were flax bleached, non-bleached, and UHMWPE. The three deposition times were chosen to be 2, 6, and 10 minutes while the three gap sizes of 10, 15, and 20 mm were selected. The three values for applied voltage were set to be 7.5, 15, and 20 V whereas the three ratios of SDBS/GNPs were 1:3, 1:2, and 2:3. The concentration of GNP in DI water was kept constant (3 g/l). The amount of SDBS/GNPs ratio was not exceeded more than 2:3 as the electrical paths among adjacent graphene nanoplatelets, and consequently

| Type of electrode | Yarn type                | SDBS/GNPs ratio | Gap size (mm) | Voltage (V) | Deposition time (min) |
|------------------|-------------------------|-----------------|--------------|-------------|-----------------------|
| Carbon & Stainless steel | Flax non-bleached     | 1/3             | 10           | 7.5         | 2                     |
|                  | Flax bleached          | 1/2             | 15           | 15          | 6                     |
|                  | UHMWPE                 | 2/3             | 20           | 20          | 10                    |
the electrical transport on the coatings, would be weakened by the existence of surfactant molecules [19,20]. As a result, a large amount of surfactant would lower the electrical conductivity in accordance with the previous studies [19,20]. In addition, the hydrophobic nature of GNPs alone would result in their agglomeration in water. Subsequently, we chose the three ratios for SDBS/GNPs to be 1:3, 1:2, and 2:3. It is noteworthy that we could not select more ratios of SDBS/GNPs in accordance with the layout of L_{18} table (Figure S4). The interaction effects of the parameters were not taken into account in this study since the focus on the effects of the major parameters on the electrical conductivity and having a simpler analysis were of primary concern.

2.4. Signal to noise ratio (S/N) and pareto ANOVA

In the Taguchi analysis, signal-to-noise ratio (S/N) was selected instead of using the mean values of the electrical properties. The S/N ratio considers both the variation and average of experimental results. The desirable responses were set to maximise the electrical conductivity of the yarns coated with f-GNPs. Therefore, the ‘larger-the-better’ characteristic (Eq.(1)) was used to determine the combination of the factors to achieve the highest output as suggested in [23,24]:

\[
\frac{S}{N} = -10 \log \left( \frac{1}{n} \sum_{i=1}^{n} \frac{1}{y_i^2} \right)
\]  

where \( \frac{S}{N} \) denotes the signal-to-noise ratio based on improved output (electrical conductivity), \( n \) is the number of samples for each experimental trial, and \( y \) is the output (electrical conductivity) value [23,24].

A Pareto analysis of variance as the simplified version of ANOVA using the Pareto principle was conducted, as it can effectively analyse the results of parameter design [23,24]. More importantly, this tool can determine the significant factors and the optimum factor levels. The percentage of contribution of each

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Table 2. L_{18} Taguchi table, showing the settings for each experimental setup.

| Set | Deposition time (min) | Voltage (V) | Gap size (mm) | SDBS (g) | Base material | Type of electrode |
|-----|-----------------------|-------------|---------------|----------|---------------|------------------|
| 1   | 2                     | 7.5         | 10            | 1        | Flax non-bleached | Stainless Steel  |
| 2   | 6                     | 15          | 15            | 1.5      | Flax non-bleached | Stainless Steel  |
| 3   | 10                    | 20          | 20            | 2        | Flax non-bleached | Stainless Steel  |
| 4   | 6                     | 15          | 10            | 1        | Flax bleached   | Stainless Steel  |
| 5   | 10                    | 20          | 15            | 1.5      | Flax bleached   | Stainless Steel  |
| 6   | 2                     | 7.5         | 20            | 2        | Flax bleached   | Stainless Steel  |
| 7   | 10                    | 7.5         | 15            | 1        | UHMWPE         | Stainless Steel  |
| 8   | 2                     | 15          | 20            | 1.5      | UHMWPE         | Stainless Steel  |
| 9   | 6                     | 20          | 10            | 2        | UHMWPE         | Stainless Steel  |
| 10  | 6                     | 20          | 20            | 1        | Flax non-bleached | Carbon plate    |
| 11  | 10                    | 7.5         | 10            | 1.5      | Flax non-bleached | Carbon plate    |
| 12  | 2                     | 15          | 15            | 2        | Flax non-bleached | Carbon plate    |
| 13  | 2                     | 20          | 15            | 1        | Flax bleached   | Carbon plate    |
| 14  | 6                     | 7.5         | 20            | 1.5      | Flax bleached   | Carbon plate    |
| 15  | 10                    | 15          | 10            | 2        | Flax bleached   | Carbon plate    |
| 16  | 10                    | 15          | 20            | 1        | UHMWPE         | Carbon plate    |
| 17  | 2                     | 20          | 10            | 1.5      | UHMWPE         | Carbon plate    |
| 18  | 6                     | 7.5         | 15            | 2        | UHMWPE         | Carbon plate    |
factor to understand the most significant factors was evaluated by this tool [23,24]. Thus, the non-significant factors can ultimately be recognised. This tool can assist to optimise the experimental plans to enhance the electrical conductivity of the f-GNPs coated yarns.

3. Results and discussion

3.1. Fourier transform infrared spectroscopy (FTIR)

The ATR-FTIR spectra of the GNPs, SDBS, and the f-GNPs with SDBS are shown in Figure 2. The presence of O-H stretching at about 3400 cm$^{-1}$ is clear on the three spectra. The peaks at 2854, 2926, and 2957 cm$^{-1}$ are induced by the C-H stretching in SDBS. In addition, the presence of peaks for the spectra of f-GNPs and SDBS at 1030 and 1200 cm$^{-1}$ are related to the symmetric and asymmetric stretching vibrations of the –SO$_3$– groups. Moreover, the peaks appearing at 1601 and 1409 cm$^{-1}$ are due to the vibration of the phenyl groups of SDBS [25–28]. The results of ATR-FTIR clearly confirm the interaction of SDBS and surface of GNPs and agree well with the results of previous studies [25–28].

Furthermore, the ATR-FTIR spectra of flax and coated flax yarns, Figure S5(a), indicate the peaks at 3346 cm$^{-1}$ related to O-H groups. The peaks at 2915, 1734, 1429, 1370 cm$^{-1}$ are assigned to the C-H vibration, C = O stretching, –CH$_3$ asymmetric, and C-H symmetric deformation of lignin, respectively. In addition, a peak corresponding to COO$^-$ asymmetric stretching is observed at about 1620 cm$^{-1}$. The broad peak at 1058 cm$^{-1}$ is related to the C-O stretching [29,30]. Figure S5(b) exhibits the spectra related to UHMWPE and the f-GNPs coated UHMWPE. The obvious peaks can be explained as follows: the peaks at 2915, 2847, 1471, and 716 cm$^{-1}$ correspond to the asymmetric stretching vibration of C-H, the symmetric stretching vibration of C-H, the in-plane bending vibration of

![Figure 2. ATR-FTIR results of GNPs, SDBS, and SDBS f-GNPs.](image-url)
C-H, and the rocking vibration, respectively [31]. The spectra related to the f-GNPs coated flax and UHMWPE with the highest electrical conductivity showed similar peaks to those of neat flax and UHMWPE, respectively.

3.2 Thermogravimetric analysis (TGA)

The thermal properties of the conductive flax and UHMWPE yarns were investigated in order to understand their behaviour at higher temperatures to check their possible usage as heating elements. Therefore, TGA was conducted to evaluate the thermal stability of the yarns with and without coating. The thermal stability of the yarns decreased with the presence of f-GNPs coating due to the presence of functional groups on the surface of f-GNPs (Figure 3) [32]. In addition, the weight loss of samples in the beginning of the test was due to the evaporation of water remaining in samples. A dramatic weight loss occurred for the flax based sample from Set 4 before silver coating at about 270 °C, whereas the neat flax sample started degrading at about 320 °C. This behaviour could be attributed to the pyrolysis of the functional groups on the surface of f-GNPs [32].

Regarding the sample after coating with silver, it started degrading at about the same temperature as the neat sample. After a temporary stabilization, it started degrading again at about 450 °C. On the other hand, the UHMWPE sample showed a similar behaviour to that made of flax. Figure 3 shows that the neat UHMWPE yarn begins degrading at about 500 °C while the f-GNPs coated UHMWPE yarn degrade at approximately 300 °C. Moreover, the decomposition temperature of the silver-coated sample of UHMWPE was found to be approximately 400 °C. All samples exhibited excellent thermal stability at high temperatures (above 500 °C). The results of TGA demonstrate the feasibility of fabricating heating wires, based on flax and UHMWPE.

3.3 Electrical conductivities of various yarns

Figure 4 indicates the electrical conductivity values of various conductive yarns prepared in accordance with the setup in Table 2. The average electrical conductivity values of the neat flax and UHMWPE yarns were found to be $10^{-8}$ and $2 \times 10^{-17} \text{ S.m}^{-1}$, respectively. The values of electrical conductivity of the conductive yarns ranged from 0.02 to 0.56 S.m$^{-1}$. The highest average value of the electrical conductivity was obtained for Set 4 samples while the lowest average value of electrical conductivity was found for Set 8 samples. In the case of using UHMWPE, the samples of Set 16 showed the highest average value of electrical conductivity to be about 0.41 S.m$^{-1}$. In addition, the lowest average value for the same of conductive flax yarns was found to be 0.03 S.m$^{-1}$ (Set 15). The changes in the experimental setup for each set caused considerable changes in the electrical properties of the yarns, for which the averaged values were used later to understand the most influencing factors to achieve the highest electrical properties of the yarns coated by the EPD technique.

SEM images were taken to confirm the existence of the percolative networks of GNPs on the fibre surfaces of flax and UHMWPE, and study the surface morphology. Figure 5 exhibits the SEM image of the yarns coated with f-GNPs using the EPD technique and their images prior to the coating process. The surface of the neat bleached flax and UHMWPE yarns can be seen in Figure 5 (a-c). The conductive yarns of Set 4 represents a
relatively homogeneous distribution f-GNPs on the surface of the flax yarns leading to a higher value of electrical conductivity, Figure 5 (d). On the other hand, after the EPD process, the yarns of Set 16 shows the lack of interconnected conductive particles on the surface of UHMWPE yarns, resulting in a lower value of electrical conductivity compared to those of Set 4, Figure 5 (e) [33–35]. To understand the mechanism of coating, cross-sectional SEM images were taken after the samples were fractured in liquid nitrogen. Figure 5 (f) depicts the distribution of f-GNPs on the surface of a natural fibre yarn. The f-GNPs mostly covered the outer parts of the yarn while a lower number of particles penetrated through the fibres. This shows that f-GNPs form more percolative networks on the outer surfaces of the fibres.
Figure 4. Average values for electrical conductivity of the yarns with regard to the Set of experiment after EPD process.

Figure 5. SEM image of bleached flax (a, b), UHMWPE (c), f-GNPs coated flax from Set four (d), and UHMWPE coated with f-GNPs from group 16 (e) distribution of f-GNPs on the surfaces of a yarn (f).
3.4. Evaluation of significant factors

In the Taguchi method, the analysis of the $\frac{S}{N}$ ratios with the conceptual approach would lead to the identification of the significant factors. Furthermore, the Pareto ANOVA tool can effectively determine the preferred combination of factors in which the highest electrical conductivity is achieved. In other words, the effect of noise factors on the performance considering the variation in response data and the deviation of average to the target values are measured by $\frac{S}{N}$ ratios. Here, the $\frac{S}{N}$ ratio equation based on the ‘larger-the-better’ characteristics was chosen to maximise the output values of electrical conductivity. The normalised value of electrical conductivity for each set of sample was derived and used for calculation of $\frac{S}{N}$ ratios. The Pareto ANOVA tool identified the contribution of each factor including the amount of SDBS to GNP ratio, the magnitude of applied voltage, yarn type, the deposition time, the gap size between the electrodes, and the type of electrode on the electrical properties of the yarns. The results of this analysis are depicted in Figure 6. The ratio of the SDBS to GNPs has the most significant effect on the enhancement of the electrical conductivity with a contribution percentage of 56.68%. This factor is the most critical one due to the presence of anionic charges on the surface of the f-GNPs, resulting in their migration towards the location of the yarns (anode) during deposition. Moreover, the magnitude of applied voltage played an important role on improving the electrical properties of the coated yarns, where its contribution percentage was found to be 16.55%. The third important factor was recognised to be the type of fibre by the contribution percentage of 15.41%. The least important factor was found to be the electrode material with only 2.27% of contribution.

3.5. Determination of preferred combinations of factors

The ‘higher-the-better’ characteristic in the Taguchi method expresses that a higher summation of $\frac{S}{N}$ ratio results in a better response of factorial effects. This analysis would identify the best level for each factor based on the summation of $\frac{S}{N}$ ratios such that the

![Figure 6](image_url)

Figure 6. Pareto ANOVA analysis for understanding the most contributed factors to enhance the electrical conductivity of the yarns by EPD method.
combination of these levels would be selected as the preferred combination of factors in accordance with the Taguchi method. The results of this analysis are presented in Figure 7. The summation of $S/N$ ratios to enhance the electrical conductivity of the conductive yarns are depicted. The plot shows that in the case of the type of electrode, the higher summation of $S/N$ ratio was corresponded to the carbon sheet electrode. In addition, UHMWPE showed the highest summation of $S/N$ among all types of yarns. The optimum amount of SDBS to GNPs was found to be 1:3 using EPD coating process due to the highest value of $S/N$ ratio for this level. This could be due the fact that higher amounts of SDBS could deteriorate the electrical conductivity of the GNPs and their network [19]. The results determine that the gap size of 10 mm, magnitude of applied voltage of 7.5 V and the deposition time of 6 min possess the highest value of summation of $S/N$ for each of the factors. Thus, the combination of these levels could produce the preferred combination of the factors in which the highest electrical conductivity based the conditions of the experiment would be obtained. As a result, the preferred combination of the factors is using a ratio of 1:3 for SDBS to GNP, applied voltage of 7.5 V, the gap distance of 10 mm, 6 min of deposition time, UHMWPE as the yarn material, and carbon sheet electrodes. The samples were fabricated following this setting and the average value of electrical conductivity was found to be 0.64 S.m$^{-1}$.

3.6. Electrothermal behaviour of the conductive natural fibre yarns

Joule heating effect is defined as the heat dissipated as the input electric power applied on an electrically conductive material [36]. In this part, we selected a conductive flax yarn from Set 4 to investigate its electrothermal behaviour. Considering the level of electrical conductivity of the samples of this Set, we enhanced the level of conductivity by a simple dip-coating of a f-GNP coated flax yarn in silver paste; hence, its resistance

![Figure 7. Sum of S/N ratio at different factorial level for enhancing the electrical conductivity of the f-GNPs coated yarns.](image-url)
dramatically decreased. The measured resistance between the two ends of wires with a
gap of 15 mm was found to be ~ 20 Ω. At this stage, the electrothermal behaviour of the
conductive yarn with regard to different voltages (1, 2, and 3 V) was assessed by
monitoring the output surface temperature on the sample by a thermometer. Figure 8
depicts a typical temporal temperature change of the sample under different applied
voltages. The surface temperature of the sample increased as the voltage increased and
reached the steady-state stage with the maximum temperature of 102 ºC within 36 s for
the case of applying 3 V and decreased to the ambient temperature after voltage cut off
in about 42 s. This behaviour was identical for the other applied voltages (Figure 8). The
electrothermal performance of our conductive flax yarns outperformed some of the
recently reported heaters, as compared in Table S1.

The temperature responsiveness and energy efficiency parameters for various applied
voltages were assessed with the aid of the experimental time-dependant temperature
curves (Figure 8). This plot can be divided to three different stages including elevating
temperature (heating process), steady-state temperature (equilibrium), and lowering
temperature to the ambient temperature (cooling process). The first stage of the
temperature time-dependent profiles can be expressed in the following Eq. (2) in
accordance with the previous studies [36–38]:

$$\frac{T_t - T_i}{T_{max} - T_i} = 1 - \exp\left(-\frac{t}{\tau_g}\right)$$

where $T_i$, $T_{max}$, and $T_t$ denote the initial ambient temperature, the steady-state
maximum temperature, and an arbitrary temperature at time $\tau_g$, respectively. This
parameter was calculated by fitting curves on the experimentally obtained results and
found to be 6.55 ± 2.75 s. Subsequently, the maximum temperature at any applied
voltage occurred at the steady-state stage. The heat generated by the input electric
power equals the heat loss through radiation and convection in accordance with the

![Figure 8](image-url)

Figure 8. Time-dependent temperature changes of the conductive flax sample at different applied
voltages from 1 to 3 V.
conservation law of energy. Therefore, the heat loss can be evaluated by Eq. (3) found in the previous studies [36–38]:

\[ h_{r+c} = \frac{P_{in}}{T_{max} - T_i} = \frac{V^2}{R} } \frac{1}{T_{max} - T_i} \]  

(3)

where \( R \) is the resistance of the flax yarn and \( V \) is the applied voltage. Accordingly, the \( h_{r+c} \) value was calculated to be 3.93 ± 1.51 mW°C\(^{-1}\). The third stage of the temperature time-dependent profile can be fitted by the following formula given in Eq. (4), as suggested in the previous studies [36–38]:

\[ \frac{T_t - T_f}{T_{max} - T_f} = \exp\left(-\frac{t}{\tau_d}\right) \]  

(4)

where \( T_t \) represents the final ambient temperature and \( \tau_d \) a time constant. The value of \( \tau_d \) was found to be 34.98 ± 10.67 s through fitting experimental time-dependent temperature decay data. As can be inferred from these data, the relatively low value of \( \tau_d \) demonstrates the relatively higher thermal and electrical conductivity of the sample compared to the one reported in the literature, which results in a rapid temperature response of the sample to the applied voltage [37]. In addition, a lower value of \( h_{r+c} \) indicates a relatively high electrical conductivity with higher electric energy efficiency due to the lower amount of electrical energy to obtain the maximum temperature at a given voltage.

Figure 9 depicts the increment in \( T_{max} \) with regard to the applied voltage that are in-line with the change of input electric power with applied voltage, which can be expressed as \( P_{in} = \frac{V^2}{R} \). Here, \( P_{in} \) is the input electric power, \( V \) is the applied voltage, and \( R \) represents the electrical resistance. Consequently, the \( T_{max} \) for various cases

![Figure 9](attachment:figure9.png)
increased in a linear manner as the $P_{in}$ increased. By fitting a tread-line on the plot, $T_{max}$ can be well-defined with regard to the $P_{in}$ using Equation (5);

$$T_{max} = 129.96P_{in} + 45.44$$

(5)

4. Conclusions

In this study, electrically conductive yarns (flax and UHMWPE) were developed using dispersions containing SDBS functionalised GNPs in DI water. The EPD technique was employed to coat the conductive micro/nanoparticles on the surface of the yarns. Since a number of parameters effect the electrical conductivity during EPD process, a parametric study on enhancement of electrical conductivity of the yarns was conducted via the Taguchi method. Later, the most contributed factors were identified by Pareto ANOVA analysis and it was found that the amount of SDBS could play a key role in enhancing the electrical conductivity of the yarns. The preferred combination of factors through Taguchi method was found to be using a ratio of 1:3 for SDBS to GNP, applied voltage of 7.5 V, the gap distance of 10 mm, six minutes of deposition time, UHMWPE as the yarn material, and carbon sheet as electrodes. The average electrical conductivity value of the fabricated samples considering the preferred combination in EPD setup was found to be 0.64 S.m$^{-1}$. A flexible heating wire based on a conductive flax yarn dip-coated with silver paste was characterised and the maximum surface temperature of the wire was found to be 102 ºC with the input power of 0.45 W (or applied voltage of 3 V) between the gap of 15 mm. The performance of the wire suggested its potential applications in wearable heaters and other warming scenarios.

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Disclosure statement

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