Engineering Graphene Flakes for Wearable Textile Sensors
via Highly Scalable and Ultrafast Yarn Dyeing Technique

Shaila Afroj,¹,²‡ Nazmul Karim,¹‡* Zihao Wang,² Sirui Tan,³ Pei He,³,⁴ Matthew Holwill,¹,² Davit Ghazaryan,⁵,² Anura Fernando³ and Kostya S Novoselov¹,²*  

¹National Graphene Institute (NGI), The University of Manchester, Booth Street East, Manchester, M13 9PL, UK  
²School of Physics & Astronomy, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK  
³School of Materials, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK  
⁴School of Physics and Electronics, Central South University, Changsha 410083, China  
⁵Department of Physics, National Research University Higher School of Economics, Moscow, 105066, Russian Federation  

*Corresponding author: mdnazmul.karim@manchester.ac.uk, (N.K.), kostya@manchester.ac.uk (K. S. N.)  

‡ These two authors contributed equally as joint first author
S1. Methodology

Figure S1 represents the process sequence in order to manufacture graphene-based textile sensors in a scalable quantity. We engineer the graphene flakes and optimise the processing parameters in order to find the best formulation for our applications. We then use laboratory scale yarn dyeing machine in order to coat a batch of 100% cotton yarn. This could readily be scaled up into an industry scale yarn dyeing machine that could potentially produce tonnes (~1000 kg/hr) of graphene-based conductive yarns. We then integrate graphene coated yarn into knitted fabric structure which could then be connected to a self-powered RFID or a low-powered Bluetooth in order to transmit the data wirelessly. The wearable garments thus produced and integrated with graphene textiles sensors can monitor the body temperature wirelessly and send to a mobile device.

Figure S1 | Graphical representation. Engineering graphene flakes to wearable textile sensors via highly scalable and ultra-fast yarn dyeing technique. Illustration by Kazi Farhan Hossain Purba and used with permission from the artist.
S2. Graphene Materials Synthesis and Characterisation

One of the major challenges with graphene’s commercial development is its poor colloidal stability in common solvents. The dispersibility of the graphene derivatives could be arranged as follows: GO > rGO > G. As GO is highly functionalized, therefore it provides higher yield, good colloidal stability and excellent dispersibility. After reduction to rGO, the dispersibility becomes almost the same as G; however the fact is that rGO disperses more effectively in polar solvents than G may be due to the presence of some residual functional groups even after reduction. The dispersibility of rGO could be improved by introducing an energy barrier to aggregation, through either covalent or non-covalent interactions. We synthesize graphene oxide using a modified Hummers method and chemically reduce GO to rGO by modifying our previously reported methods. We use ascorbic acid (AA) and sodium hydrosulphite (SH) as reducing agents and functionalise the surface of rGO flakes using PSS/PVA to have better dispersibility and prevent agglomeration. We also exfoliate graphene-based inks (G) using a highly scalable microfluidization technique. We use sodium dexooycholate (SDC) as a surfactant in order to disperse graphene flakes in water through non-covalent bonding.
Figure S2 | Flake size distribution of rGO and G flakes. a) rGO (SH) PSS, b) rGO (AA) PSS, c) rGO (AA) PVA and d) G Flakes. The statistical analysis of the optical images of 100 flakes show that the mean lateral dimension of GO is ~5.85 µm. After reduction with sodium hydrosulphite (SH) and L-Ascorbic Acid (AA), the flake size is reduced to ~4.86 µm, may be due to the stresses during pre-mixing and centrifugation steps in post-washing cycles. We also obtain lower flake size for G flakes (~1.45 µm), may be again due to the extremely high shear rate (10^8 s^-1) it is subjected to during the microfluidization of graphite.
Figure S3 | Flake thickness distribution of rGO and G flakes. a) rGO (SH) PSS. b) rGO (AA) PSS. c) rGO (AA) PVA and d) G Flakes. The statistical analysis of the AFM images of 100 flakes show that the mean flake thickness of GO synthesized by modified Hummers method is ~2.07 nm. After reduction with sodium hydrosulphite (SH) and L-Ascorbic Acid (L-AA) in the presence of polymers, the flake thickness increases slightly to ~2.2 nm. This indicates the presence of single to few layers of GO and rGO flakes. Moreover, the flakes thickness increased after reduction may be due to the presence of cross-linking polymers (such as PVA and PSS). After microfluidization (20 cycles) of graphite flakes, the obtained G flakes (figure S2d) become thinner with ~20% of G flakes are <10 nm in thickness, however few layers graphene or graphitic dispersions are obtained with microfluidization.
Figure S4 | Raman spectra. a) GO and rGO. b) G flakes. The Raman spectra of GO and rGO shows characteristic peaks at ~ 1344.78 cm$^{-1}$ and 1605.95 cm$^{-1}$, corresponding to D and G bands. The intensity ratio of the D to G band ($I_D/I_G$) is increased after reduction, from ~0.98 for GO to upto ~1.75 for rGO, Table S1, which suggests recovery of hexagonal carbon atoms and the generation of a large number of sp$^2$ domains in rGO. Raman spectra of exfoliated G flakes displays a typical spectrum like liquid phase exfoliated graphene after 20 cycles, with characteristics D peak at 1350 cm$^{-1}$, G peak at 1582 cm$^{-1}$ and an asymmetric 2D band at 2730 cm$^{-1}$, Figure S5b.

S3. Stability of Graphene Dispersions

Although the reduction of graphene oxide is a popular approach, a major problem associated with this route is that rGO aggregates in aqueous solution due to its hydrophobic nature, especially in higher concentration. To prevent re-stacking of graphene flakes, surface characteristics can be modified by adding the functionality to the surface. These functional groups can be added via either covalent bonding or non-covalent interactions.$^4, 10$ Covalent bonds are usually achieved by atom doping or reaction with the residual functional groups of graphene; whereas, non-covalent bonds are generally formed by van der Waals force, hydrogen bonding, electrostatic interaction, p–p stacking interactions and coordination bonds. When graphene’s unique electrical property and large surface area are expected, the
modification by forming non-covalent bonds is typically preferred. They are comparatively weaker than covalent bonds but multiple non-covalent bonds working in harmony can yield highly stable modifications. Moreover, these non-covalent bonds are easy to achieve over the entire graphene surface. On the other hand, covalent bonding is commonly preferred when stability and the strong mechanical properties of modified graphene are required.

Stankovich et al.\textsuperscript{11} reported that the stable rGO dispersion can be prepared by the surface modification using poly (styrenesulfonate) (PSS). Since then, a significant amount of works\textsuperscript{12-15} has been carried out to uncover the prospect of functionalised rGO. Wei et al.\textsuperscript{12} used several modification media such as poly(styrenesulfonate) (PSS), polyelectrolyte, polyaniline and ionic liquid to prepare high quality, free-standing graphene films. The organic functional groups with different polarities resulted in very different physiochemical properties such as surface tension, adhesion and wettability. The surfactant (PSS anions) functionalised graphene dispersion presented the best wettability, and great adhesion to the substrate. Swain et al.\textsuperscript{15} used two cross-linking polymers, Polyvinylpyrrolidone (PVP) with the ring structure and poly vinyl alcohol (PVA) with the linear chain, for preparing functionalised graphene dispersion. The polymers cross-linked and protruded into the solvent to achieve better solubility which was stable for over six months. Moreover, they stabilized sterically to avoid the agglomeration of rGO sheets.

We used PSS and PVA in order to modify the surface of rGO flakes and make it stable in water. Figure S5-7 shows the aggregation of rGO flakes without or with a very small amount of polymer (1:0 and 1:1). However, the addition of polymers (1:5 and 1:10 ratios) significantly improves the dispersibility of rGO.\textsuperscript{6} The presence of polymer (PVA and PSS) plays an important role in stabilizing the rGO dispersion. Moreover, the stability of the dispersions improves with the increase of rGO reduction time, may be due to better functionalization of the surface of rGO flakes with increased reaction time.
Figure S5 | Stability of rGO (AA) PSS dispersions. Images showing dispersity of rGO inks at various reduction time and amount of polymer (PSS)

Figure S6 | Stability of rGO (AA) PVA dispersions. Images showing dispersity of rGO inks at various reduction time and amount of polymer (PVA)
**Figure S7** | **Stability of rGO (SH) PSS dispersions.** Images showing dispersity of rGO inks at various reduction time and amount of polymer (PSS)

**S4. The Degree of rGO Reduction – Raman and XPS Analysis**

We reduce GO to rGO chemically using Na$_2$S$_2$O$_4$ and L-AA. Figure S8 shows the change of the colour of GO dispersions to opaque black almost immediately, which is not the case for L-AA. This confirms faster and efficient reduction of GO with Na$_2$S$_2$O$_4$, which is in agreement with previous studies.\textsuperscript{16, 17}

**Figure S8** | **Reduction rate of GO with SH.** Image shows GO (golden brown) turns black immediately after adding Sodium Hydrosulphite
We then analyze the $I_D/I_G$ ratio of various rGO formulations in order to observe the degree of reduction and find a best rGO formulation. In order to capture the Raman spectra, clean samples were prepared on oxidised Si substrates (with 290 nm thick SiO$_2$ layer) by drop casting a very low concentration (apprx 0.1 mg/mL) of rGO dispersion. Raman spectra were captured using Renishaw Raman System equipped with 633 nm laser and spectra were taken at 5 different locations of each sample.

All sp$^2$ bonded carbon show a common Raman shift where the G peak is at ~1580 cm$^{-1}$ and the D peak is at around ~1360 cm$^{-1}$.$^{18}$ The D peak is the disorder band or defect band that shows a spectrum due to the breathing modes of six-atom rings and observed when defects are present.$^{19}$ Moreover, the increment in the intensity ratio ($I_D/I_G$) indicated the generation of large number of sp$^2$ domain and formation of rGO from GO.$^{15}$ Table S1 shows mean of $I_D/I_G$ ratio of rGO formulation that could be used to observe the degree of reduction with various reduction time and reducing agents.

**Table S1 | $I_D/I_G$ ratio.** The rGO samples reduced at various reduction times with different reducing agents and different amount of polymers.

| Reduction time | rGO (AA) PSS 1:5 | rGO (AA) PSS 1:10 | rGO (SH) PSS 1:5 | rGO (SH) PSS 1:10 | rGO (AA) PVA 1:5 | rGO (AA) PVA 1:10 |
|----------------|-----------------|-------------------|-----------------|-----------------|-----------------|-----------------|
| 12 hrs         | 1.53            | 1.52              | 1.71            | 1.73            | 1.55            | 1.63            |
| 24 hrs         | 1.51            | 1.55              | 1.73            | 1.75            | 1.55            | 1.58            |
| 48 hrs         | 1.52            | 1.55              | 1.64            | 1.70            | 1.57            | 1.60            |
| 72 hrs         | 1.55            | 1.49              | 1.64            | 1.63            | 1.67            | 1.63            |

Table S1 shows that a better reduction can be achieved (maximum $I_D/I_G$ ~ 1.73) by using Na$_2$S$_2$O$_4$ compared to L-AA. However, $I_D/I_G$ decreases after 24 hours while reduced with Na$_2$S$_2$O$_4$. On the other hand, by using L-AA a better reduction can be achieved while reduced in presence of PVA compared to PSS. Although in both cases, the degree of reduction time
does not show a clear trend, in most of the cases better reduction is achieved with higher GO to polymer ratio (GO: polymer in 1:10 ratio).

We also use XPS analysis in order to confirm the degree of reduction by observing the change in carbon content and the reduction of oxygen containing groups in GO. A Kratos Axis system spectrophotometer was used to perform the XPS analysis. The inks were drop casted on PEL paper, dried at 100 °C temperatures for 5 minutes and attached to the sample holder using a double sided tape. The samples were irradiated using monochromatic Al Kα X-rays (1486.6 eV) with a power of 150 W and wide scan spectra were recorded with pass energy of 160 eV from which the surface composition (C and O) was determined. High resolution carbon (1s) and oxygen (1s) spectra were recorded with pass energy of 40 eV and the binding energy (BE) values were calculated relative to the Carbon (1s) photoelectron peak at 285.0 eV. All samples were analysed in duplicate and data was analysed using the CASA XPS software.

XPS analysis further confirms the degree of GO reduction. Wide scan XPS spectra show a sharp peak at 533 eV for GO, representing the domination of oxygen containing components like C-O and C=O in GO.\(^{20}\) When GO is reduced to rGO, a sharp decrease in the intensities of all of the oxygen related peaks are observed, indicating the restoration of C-C π bond in rGO samples.\(^{5}\) Consequently, the C/O of GO 2.41 increases upto ~6.59 in rGO which indicates the reduction of GO to rGO. The C/O ratio shows a higher degree of reduction can be achieved by Na\(_2\)S\(_2\)O\(_4\) upto 24 hours reduction time, which is in agreement with the results from Raman analysis. Whereas for L-AA, C/O is found to be almost similar for all the reduction times.
The high resolution spectrum for GO shows two main peaks that can be fitted to four components arising from C–O groups (hydroxyl and epoxy, $\sim$286.5 eV), the C–C bond ($\sim$284.6 eV), C=O (carbonyl, $\sim$288.3 eV) groups and O–C=O (carboxyl, $\sim$290.3 eV) groups.$^{21-23}$ After reduction, the peaks associated with the oxygen functional groups significantly diminished, Figure S9-10. The small bump around 288.5 eV indicated some residual oxygen functionality and they are within the ketone and carboxyl carbon region.

**Figure S9**|High resolution XPS spectra. rGO reduced by Na$_2$S$_2$O$_4$ with 1:10 GO to polymer (PSS) ratio. a) 12 hours. b) 24 hours. c) 48 hours. d) 72 hours.
**Figure S10** | High resolution XPS spectra. rGO reduced by L-AA with 1:10 GO to polymer (PSS) ratio. a) 12 hours. b) 24 hours. c) 48 hours. d) 72 hours.

**S5. Temperature Dependence of rGO Resistances and I-V Curves**

As discussed earlier, we make 48 formulations in order to optimize the GO reduction for both AA and SH using various reduction time (12, 24, 48, 72 hrs) and four stoichiometric ratios (1:0, 1:1, 1:5 and 1:10) between GO and polymers (PSS/PVA). We drop-cast graphene dispersions on Si/SiO$_2$ wafer using a micropipette and dry them at room temperature.

We then measure the conductivity of graphene inks from I-V curves using a 2–probe measurement at various temperature ranges (150K to 280K), Figure S11-15. We extract the resistance for each sample from the slope of I-V curves (insets). The insets figures show linear I-V characteristics which indicate the ohmic nature of rGO. As seen from Figure S11-
15, the resistance (R) decreases monotonically with the increase of temperature up to ~200K and then become linear for almost all samples, which is similar to the behavior of an intrinsic semiconductor. Previous studies have reported a similar temperature dependence of carbon nanotubes\textsuperscript{24} and graphene\textsuperscript{25}; however, the negative resistance co-efficient of these samples may not be intrinsic, perhaps it could be due to the presence of high density of defects in rGO.\textsuperscript{26}

\textbf{Figure S11|Electrical transport measurements.} Temperature dependence of the resistances rGO (SH) samples prepared with 1:5 polymer (PSS) ratio with various reduction time.
**Figure S12** | Electrical transport measurements. Temperature dependence of the resistances rGO (SH) samples prepared with 1:10 polymer (PSS) ratio with various reduction time.

**Figure S13** | Electrical transport measurements. Temperature dependence of the resistances rGO (AA) samples prepared with 1:5 polymer (PSS) ratio with various reduction time.
Figure S14 | Electrical transport measurements. Temperature dependence of the resistances rGO (AA) samples prepared with 1:10 polymer (PSS) ratio with various reduction time.

Figure S15 | Electrical transport measurements. Temperature dependence of the resistances rGO (AA) samples prepared with 1:5 polymer (PVA) ratio with various reduction time.
Table S2. Fitting parameters. The reduction conditions of rGO inks and the fitting parameters used for the investigated devices for Mott’s variable range hopping

\[ G = G_0 \exp \left( -\frac{T_0}{T} \right)^\alpha \]

| Reducing Agents and Polymer | Reduction Time (hrs) | GO to Polymer Ratio | \( T_0 \) (K) | \( G_0 \) (S) |
|-----------------------------|----------------------|---------------------|----------------|-------------|
| L-AA and PVA                | 12 01:05             | 978                 | 6.71E-08       |
|                             | 12 01:10             | 1215                | 1.12E-08       |
|                             | 24 01:05             | 732                 | 3.63E-08       |
|                             | 24 01:10             | 1011                | 2.97E-08       |
|                             | 48 01:05             | 834                 | 3.07E-07       |
|                             | 48 01:10             | 827                 | 1.54E-07       |
|                             | 72 01:05             | 792                 | 3.66E-07       |
|                             | 72 01:10             | 707                 | 8.38E-08       |
| SH and PSS                  | 12 01:05             | 1328                | 1.03E-07       |
|                             | 12 01:10             | 1156                | 1.01E-07       |
|                             | 24 01:05             | 955                 | 3.36E-07       |
|                             | 24 01:10             | 708                 | 1.51E-07       |
|                             | 48 01:05             | 873                 | 4.56E-08       |
|                             | 48 01:10             | 882                 | 1.55E-08       |
|                             | 72 01:05             | 861                 | 2.02E-08       |
|                             | 72 01:10             | 868                 | 1.96E-09       |
| L-AA and PSS                | 12 01:05             | 823                 | 1.06E-07       |
|                             | 12 01:10             | 1685                | 1.08E-07       |
|                             | 24 01:05             | -                   | -              |
|                             | 24 01:10             | 1178                | 5.19E-07       |
|                             | 48 01:05             | 834                 | 5.53E-07       |
|                             | 48 01:10             | 902                 | 6.52E-07       |
|                             | 72 01:05             | 950                 | 9.61E-07       |
|                             | 72 01:10             | 907                 | 5.31E-07       |
S6. Wash Stability

Figure S16 | SEM images of unwashed and washed samples. a) SEM image of rGO (SH) dyed (coated) cotton yarn (1000x magnification). b) SEM image of rGO (SH) dyed (coated) cotton yarn after 10 washing cycles (1000x magnification). c) SEM image of G flake dyed (coated) cotton yarn (1000x magnification). d) SEM image of G flake dyed (coated) cotton yarn after 10 washing cycles (5000x magnification).

As evident from the wash stability tests, GO provides better durability than G flakes due to the presence of residual functional groups, Figure S9-10. SEM images of rGO coated yarn shows uniform coating of rGO flakes on the surface, Figure S16a. After 10 washing cycles in a home laundry machine, the coating still exist on dyed fibres with only small number of unfixed flakes are removed the surface, Figure S16b. Therefore rGO coated yarns are still conductive even after 10 washing cycles. In contrast, G flakes are removed from the yarn.
surface after few washing cycles. This may be due to the absence of the residual functional
groups in G flakes as seen from the high resolution XPS spectrum. Figure S16c shows large
amount of G flakes on the fibre surface, which are removed after 10 washing cycles with only
few flakes left on the surface, figure S16d.

**S7. Knitted Graphene-Based Temperature Sensors**

*Figure S17| Knitted graphene yarn. a, front and b, the end elevation of the graphene yarn
knitted stitch*

Wearable knitted temperature sensor is designed to work under un-stretched condition. In this
state, the head of the loop binds the legs of the loop together causing the electro-conductive
pathway to be approximately a straight line. This causes the width of the sensor fabric to be
the length of the conductive path way for a single knitted course. Therefore, according to the
Holm’s theory:

\[
R_c = \rho l_{\text{width}} + \frac{\rho}{2} \sqrt{\frac{\pi H}{nP}}
\]

\[
R_i = \frac{\rho l_{\text{width}}}{N} + \frac{\rho}{2} \sqrt{\frac{\pi H}{nP}}
\]

Where

\(\rho = \text{Electrical resistivity of the graphene yarn}\)
$l_{\text{width}} =$ Width of the knitted sensor fabric in the knitted weft direction

$R_c =$ Resistance of a single knitted course of graphene yarn in ohm

$n =$ Number of contact resistance points in the graphene yarn knitted course

$R_s =$ Total resistance of the graphene yarn sensor in ohm

$H =$ Material hardness

$N =$ Number of knitted courses of graphene yarn in the sensor

$P =$ Contact pressure
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