

The Effect of Graphene Oxides Sheets on the Mechanical Properties of Graphene Fibres*

Heng Zhai, Yi Li*, Yang-Yang Fan

School of Materials, University of Manchester, Oxford Road, Manchester, M13 9PL, UK

Abstract

Graphene Oxides (GOs) of different sheet size were prepared by a novel modified Hummers' method. The heating time of expandable graphite, and oxidizing time of expanded graphite are demonstrated in order to have an influence on GO sheet size. Over-heated graphite in the microwave causes the decrease of GO sheet size, leading to a reduction in the performance of graphene fibres' mechanical properties. The oxidation mechanism was proposed by analyzing the chemical structure model of GO and evaluating its functional groups. Aggregation of GO sheets leads to the increase of internal shear stress, thus changing the viscosity of GO dispersion. Viscosity is another characteristic relating to the forming of graphene oxide liquid crystals (GO LC) and its phase transformation, which affects the mechanical properties of graphene fibres in the process of wet-spinning.

Keywords: GO dispersion; GO size; heating time; oxidation time; GO LC; viscosity; mechanical properties

1 Introduction

Graphene, as the elementary component part of all the graphene-based materials, is a flat two-dimensional (2D) monolayer of sp^2 -hybridized carbon atoms tightly piled into a honeycomb lattice [1]. It is a rapidly rising star in the field of material science and condensed-matter physics [2]. Due to its extraordinary electronic, mechanical and thermal properties it has attracted continuous attention, which mainly arises from its unique atomic thickness and 2D structure [3]. Nevertheless, studies of graphite tend to utilise increasingly fewer layers for some time. Indeed, it was Geim and his co-workers at the University of Manchester who first isolated single layer samples from graphite in 2004 and discovered a new page for the development of graphene [4]. From then on, promising applications for graphene in electronic devices, sensors and composites have led to an explosion in interest. Among all the applications, the outstanding properties of graphene fibres are optimized in macroscopic aspects by one-dimensional graphene (1D) fibres.

*Project supported by EU Horizon 2020 and University of Manchester (UMRI project "Graphene-Smart Textiles E-Healthcare Network", 644268-ETEXWELD-H2020-MSCA-RISE-2014, AA14512, R1119938.s).

*Corresponding author.

Email address: henry.yili@manchester.ac.uk (Yi Li).

However, graphene cannot be used to fabricate graphene fibres directly due to inherent deficiencies, such as limited dispersibility in common solvents, which hinders development of the direct scalable assembly of graphene. Whereas, the most accessible precursor of graphene, which is named as chemically oxidized graphene or graphene oxide (GO), with abundant oxygen functional groups on its basal planes and edges, is able to be completely exfoliated. Afterwards, colloidal dispersions of a single sheet are formed in water or other polar organic solvent [5]. The superiority of GO, such as long-term solubility, easiness to handle, and scalable production with low cost makes it reliable in assembling 2D sheets into macroscopic graphene-based architectures [1]. Wet-spinning seems to be the most effective, easy-handle and fastest method to produce large-scale graphene fibres without by-products. The continuous process of fabrication from graphene and graphene oxide to graphene fibres via wet-spinning ensures that high-performance graphene fibres have become widely applied in various domains.

Until now, research efforts have been driven by the application of high-performance 2D films, paper, and membranes in electric double-layer capacitors and pseudocapacitor stretchable electrodes, biosensors, and biomedical applications due to their respectable mechanical strength, electrical conductivity, signal-to-noise ratio and potential biocompatibility [6]. Regarding to three-dimensional (3D) graphene structures, it is evident that achievements in 3D graphene have been made in electrodes of supercapacitors, supercapacitors, conductors with flexibility, and biosensors [7]. Compared with 2D and 3D graphene materials, a one-dimensional (1D) graphene fibre was believed to show high electrical conduction, ultrahigh mechanical strength after stretching, functional possibility, unique flexibility and weavability [8]. Nowadays, 1D graphene fibres have already been prepared and confirmed for possessing multiple superior properties in comparison to conventional large-scale materials.

Here, we report a way of fabricating graphene fibres through wet spinning. By altering the duration of heating process of expandable graphite and oxidizing process of expanded graphite, different mechanical properties of graphene fibres could be achieved. Among them, graphene oxide dispersion synthesized by 30 seconds' heating in microwave and 24 hours' oxidation process could be used to spin the most robust graphene fibres, thereby is endowed with tensile strength as 24.16 MPa and Young's modulus as 4.49 GPa. Besides, the particle size and viscosity of graphene oxide dispersion are also tested to explain the effect to mechanical properties of graphene fibres.

2 Experiment

2.1 Synthesis of GO

As an important intermediate to fabricate graphene fibres, GO has a crucial role in the overall properties of graphene fibres. The synthesis of GO is based on the modified Hummers' method [9]. Expandable graphite is severed as the raw material.

0.5 g expandable graphite was heated at 750 W in the microwave oven for 20, 30 and 40 seconds respectively. Then, 100 ml of 98% concentrated sulfuric acid and 5 g of potassium permanganate were added to the expanded graphite. The oxidation time of expanded graphite is 24 or 48 hours. The following steps were observed by the Hummers' method. Finally, six groups of GO dispersions were obtained:

Table 1: Six groups of GO by different time settings of heating and oxidation

Oxidation	Heating		
	20 s	30 s	40 s
24 h	(20 s, 24 h)	(30 s, 24 h)	(40 s, 24 h)
48 h	(20 s, 48 h)	(30 s, 48 h)	(40 s, 48 h)

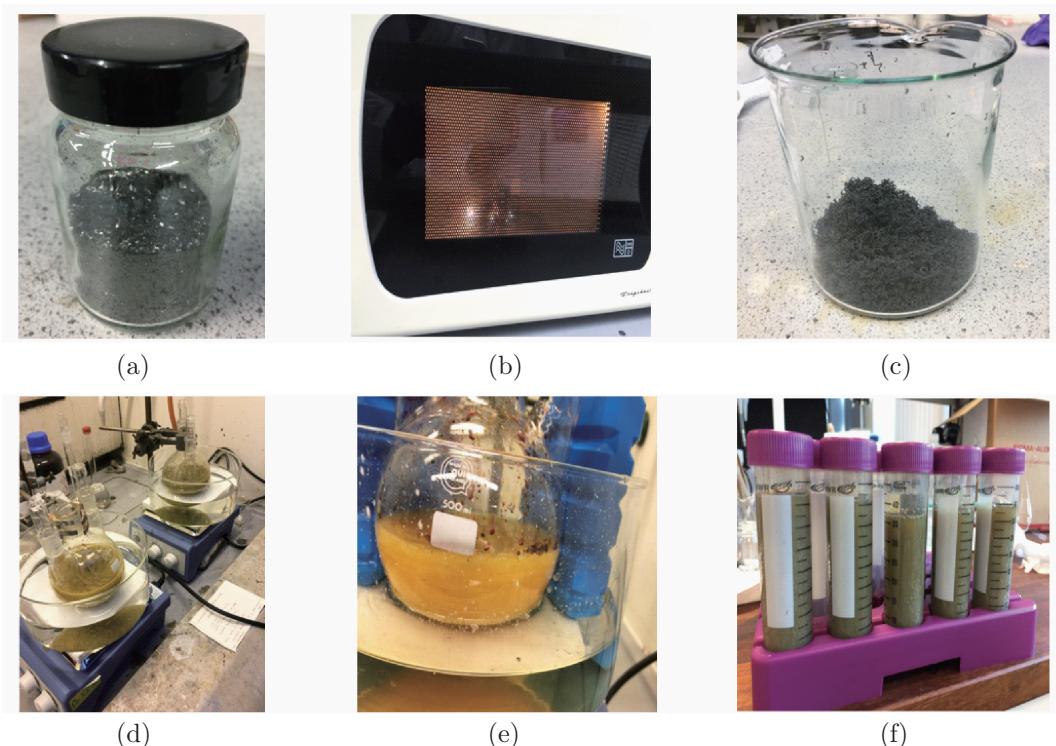


Fig. 1: (a) Expandable graphite; (b) Expandable graphite being heated in microwave oven; (c) Expanded graphite, (d) Expanded graphite under oxidation; (e) Colour changes to blonde after adding H_2O_2 under an ice bath; (f) Final products of graphene oxide dispersion

2.2 Centrifuge

The GO dispersion contains many unsolvable particles, such as Mn_2O_7 and graphite, and unreacted solvent such as hydrochloric acid and hydrogen peroxide. Therefore, it is very necessary to remove those particles and solvents. The specific parameters for centrifuge are in the following table:

Table 2: Settings for centrifuge

Speed	14 500 r/min
Time	20 mins
Repeat times	5

Notice: Between the fourth and fifth centrifuge, add one more centrifuge with a speed of 3000 r/min, and keep the upper dispersion, get rid of the lower dispersion

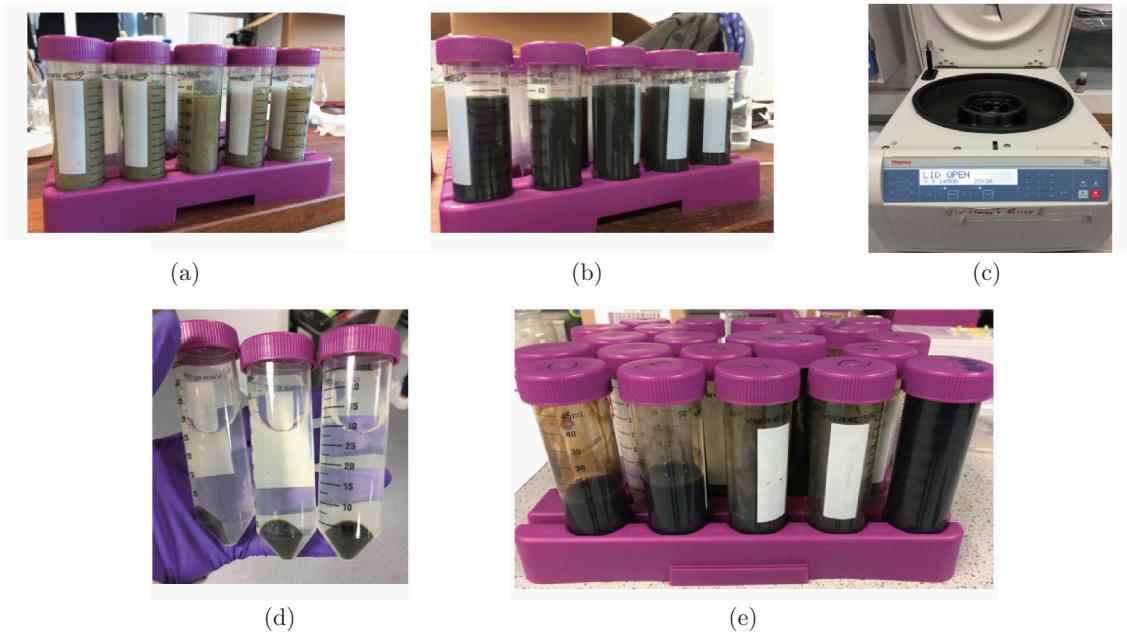


Fig. 2: (a) After first centrifuge; (b) After second centrifuge; (c) Centrifuge settings; (d) After the final centrifuge; (e) Combinations of the same group for GO dispersion

2.3 Wet Spinning

Coagulation bath chosen for this experiment was an ethanol/water mixture with 5%wt CaCl₂ solution added, which has been tested to be effective. The drawing ratio was set to be 1.5 using the apparatus shown below:

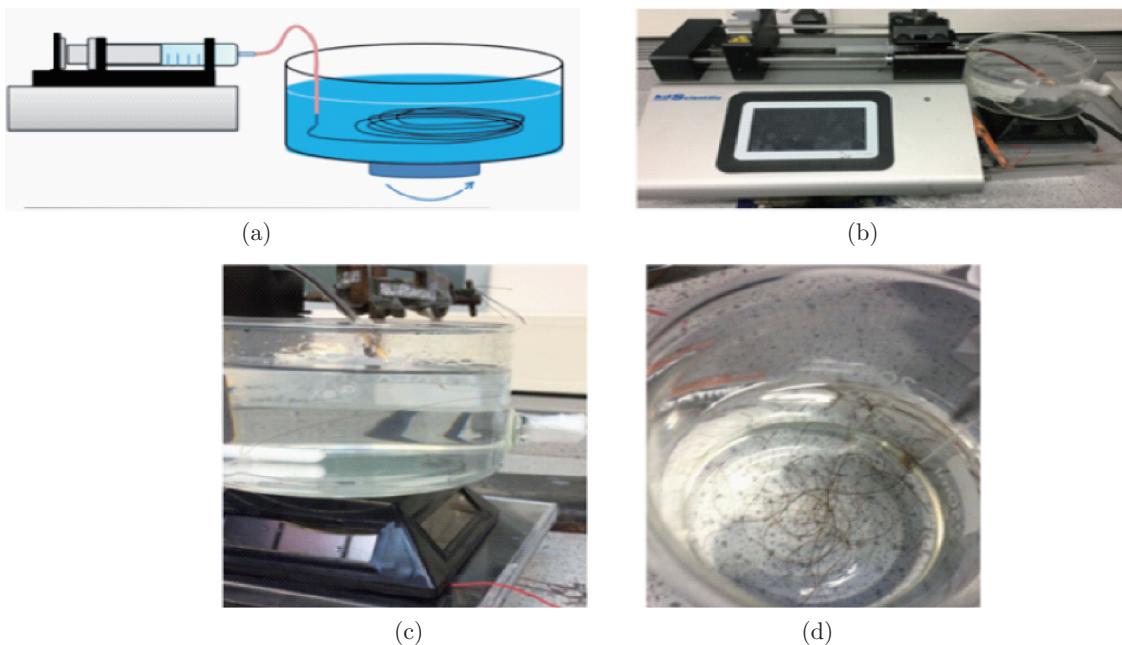


Fig. 3: (a) Design of wet-spinning apparatus; (b) Wet-spinning apparatus in the laboratory; (c) Graphene oxide fibres (GOFs) emanate from needle and are drawn by the rotating plate; (d) GOFs in water

2.4 Reduction of GOFs

Hydroiodic acid was used for the reduction of GOFs. The specific methodology is to immerse GOFs in hydroiodic acid at 90 °C for 8 hours.

3 Results & Discussion

3.1 Graphene Oxide

3.1.1 Mean Size

Measuring for size distribution is under a scanning electron microscope (SEM). Before measurement, the six groups of GO dispersion, which were synthetized under 20, 30 or 40 seconds' heating and 24 or 48 hours' oxidation, should be re-prepared with the same concentration at 100 µg/ml. After the GO was spin-coated to the surface of silicon, images relating to the size distribution of the GO sheet were scanned by SEM.

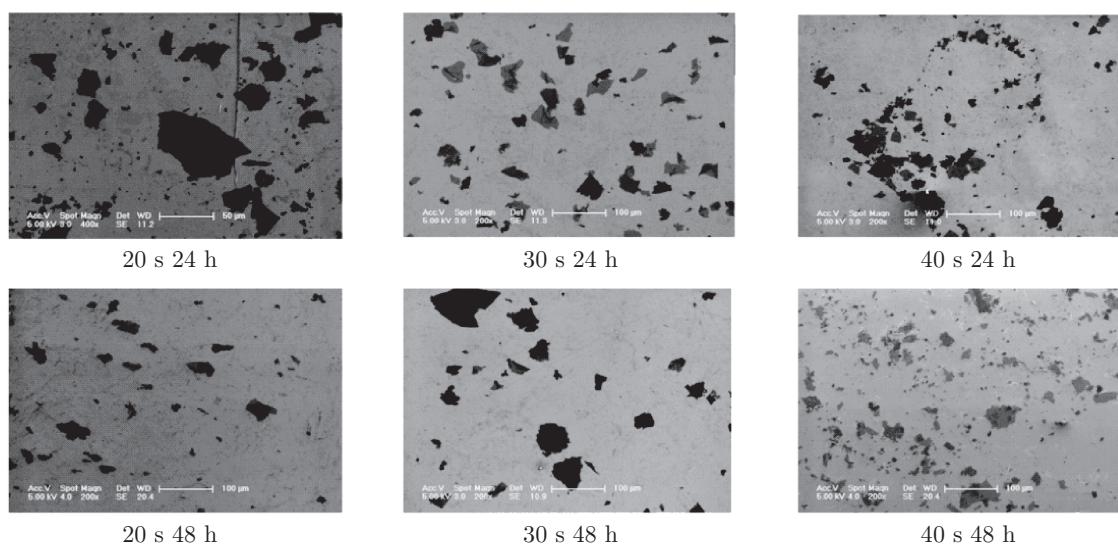


Fig. 4: Images of the GO sheet under different heating and oxidation times by SEM

Each GO sheet size was measured and calculated using ImageJ software. Figure 5 accurately demonstrates the differences of size between groups:

From the column chart in Fig. 5, it is easy to identify that regardless of whether the oxidation time was controlled for 24 hours or 48 hours, sizes of the GO sheets changes in corelation to a change in time while the expandable graphite was being heated. This variation trend was found to follow a normal distribution if more variables for heating time are added such as 10, 25, 35 and 50 seconds. The maximum value occurred when the expandable graphite was heated for 30 seconds. This indicated that 30 seconds' heating may contribute most to the increase of the GO sheets' size, and this influence seems to be significant. However, when the heating time was reduced to 20 seconds or was increased to 40 seconds, the size of the GO sheets decreased dramatically, but remained at almost the same size.

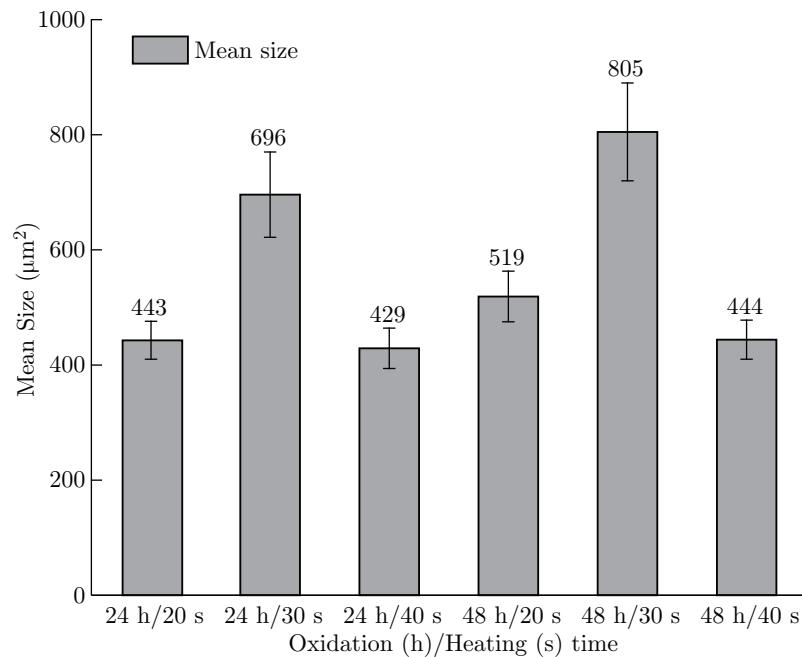


Fig. 5: Mean size of GO sheets in different groups

When the heating time was fixed, the size of GO sheets exhibited a slight growing trend along with the increase in oxidation time. Moreover, aside from the mean size, which exceeded $100 \mu\text{m}^2$ when 30 seconds' heating was employed, the size of the other two groups also showed a slight difference. Therefore, the oxidation time was concluded not to be the key factor that affect the size of the GO sheets when comparing the heating time.

3.1.2 Size Distribution

Since the shape of the GO sheet resembles a circle, the diameter of each particle can thus be calculated. The frequency of the particle-size distribution together with its corresponding SEM images is shown in Fig. 6.

The histograms in Fig. 6 suggest that the size distribution for both GO dispersions synthesized by 30 seconds' heating, 24 hours' or 48 hours' oxidation may follow a normal distribution roughly. However, the group '30 s 24 h' has most particles clustering in diameter intervals [12.5, 17.5], while [17.5, 22.5] are shown for the group '30 s 48 h'. Thus, clustering means an extra 24 hours' oxidation could increase GO sheet size by 5 μm on average in terms of diameters.

3.1.3 Discussion

(1) Data Analysis

Data analysis is assisted by Minitab software.

1) Interval Plot & Analysis of Variance

An interval plot is a graphical summary of the distribution of a sample that shows the sample's central variability and tendency. The default interval plot exhibits a mean symbol with a 95% confidence interval bar. Analysis of variance (ANOVA) tests the hypothesis that the means of two

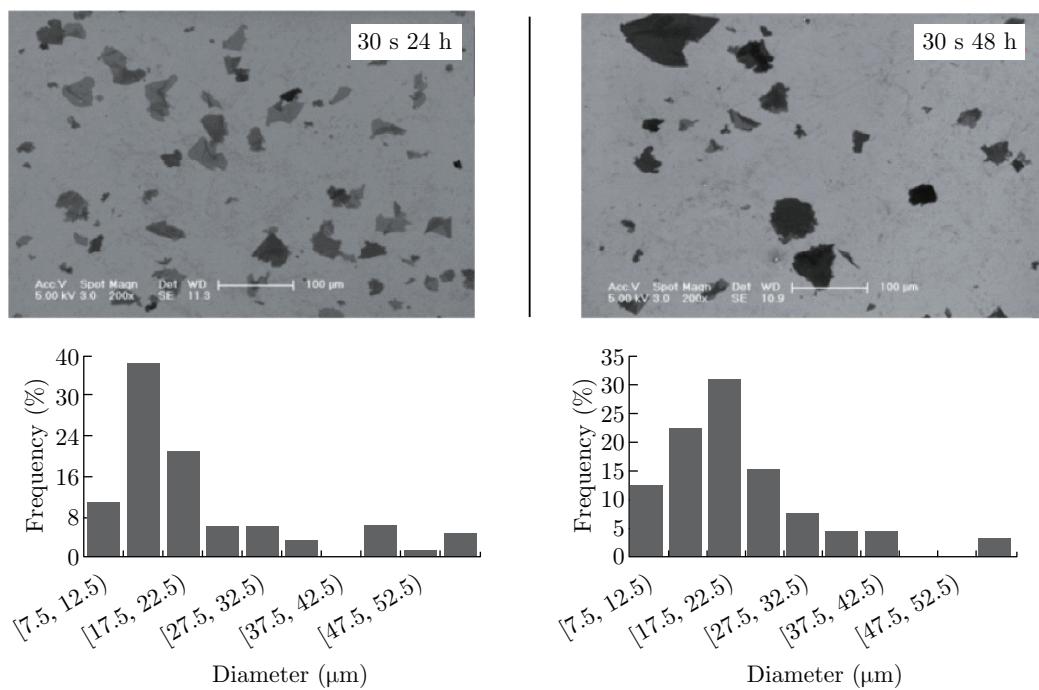


Fig. 6: SEM images and corresponding histogram of GO size distribution (under the SEM image)

or more variables are equal. ANOVA assesses the importance of one or more factors by comparing the means of response variables at the different factor levels. ‘P-value’ is used to examine the significance of investigated variables; if ‘P-value’ is less than 0.05, the corresponding variable can be concluded to have a significant effect on the dependent variable at a 5% significance level. ‘F-value’ is used to determine which factor has a more significant effect. The higher the ‘F-value’, the more significant is its contributory effect.

By examining the P-values circled in Table 3, variable heating time was found to be significant at a 1% level of significance, while the variable oxidation time was found not to be significant at a 5% level of significance. The results suggested that heating time does have a significant effect on the size of GO sheets but oxidation time failed to be concluded to have a significant effect. Based on this conclusion, the remaining article will focus more on the effects of heating time by redesigning and repeating the experiment.

Table 3: Analysis of Variance for Mean Size

Source	F	P
Heating Time	45.18	0.000
Oxidation time	5.28	0.061
Heating*Oxidation Time	0.89	0.460

2) Main Effect Plot & Interaction Plot

Main effect plots in Fig. 7 were used to investigate the differences among the mean sizes affected by heating time and oxidation time respectively. Interaction plot displays the interaction between heating time and oxidation time. A steeper slope suggests a higher significant interaction.

By visual examination of Fig. 7, the mean size of GO sheets was found to increase initially and

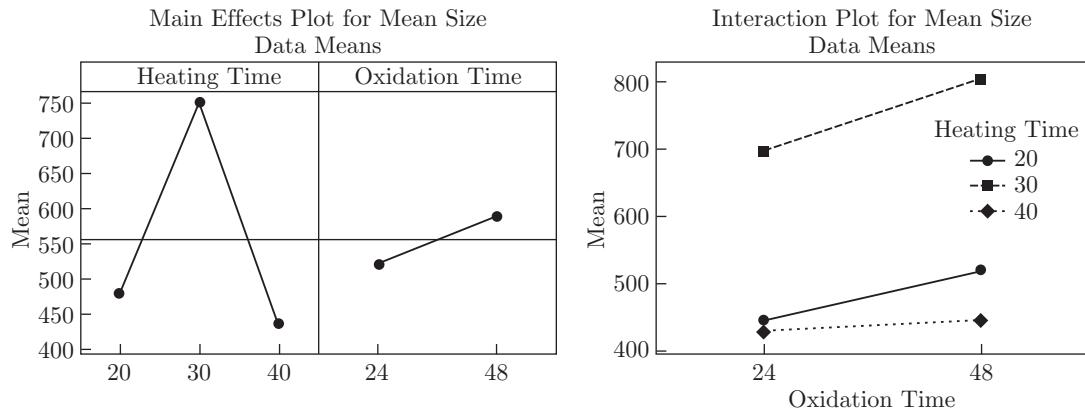


Fig. 7: Main effects plot & interaction effects plot

then decrease when the heating time increased. However, the mean size of GO sheets maintains a continuous increase through extending the oxidation time. The unparallel lines of the interaction plot mean that the interaction between heating time and oxidation time jointly influence the size of GO sheets.

3) Residual Plot Analysis

A residual plot is a graph that is used to examine the goodness-of-fit in regression and ANOVA. The normal probability plot of residuals is used to examine the assumption that the residuals are normally distributed if there is a linear relationship between the residuals and theoretical percentiles. Histograms of residuals are used to determine whether the variance of residuals is normally distributed. The plot of residual versus fits is used to examine the consistency of the variance of the residuals. The plot of residual versus order is employed to examine whether the residuals are independent and identically distributed (IID) or not. The residuals of Mean Size are concluded to be normally distributed and there are no outliers in the data, which indicates that the measured data is relatively reliable.

(2) Mechanism

1) Effect of Microwave Heating

Expandable graphite is a synthesized intercalation compound of graphite that expands or exfoliates when heated. It is manufactured by treating flake graphite with various intercalation reagents that migrate between the graphene layers in a graphite crystal and remain as stable specials. When expandable graphite is heated in the microwave, the rapidly increasing temperature makes these intercalation compounds decompose into gaseous products, resulting in high intergraphene layer pressure. This pressure develops enough force to separate graphite basal planes in the “c” axis direction [11]. Nevertheless, with the continuous increase of heating time in the microwave, the connecting point between the expanded graphite worms have a more concentrated field density, resulting in the electrical gas breakdown of expanded graphite, which decreases the size of expanded graphite; this is called ablation of expanded graphite [12]. On the other hand, overheating oxidizes expanded graphite, leading to a destroyed structure of expanded graphite and subsequently a decrease of size. These mechanisms are believed to be capable to convincingly demonstrate why the mean size of GO sheets increases first before decreasing when heating time continuously increasing.

2) Effect of Oxidation Process

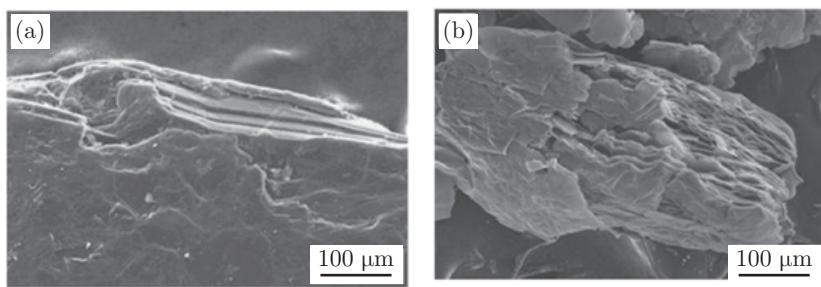


Fig. 8: SEM image of graphite (a), expandable graphite (b)

Based on the modified Hummers' method to synthesize GO dispersion in this experiment, the conversion process of expanded graphite to GO can be identified as three independent steps [10].

The first step is to convert graphite to the sulfuric acid-graphite intercalation compound (H_2SO_4 -GIC), which is regarded as the first intermediate. The formation of H_2SO_4 -GIC begins immediately after graphite is exposed to the acidic oxidizing medium, which makes the graphite more reactive to oxidation without changing the graphene structure. As a result, this step will be completed very abruptly, within 3-5 minutes.

The second step is the conversion process of GIC for the oxidized form of graphite, which is defined as pristine graphite oxide (PGO). The second intermediate is formed at this step. During this process, under strong oxidation of $KMnO_4$ and H_2SO_4 , a substantial number of phenolic groups are initially produced at the edges and then become defect on the basal plane under strong oxidation of $KMnO_4$ and H_2SO_4 . The double bonds are transformed into single bonds simultaneously. With the oxidized process proceeding, two reactions will take place in phenolic groups: most of the phenolic groups will be condensed into forms of C—O—C linkages, while a small number of phenolic groups will be oxidized into quinone groups. Then, ketone groups convert to COOH [13]. This process will last for several hours or even a couple of days depending on the graphite source. Moreover, the oxidation process of carbon atoms in a graphene layer with other specials might lead to a loss of atoms and the creation of defects; with the increase of oxidation time, the defects increase and the electrostatic repulsive force between GO reduces. Finally, there is also a slight increase to the size of GO sheets and this increase is consistent with colloidal theory [15, 16].

The final step is to convert PGO to GO by a reaction between PGO and water.

3.2 Viscosity

3.2.1 Viscosity Measurement

The viscosity of graphene oxide is a measure of its resistance to gradual deformation by sheer stress. This resistance is the kind of force formed by the attraction between particles or nanolayers in GO dispersions. Based on the research on rheological and polarized optical microscopy, a rational relation among GO sheet size, GO viscosity, GO concentration, and GO liquid crystal can be discussed, leading to an understanding of GF's spinnability [17].

GO dispersion is a type of non-Newtonian fluid. This means that its viscosity varies with changes of the shear rate. The viscosity measurement of GO dispersion is under a rheometer.

GO dispersion used for measurement has the same concentration (5 mg/ml) in all 6 groups. The results are shown in Fig. 9 below:

Figure 9(a) indicates that under the same oxidation time of 24 hours, with an increase of heating time from 20 seconds to 40 seconds, the viscosity keeps rising at the same shear rate and this growth rate keeps decreasing when the shear rate accelerates. When the shear rate is greater than 1 S^{-1} , the viscosity will no longer change and will remain at a low value. Figure 9(b) suggests that viscosity of GO dispersion is affected by a variable oxidation time. A negative effect of a relatively higher oxidation time for viscosity of GO dispersion is also detected, in contrast to a higher heating time.

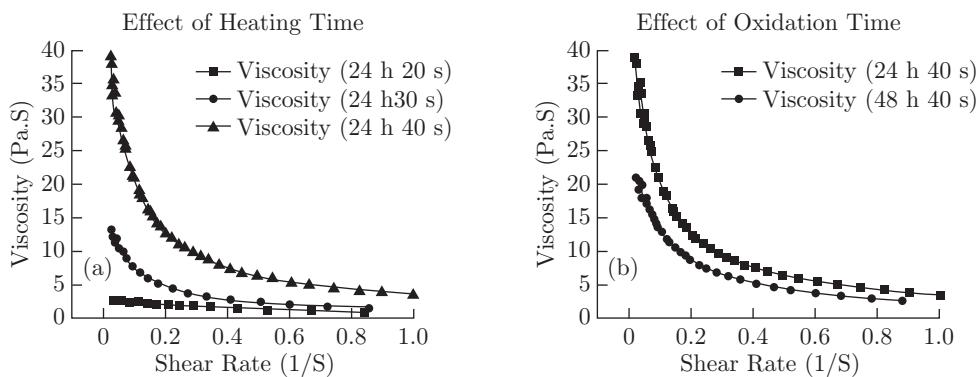


Fig. 9: (a), (b) Viscosity affected by heating time and oxidation time

3.2.2 Discussion

(1) Mechanism of Viscosity

GO dispersion, as a non-Newtonian liquid, has a property of abundant aggregation and rheological behaviour. The aggregation of GO sheets leads to an increase of internal shear stress due to the greater force required to dissipate the aggregated particles in the media, hence to increase viscosity. The aggregation of GO in water contains five sorts of forces including: electrostatic interaction, Van der Waals attraction, hydrogen-bond interaction, π - π stacking, and the collision of water molecules [14]. Furthermore, except for forces, the pH of solution, electrical surface surge and functional groups of GO will impact on aggregation of GO as well. What has been discussed regarding the oxidation of expanded graphite by modified Hummers' method indicates that the GO sheets contain large amounts of hydroxyl and carbonyl functional groups, resulting in a reduction in the electrostatic repulsive force between GO sheets [15]. The aggregation of GO sheets increases internal shear stress, leading to the increase of viscosity. This process can be used to explain how viscosity is affected by the oxidized process of graphite (Fig. 9(a), (b)).

(2) Viscosity & Spinnability

Viscosity refers to the behaviour of graphene oxide liquid crystals (GO LC) as well. The phase transformation of GO LC results in the spinnability of GFs being affected. Rouhollah Jalili found that the spinning of continuous, strong and robust GO fibres could be conducted at concentrations of as low as 0.075wt% through knowledge gained from straightforward formulation of GO LC 'inks' in a series of processable concentrations [17].

There are two prerequisites required for the formation of liquid crystals, which are well-dispersed

in solution and a higher aspect ratio [18]. GO is proved to demonstrate good dispersibility in various solvents. Therefore, a higher aspect ratio is the key to the forming of GO LC. The increasing viscosity of GO dispersion contributes to a higher shear stress, which results in the sliding of lamellae from GO sheets. GO sheets become thinner and thinner as a result of more lamellae sliding out. A thinner GO sheet makes a higher aspect ratio, which leads to GO LC being formed. As a consequence, the alignment of GO LC improves the spinnability of GO dispersion for GOFs.

3.3 Mechanical Property

3.3.1 Tensile Strength & Strain

The theoretical equation known to calculate tensile strength is $\sigma = F_b/S_0$ (F_b denotes the breaking force of graphene fibre and S_0 denotes its cross-sectional area). The equation for strain is $\varepsilon = \Delta L/L \times 100\%$ (ΔL is the length difference and L is the original length of graphene fibre, which is measured as 11 mm).

From the Tensile Strength chart in Fig. 10, it is clearly observed that under the same oxidation time of 24 or 48 hours, the heating time for 30 seconds always has the highest tensile strength and the specific value of strength in the group ‘30 s 24 h’ can reach as high as 24.16 MPa, which is seven times larger than ‘20 s 24 h’. This indicates that the variable heating time of expandable graphite has a huge influence on tensile strength. The overall variation trend is approximately a normal distribution, proving that the heating time of 30 seconds may be the most favorable. But in terms of the effect of oxidation time, the tensile strength has an obvious downtrend with an increase of oxidation, except for a heating time 20 seconds, which increases slightly. This heterogeneity might be the consequence of the interaction of heating and oxidation or unexpected errors in the data. Details will be explained in the discussion part of this article.

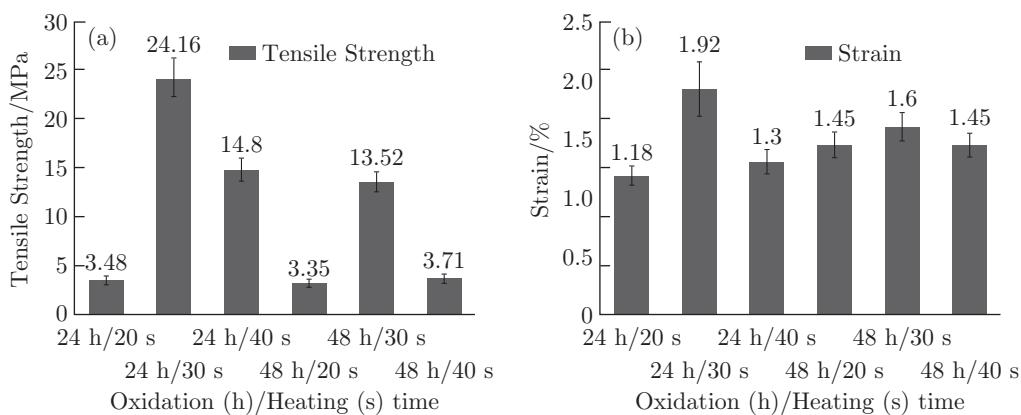


Fig. 10: (a) Tensile strength of graphene fibres, (b) Strain of graphene fibres

The Strain chart indicates that increasing oxidation time contributes to the increase of the strain of the graphene fibre. Combining the results of the Tensile Strength chart and Strain chart; the higher the tensile strength, the lower the strains of GF can be confirmed, hence, GFs can be concluded as an elastic but not perfectly elastic material as it is in according with Hooke's Law at the beginning of stretching but not later on.

3.3.2 Stress-strain Curve & Young's Modulus

When graphene fibres are being stretched under the mechanical test machine, the change of loading and elongating is a continuous process actually and the stress-strain curve shows a continuous progression until the graphene fibre breaks. Figure 11(a) & (b) indicates that the effect of heating under 24 and 48 hours' oxidation. The results are similar to what has been summarised earlier in this article. Fibres with heating for 30 seconds always exhibited a superior mechanical property. Furthermore, mechanical properties were found to be the best when synthetized fibres are under 30 seconds' heating and 24 hours' oxidation. The results of the effects of oxidation under 30 and 40 seconds' heating are presented in Fig. 11(c) & (d). The increase of oxidation time was found to have a negative effect on the mechanical property of GFs.

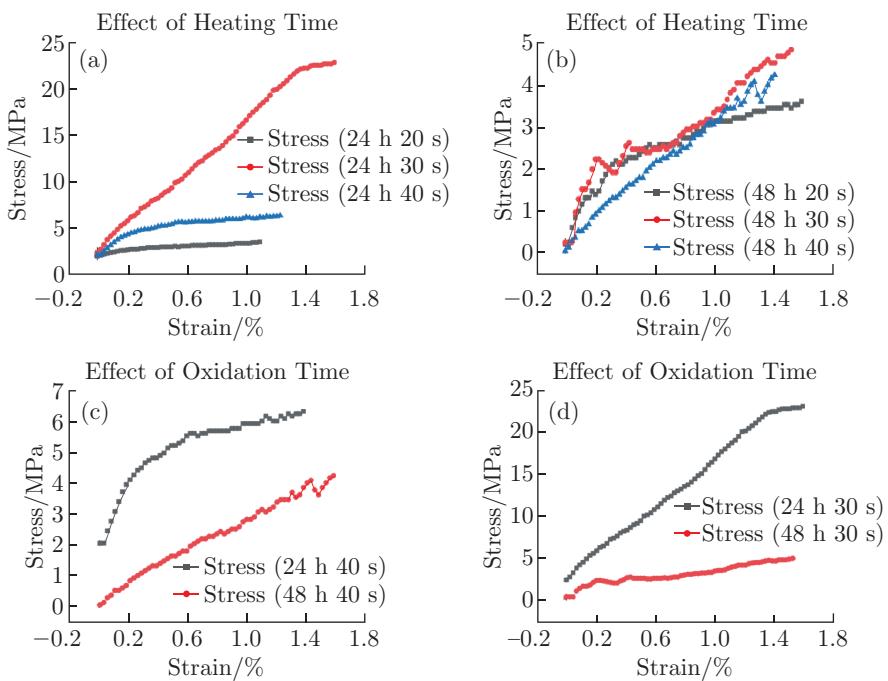


Fig. 11: (a) & (b) Effects of heating time under 24 and 48 hours' oxidation; (c) & (d) Effects of oxidation time under 30 and 40 seconds heating

Young's modulus is the ratio of stress and strain of a material under its elastic limits. Based on the definition, the slope of the stress-strain curve at its linear elastic part is the Young's modulus apparent in Figure 11. And the calculated values are presented in Table 4.

Table 4: Young's modulus

	20 s	24 h	30 s	24 h	40 s	24 h	20 s	48 h	30 s	48 h	40 s	48 h
Young's modulus (GPa)		3.23		4.49		4.06		1.12		1.43		0.44

From the table, fibres synthetized under 24 hours' oxidation exhibit a higher Young's modulus than when synthetized under 48 hours' oxidation. More importantly, 30 seconds of heating shows a better Young's modulus when comparing to 20 and 40 seconds.

3.3.3 Discussion

According to the literature review, various factors dominate the tensile strength of GFs. These factors were concluded as Van der Walls force, hydrogen bonds or chemical bonds between GO sheets. An additional factor is the entanglement of twisted graphene sheets [19]. It is predicted that the displacement of GO nanosheets and stretching of folded nanosheets cause the plastic deformation of GOFs, which weaken the mechanical property of GFs. Considering these factors, three strategies are taken to improve its mechanical property effectively: (1) through introducing high energy bonds to increase the interlayer interaction; (2) reducing structural defects; (3) advancing the regular alignment of graphene paralleled in the fibre axis [8].

(1) GO Sheet Size & Mechanical Property of GFs

As has been discussed above, the ablation of expanded graphite can decrease GO sheet size as the heating time in the microwave keeps rising. This process results in a larger GO sheet size synthesized under 30 seconds' heating in the microwave. The enlarged GO sheets increase the interaction between GO sheets through more chemical bonds and hydrogen bonds created between the GO sheets, which makes GFs with 30 seconds' heating in the microwave during the fabricated process possess the highest tensile strength. On the other hand, with oxidation time added, GO sheets break, thereby generating the structural defect. This will also reduce the mechanical property of GFs.

(2) Wet Spinning & Mechanical Property of GFs

It has been demonstrated by previous discussion in this article that viscosity of GO dispersion relates to its ability of spinning. An alternative factor, coagulation bath, is considered to determine the mechanical property of GFs in the wet-spinning process. In this experiment, CaCl_2 was chosen for the coagulation bath in interest of its superior ability of spinning. The mechanism is that a higher CaCl_2 concentration induced diffusion of CaCl_2 cations and the diffusion accelerated the form cross-linking between GO sheets. When LC GO was squeezed out due to coagulation, GOFs gradually precipitated. Meanwhile, Ca^{2+} from coagulation formulation went into GOFs, generating the forming of cross-links between GO sheets. Thus, the mechanical properties of GOFs were significantly enhanced. However, the cross-links were formed at the early stage of the diffusion. The ultra-high concentration of Ca^{2+} might therefore cause an increase of residuals and not contribute to further enhancement of mechanical properties.

4 Conclusion

By contrasting the measured characteristics of six groups' GO dispersion and GFs, it was found that GO dispersion synthesized by 30 seconds' heating in the microwave has a larger GO sheet size compared with 20 seconds' and 40 seconds' heating. The difference of GO sheet sizes could be explained by the theory of expanded graphite's ablation, which believes that over-expanded graphite going through electrical gas breakdown does result in a decrease on sizes. The increasing sizes of GO sheets also contributes to the interaction between GO Nanosheets with additional hydrogen bonds and chemical bonds being formed. This firm interaction between GO Nanosheets conduces to the enhancement of GF's mechanical properties at macroscopic levels. As a result, GFs synthesized with 30 seconds' heating exhibit superior mechanical properties. Based on the heating time of expandable graphite, it is proved to be a crucial factor to affect the mechanical

property of GFs. On the other hand, the increase of oxidation time does change the functional groups in the GO sheets' basal plane into abundant oxygen functional groups such as carboxyl groups, hydroxyl groups, and epoxide groups. These abundant functional groups weaken the interactions of Van der Waals force between conjugated structures of graphene, leading to the decrease of the mechanical properties of GFs.

Likewise, the wet-spinning process of GO dispersion has a substantial influence on the mechanical properties of GOFs as well. Viscosity is demonstrated to affect the forming of graphene oxide liquid crystals and its phase transformation. In addition, the coagulation bath containing Ca^{2+} generates the cross-links between GO Nanosheets. These two factors are confirmed to be the core points related to the mechanical properties of GFs.

5 Further Study

Based on the study of fabrication of graphene fibres via wet spinning it is concluded that the properties of GO dispersion and the wet-spinning process does affect the mechanical properties of GFs. Prevailing conclusions of a sufficiently large number of researches have been given regarding GO dispersion in literatures on the effect of heating time and oxidation time. Further study for this article could focus on how to improve mechanical properties by optimizing the wet-spinning process of GFs. In wet-spinning, the diameter of needles and flow of GO from the needle will influence the speed of GOFs emanating from the needle, which could affect the drawing ratio of GFs. Most importantly, the selection of coagulation bath will also have a considerable influence on GFs. Hence, the next research priority for the authors is to focus on the wet-spinning process.

Acknowledgement

We are grateful for the support of the EU Horizon 2020 and University of Manchester through projects with project codes 644268-ETEXWELD-H2020-MSCA-RISE-2014, AA14512 (UMRI project “Graphene-Smart Textiles E-Healthcare Network”) and R1119938.s

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