Graphene-Material Based Nanocomposite-Coated Optical Fibres: A Multi-Functional Optical Fibre for Improved Distributed Sensing Performance in Harsh Environment

Kenny Hey Tow, Sandy Alomari, João M. B. Pereira, Tiago Neves, and Åsa Claesson

***Abstract*—The optical fibre coating is essential to ensure high performance and reliability of the optical fibre. Out of all polymer-coated fibres, polyimide coatings provide the highest temperature rating, typically rated for use in optical fibre sensing applications at 300 °C (in air), with short excursion to 350 °C. In this communication, we assess whether the inclusion of graphene-based nanoparticles, such as graphene and graphene oxide, in a polyimide coating can enhance the durability of optical fibres at high temperatures. Draw tower fabrication of optical fibres with nanocomposite polymer coating is described. Tensile strength tests, performed on aged nanocomposite-coated optical fibres, are used as an indication of their performance at harsh conditions. The results are validated and quantified by distributed temperature and humidity sensing tests performed using these fibres. The results show that this novel class of fibre is more robust to high-temperature ageing and moisture-induced strain than standard polyimide-coated fibres, when used for distributed sensing. The electrical conductivity of the nanocomposite coating is also used in a multi-sensing approach, together with distributed optical fibre sensing, to measure temperature in a reliable way using the same optical fibre.**

***Index Terms*—Graphene oxide, high temperature, nanocomposite, optical fibre sensor, specialty optical fibres.**

I. Introduction

Fibre-optic sensing increases its industrial relevance year by year. These sensors are being used extensively in areas such as aerospace, energy, medicine, infrastructure, and production. Many fibre-optic sensor applications make use of specialty optical fibres, i.e., fibres that are customized for the specific use case. The optical fibre coating is an important parameter to consider when designing a custom-made optical fibre. Although the fibre coating can sometimes play an active part of the sensor, in most cases it should be passive with the purpose of providing protection during manufacture, installation, and use of the sensor. Most optical fibres are coated with a polymer material. Due to its cost-effectiveness, flexibility, and ease of application, acrylate coating is the most employed. When the fibre-optic sensor is to be installed in harsh settings, it is common to use polyimide-coated optical fibres, for their durability at high temperatures and in other challenging environments. Polyimide coatings provide excellent long-term protection to fibres up to 300 °C in air for long period of time in harsh environment applications (shorter term to 350 °C) [1], [2], [3]. Yet, there are many use-cases where long-term, higher temperature monitoring of processes and assets using distributed fibre-optic sensing techniques would be beneficial [4], [5], [6], [7]. In these specific cases, fibres with metal coatings such as aluminum or gold, which can withstand higher temperatures, are sometimes used. However, metal coated fibres are much more expensive than fibres with more conventional coatings, more difficult to fabricate at long lengths and they exhibit non-linear differential attenuation [8], making them difficult to use for kilometre-long distributed temperature measurements. Combining the unique mechanical, optical, electrical, chemical and morphological properties of graphene-based materials with the benefits of optical fibre sensing schemes is attracting growing interest in the sensing community. However, integrating these materials into long lengths of silica fibres leads to significant transmission losses of the order of a few dB/cm [9], which is incompatible with distributed optical fibre sensing.

In this work, the graphene-based material is included in the optical fibre coating instead. In particular, we focus on polymer coatings enhanced by the introduction of nanoparticles in the polymer, so-called nanocomposite (NC) coatings. One early publication [10] reports that a nanocomposite combining graphene oxide and polyimide greatly can enhance the polyimide performance by increasing its electrical conductivity by 14 orders of magnitude, reducing the oxygen transmission rate by 93%, and increasing the Young's modulus of polyimide by almost 300%. Nanocomposite coating with graphene oxide and polyimide has also been reported to slow down the weight loss in bulk polyimide components at high temperatures [11], usually attributed to the oxygen diffusion barrier properties

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of the nanocomposite. Many nanocomposite-based optical fibre sensors have been reported in the literature for measuring humidity, pH, temperature, ions, etc. [12]. Most of them involve complex fabrication procedures (etching/tapering, stripping/recoating), or exhibit cross-sensitivities to other parameters (temperature, humidity); thereby limiting their practical implementations. Moreover, these sensors are typically single point sensors, and difficult, if not impossible, to multiplex along one optical fibre. To fully unlock the potential of NC coated optical fibres for distributed fibre sensing applications and show their commercial relevance, access to kilometre lengths of these fibres with uniform coatings are required. A continuous process for coating long lengths of optical fibres with a mixture of graphene-acrylate (30%-wt) was reported [13], using jet spray. The authors reported that, when used as temperature sensors, these fibres demonstrated fast thermal response, with thermal diffusivity 30-fold higher in the axial direction than a conventional acrylate coated silica fibre.

More recently, the fabrication of long lengths of nanocomposite-coated fibres using conventional fibre draw tower equipment was reported [14]. In this paper, we present the developed process for manufacturing long lengths of these nanocomposite-coated fibres. The mechanical strength and durability of these NC fibres after thermal ageing are discussed in Section III. As previously demonstrated in [14], the inclusion of graphene material nanoparticles to the polyimide coating improves the sensing performance of the fibre in terms of lower optical loss on ageing, and improved accuracy in the sensing data. Two examples of using these novel optical fibres for distributed sensing in harsh environments (high temperature and moisture setting) are presented in Section IV. In addition, this coating confers additional properties such as enhanced electrical conductivity of the coating material. The potential of exploiting this parameter for sensing applications is covered in Section V. The commercial relevance of developing these fibres and future applications with these fibres are discussed in the last section.

II. Nanocomposite Precursor and Optical Fibre Fabrication and Testing

A. *Preparation of PI and PI/rGO Nanocomposite Films and Their Oxygen Transmission Rate*

Polyimide (PI) based nanocomposite (NC) solutions with different contents of reduced graphene oxide (rGO) were prepared, using ultrasonic stirrer to disperse the nanoparticles into the polymer precursor. N-methyl-2-pyrrolidone (NMP) solvent was added to adjust the viscosity of the mix when needed. Using this technique, three precursors were prepared with 0, 2.5 and 5 wt% rGO in polyimide. Samples of these precursors were cast onto commercial Kapton films using a film applicator (the doctor blade technique). The films were dried and cured using 15 °C/min ramp up to 400 °C under nitrogen atmosphere. The final thicknesses of the films were measured to be $\sim$11 μm.

First, the barrier properties of these films against oxygen diffusion were measured. The oxygen barrier property of each film was determined using OX-Tran 2/21 (SL) and Ox-Tran 2/22 10x instruments, at room temperature. The results are plotted on Fig. 1. As expected, the oxygen transmission rate (OTR) decreases with increasing the concentration of rGO, with the 5 wt% rGO film having a 37% reduction of the transmission rate compared to pure PI - a significant barrier performance, in line with prior work [10]. To be fully relevant to the further experiments presented in this paper, the OTR measurements should be repeated at higher temperature (350–450 °C), out of range for our current equipment.

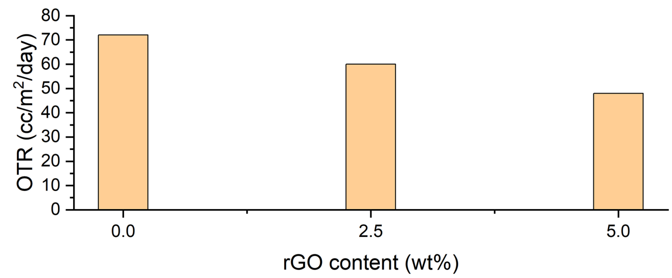

Fig. 1. Oxygen transmission rates (OTR) of pure PI and rGO/PI films with various loadings of rGO at room temperature.

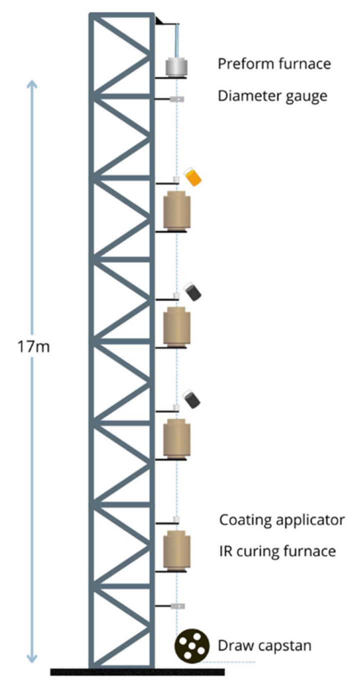

Fig. 2. Schematic picture of the optical fibre draw tower and nano-composite coating process.

B. *Fabrication of the Nanocomposite-Coated Optical Fibres*

Nanocomposite-coated optical fibres were manufactured in a 17-m high optical fibre draw tower equipped with thermal furnaces for curing the polyimide and NC coatings (Fig. 2). NC mixtures were applied via nozzles in a continuous process during fibre drawing. Several hundreds of metres of fibres were drawn for each sample, the length limited by the availability of the mixed NC precursor.

Two fibre designs were manufactured, a reference fibre with three layers of pure polyimide coating (ref) and a NC coated

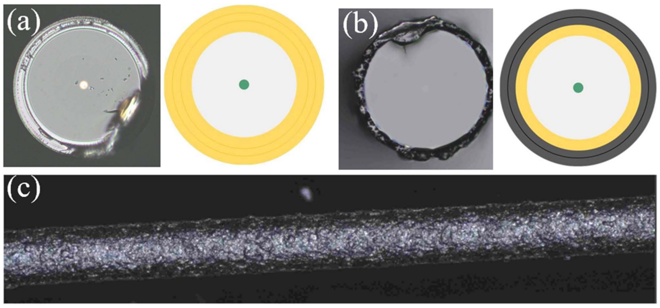

Fig. 3. Adapted from [14]. Design and cross-section of: (a) reference fibre coated with three layers of pure polyimide (left), and (b) NC coated fibre consisting of two NC layers on top of one polyimide layer (right). (c) Side view of a drawn 5 wt% rGO/PI fibre.

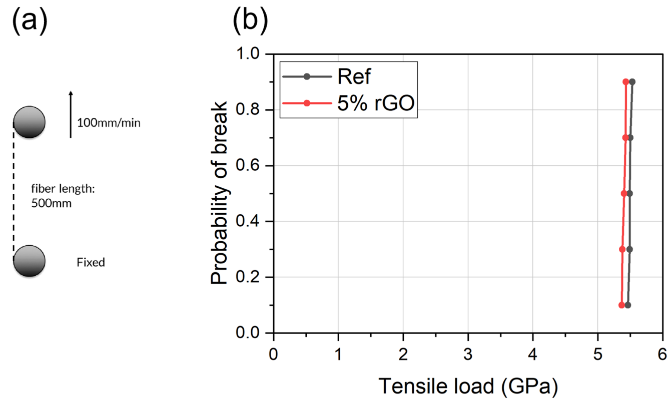

Fig. 4. (a) Schematic picture of the tensile strength test setup, (b) Weibull probability plots of the tested fibre.

fibre, respectively Fig. 3(a) and (b), with one pure layer and two NC layers. The innermost layer of the NC coated fibre is pure polyimide to assure that the glass is not damaged by the rGO material.

Fig. 3(c) shows a side view of the NC coated fibre, demonstrating the typical surface roughness of the 5wt% NC coated fibre. The attenuation losses of different samples of NC-multimode fibres were measured using a commercial OTDR. At 1300 nm, attenuation losses of around 3 dB/km were measured, which is slightly higher than that of commercially available polyimide-coated multimode optical fibres, specified to be ≤ 2 dB/km. This higher attenuation can probably be accounted to the presence of more microbends, created by the clustering of the nanoparticles in the coating material. However, this value is still quite low as compared to metal coated multimode fibres ($\sim$10 dB/km). In addition to NC coated fibres drawn from preform, NC coated fibres and fibre Bragg gratings were also manufactured by up-coating polyimide-coated fibres with NC coating layers. Standard test procedures, performed after the drawing process, showed that the NC coating did not influence the as-drawn optical properties of the fibres.

III. Tensile Strength Test Before and After Ageing

Tensile strength tests of the as-drawn PI and 5% rGO/PI (NC) coated fibres were performed at room temperature using a Lloyd LS2.5 system, adapted for linear strength tests of optical fibres, illustrated on Fig. 4(a). The gauge length was 500mm and the tension rate 100 mm/min. To pass the tensile test, the Weibull distribution of the break strength should show the break probability in a close-to-vertical line, indicating a consistent strength that remain unchanged from one sample to another [15]. The results, plotted on Fig. 4(b), show that the reference and the NC coated fibre are both strong, with a minor difference in break strength between the two fibres.

To test the durability of the fibres, samples were baked in air and their tensile strength tested after ageing. After baking for 100 hours at 300 °C in air, which is the temperature that the PI-coated fibres marketed for, neither of the fibres showed any significant strength degradation. New samples were then subjected to higher temperatures for different durations, as listed in Table I. Resulting Weibull probability plots for this series of tests (tests 2–9) are shown in Fig. 5. The latter clearly shows a different ageing behaviour of the NC coated fibre, compared to the pure PI coated fibre at these ageing conditions. In particular, the NC coated fibre shows significantly higher strength compared to the reference after ageing at: (i) 370 °C after 67.5h, Fig. 5(a); (ii) 400 °C up to 20 hours, Fig. 5(b) and (c), and (iii) 430 °C after 4.5h.

TABLE I
Ageing Parameters (Temperature and Time) for the Samples Used in the Tensile Strength Test With Fibre Ageing

Test	Furnace temperature	Time
1	300°C	100 hours
2	370°C	67.5 hours
3	400°C	4.5 hours
4		8 hours
5		20 hours
6		25 hours
7		44 hours
8	430°C	4.5 hours
9	465°C	4.5 hours

These results show that the lifetime of polyimide-coated fibres can be enhanced by introducing graphene nanoparticles in the coating. Work is ongoing to determine whether this improvement can be attributed to the lower OTR values of the NC coating and/or due to the enhanced material properties and rigidity of the PI coating due to nanoparticles.

IV. Improving Sensing Performance of Optical Fibres With Nanocomposite Coatings

This increased durability at higher temperatures can be directly exploited in the development of optical fibre sensors for harsh environments. Many distributed temperature sensing systems are designed for operation up to 300–350 °C, mainly limited by the temperature rating of PI-coated fibres. However, high-temperature monitoring of up to around 450 °C is of high interest to industry, as are ways to avoid the high cost and practical issues associated with metal-coated fibres.

When undergoing thermal degradation, the polyimide coating exerts microbends to the fibre, inducing so called "microbend

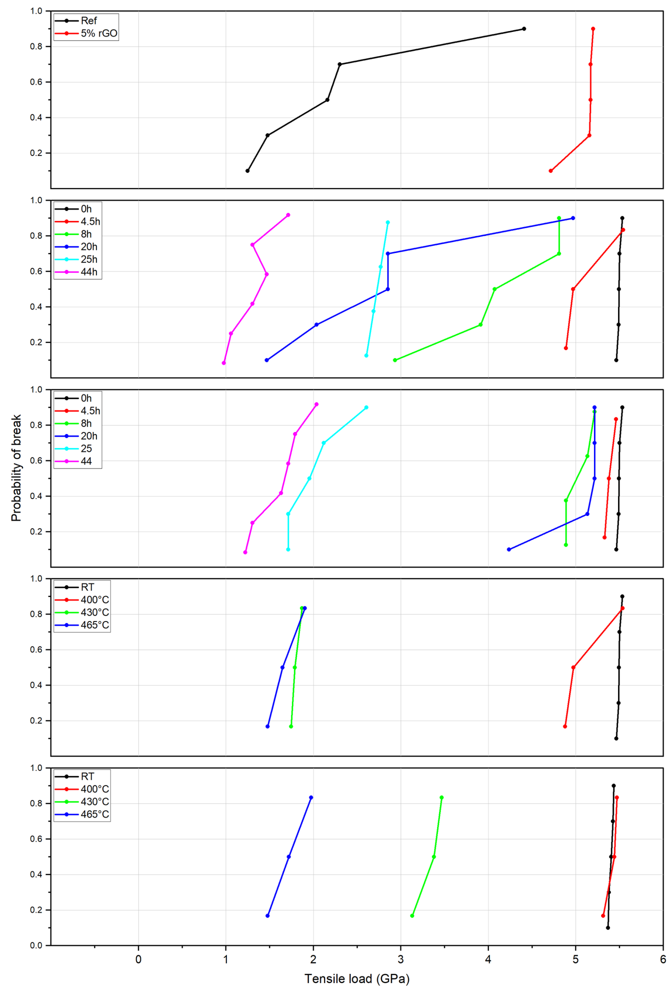

Fig. 5. Weibull probability plots of the tested fibres under different ageing conditions (a) ref (PI) and 5% wt rGO/PI fibre after ageing at 370 °C for 67.5h (b) ref (PI) fibre after ageing at 400 °C for 4.5-44 hours (c) 5% wt rGO/PI fibre after ageing at 400 °C for 4.5-44 hours (d) ref (PI) fibre after ageing 4.5 hours in air at three different temperatures (e) 5% wt rGO/PI fibre after ageing 4.5 hours in air at three different temperatures.

losses," which increase until the coating is completely removed from the glass. During this process, temperature measurement accuracy of distributed temperature sensor (DTS) systems, based on Raman scattering, might be adversely affected due to the unpredictability nature of this varying loss. Indeed, most commercial devices require optical loss budgets less than 10 dB to provide correct measurements. Hence, it is important to make sure that the additional fibre losses, brought by microbend losses along the whole length of the fibre, do not exceed this optical loss budget to ensure a sufficient signal-to-noise ratio for reliable high-temperature measurement.

A. Increasing the Sensing Performance of a Polyimide-Coated Multimode Fibre During High-Temperature Measurement

An experiment, illustrated in Fig. 6(a), was designed to evaluate if the inclusion of NC in the fibre coating could make a standard polyimide coated fibres more robust against microbend-induced optical losses during ageing [14]. The

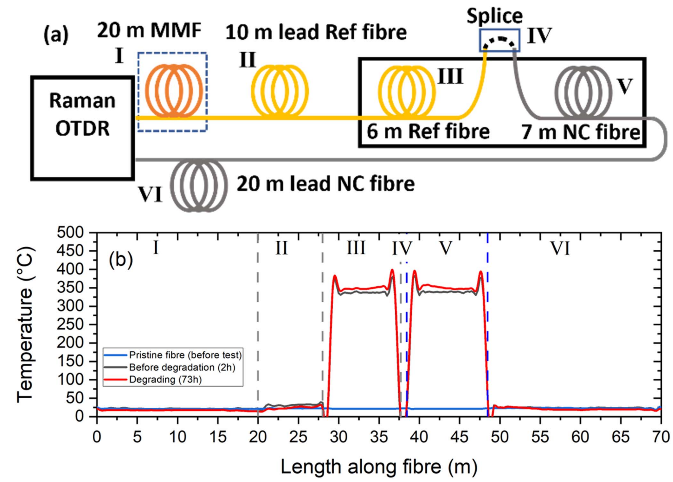

Fig. 6. Adapted from [14]. (a) Representation of the different fibre arrangement used for the ageing test. (b) Sample temperature measurements with this fibre arrangement in the Sections I, II, III, IV, V, and VI as described in the text.

fibres tested are standard graded-index 50/125 multimode fibres (MMF) with polyimide coating (Ref fibre) and the same fibre type up-coated with two thin layers of NC (5 wt% rGO) coating, referred to NC fibre in this section. A commercial Raman Optical-Time Domain Reflectometry (R-OTDR) interrogator, operating at 1064 nm, was connected to: (I) a 20 m MMF fibre, used for room-temperature referencing, (II) spliced to our reference pure PI-coated MMF (Ref). The last 6-m section of this fibre (III) was spliced (IV) to the last 7-metre section of the NC MMF (V). (III) and (V) were placed in a furnace. The rest of the NC fibre (VI) was connected to another channel of the R-OTDR to allow for both single and dual-ended measurements. A thermocouple was placed next to the Sections III and V for sensor calibration.

Sample temperature measurements, obtained from the fibre segments (I)-(VI) are shown in Fig. 6(b). The 2 hotspots – segments (III) and (V) – are clearly visible for a measurement taken at 350 °C. An example of inaccurate temperature measurements due to fibre degradation, also taken at 350 °C but after a few days of thermal ageing, is also illustrated for comparison.

To assess the potential impact that the NC coating has on the microbend losses, the optical losses at 1064 nm (the pump wavelength) of the 2 fibres under test were monitored in segments (III) and (V) during thermal ageing. The temperature was cycled 4 times over one week, 1 time to 400 °C and 3 times to 450 °C, and temperature measurements were acquired every 4 minutes, with a sampling interval of 25 cm in dual-ended configuration. This measurement technique mitigates the effect of differential attenuation along the fibre length, resulting in better accuracy in the temperature measurement. No additional signal processing was applied to the data to get a fair assessment of the measurement performance brought by the nanocomposite coatings to the fibre. The evolution of the accumulated losses over the first 5 metres ($\Delta\alpha$) of the two test fibres, measured by OTDR at the pump wavelength, is plotted on Fig. 7(a). As expected, $\Delta\alpha$ varied over time since residual strain and microbends along the fibres changed while the coating was gradually burning away.

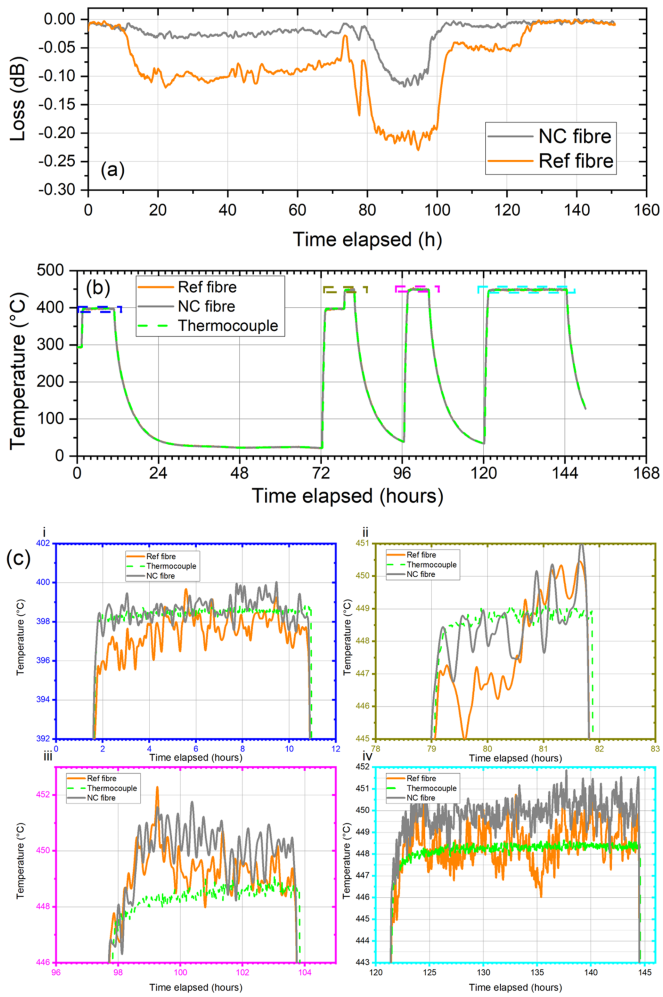

Fig. 7. Adapted from [14]. Evolution of the (a) optical losses over the first 5 metres, (b) temperature measurements, and (c) zoom on selected regions as measured with temperature cycling over 1 week for the reference and NC fibres.

TABLE II
LOSSES OVER 5 METRES ($\Delta\alpha$) OF FIBRE SAMPLE AND LONG-TERM ACCURACY OF THE TEMPERATURE MEASUREMENT DURING AGEING OF THE OPTICAL FIBRES

Cycle (Set T)	Losses		Mean temperature / *Standard deviation*		
	$\Delta\alpha_{Ref}$	$\Delta\alpha_{NC}$	*TC*	Ref	NC
Initial			22.1 (N/A)		
1 (400°C)	0.01	0.01	398.5/*0.207*	397.4/*1.24*	398.5/*0.886*
2 (450°C)	0.02	0.15	448.5/*0.904*	447.8/*1.77*	448.5/*1.17*
3 (450°C)	0.02	0.12	448.4/*0.324*	449.2/*1.62*	449.9/*1.65*
4 (450°C)	0.01	0.01	448.3/*0.243*	449.8/*1.45*	448.2/*1.30*

An increase in the loss of the reference fibre $\Delta\alpha_{\mathrm{Ref}}$ (Table II) was already observed in the first 12 hours, while the transmission of the NC fibre remained almost unchanged. Notable changes in attenuation of the NC fibre were only observed once increasing the temperature to 450 °C, at approximately 80 hours into the cycling. After 96 hours, the optical loss of the reference fibre was double that of the NC coated sample. At around 100 hours, the losses of both fibres decreased, indicating that the coating was removed from the fibres.

For a more thorough study, the temperature evolution at position 34 m (Ref) and 45m (NC) were plotted on Fig. 7(b). Due to the varying optical losses described above, the long-term temperature measurement accuracy deteriorated with cycles as shown in Fig. 7(c). For each cycle, the mean and standard deviation was calculated in the high-temperature zone and compared to the data from the thermocouple in Table II. The presence of nanoparticles in the coating mitigated the impact of differential attenuation with time, indicating that the use of NC coated fibres can increase the lifetime of the fibre and improve measurement accuracy in high temperature environment during the time the coating is degrading.

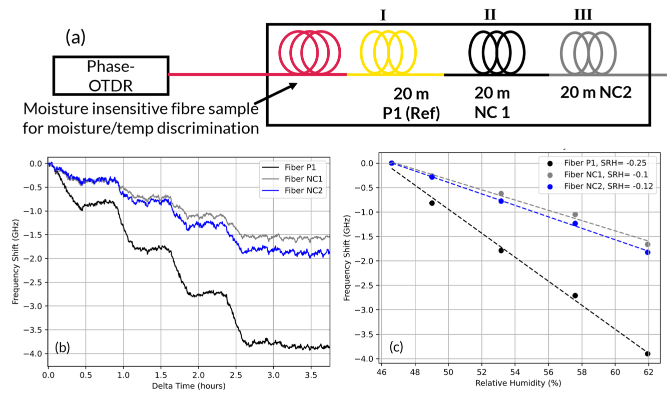

Fig. 8. Adapted from [14]. (a) Experimental scheme used to assess the moisture sensitivity of NC coated fibres. (b) Measured frequency shift corresponding to changes in RH levels and (c) humidity sensitivity for the tested fibres.

B. Reducing the Moisture Sensitivity of a Polyimide-Coated Fibre

The moisture sensitivity of polyimide is often exploited in fibre-optic humidity sensors that link humidity level to strain induced on a fibre by the swelling of the polyimide coating. This moisture sensitivity can be problematic when a polyimide coated fibre is used for monitoring small temperature changes using, e.g., fibre Bragg grating sensors. In such cases, it is crucial to decouple the measurands to ensure accurate and reliable measurements [16].

An experiment, represented on Fig. 8(a), was designed to assess if the NC coated fibres could reduce the humidity-induced strain "felt" by a polyimide coated fibre (P1, reference). The fibres used in this experiment were standard single-mode fibres. Two NC coated fibres were prepared (NC1 (5wt% rGO) and NC2 (2.5wt% rGO)). 20 metres each of P1, NC1 and NC2 were spliced together and placed on a stress-free mount in a humidity chamber. The chamber was kept at a constant temperature of 27 °C, with relative humidity (RH) level changing from 46-62% RH in four steps. A moisture insensitive fibre sample, coated with a hydrophobic silicone was used as the "pure" temperature reference to discriminate temperature changes from humidity or other environmental changes (e.g., residual strain) during the measurement [17]. The resulting frequency shifts of the fibre samples were measured by φ-OTDR. Using the frequency shift of the "pure" temperature reference, the temporal response of the three tested fibres exclusively due to varying humidity levels is plotted in Fig. 8(b). The corresponding humidity sensitivity of these fibres were determined and plotted in Fig. 8(c), as reported in [14].

As expected, the three fibres tested show different sensitivity to relative humidity (SRH). It is clear from Fig. 8(c) that the humidity-induced strain induced on the NC fibre is reduced by at least a factor 2 as compared to a conventional PI-coated fibre. This feature can have direct use in the design of high temperature PI-coated FBG sensors with reduced moisture sensitivity. Further investigation is under way to determine if the reduced moisture sensitivity was caused by an increased stiffness of the coating or less moisture absorption due to the presence of the NC in the coating.

V. Using the Electrical Conductivity of the Nanocomposite Coating for Fibre-Optic Sensing

Electrical conductivity is an interesting property for an optical fibre or its coating to possess since it allows the measurement of electrical parameters (resistivity, current, capacitance). To do so, one could co-draw metals along silica fibres, but this is very challenging due to the limited selection of material compatible with melting temperatures compatible with silica. A metallic coating can instead be used, with the caveat of having a heavier and more bulky fibre-based system since larger bending radii would be required to reduce optical losses in metal-coated fibres. This could also be detrimental for sensing applications, e.g., in avionics, requiring in-situ health monitoring of composite structures using multifunctional sensors [18]. Carbon-coated fibres are also a good option. This thin 20 nm carbon film is usually added between the fibre cladding and the external coating to protect the optical fibre against the diffusion of small molecules into the glass to protect its mechanical integrity when used in harsh environment and oil & gas applications [19], [20]. Moreover, the carbon coating is an electrically conductive medium that can be used to modify the refractive index of long lengths of fibres in a controlled way by applying a current on it [21]. To do so, the external acrylate or polyimide coating needs to be removed to contact the carbon coating, which compromises the strength of the fibre and, subsequently, the fibre component.

The NC fibre has a core that can carry light and a coating containing conductive reduced graphene oxide that can conduct electricity [22] along the whole length of the fibre. Measurement of conductivity along a section is obtained by directly contacting an electrical wire on the section of the coating under test. An electrical resistance of $\sim$ 11 kΩ/cm was measured for our NC fibre at room temperature.

This electrical conductivity can be used to heat the optical fibre in a controlled way. Two FBGs were coated with the same layers as described in Section II: one with the reference PI coating (Ref FBG) and the other one with the 5 wt% rGO NC coating (NC FBG). Current was applied through the coating along a 14 cm section of the NC fibre to heat the fibre. The wavelength shifts of both FBGs due to Joule effect were measured using an interrogator, with a sampling rate 1 kHz and frequency resolution 0.1 pm. As expected, the resulting temperature change as a function of electrical power applied to the fibre coating follows a linear dependence, as shown in Fig. 9. It is worth noting that less than 350 mW electrical power was needed to heat up the FBG, so this 14 cm sample of the fibre, from room temperature to 70 °C, which demonstrates the high potential of using the NC fibre as a current sensor or to use the coating to heat long lengths of fibre. This offers the possibility for developing compact distributed fibre sensing systems in which active heating is required, for instance for measuring wind, flow speeds. [23], [24].

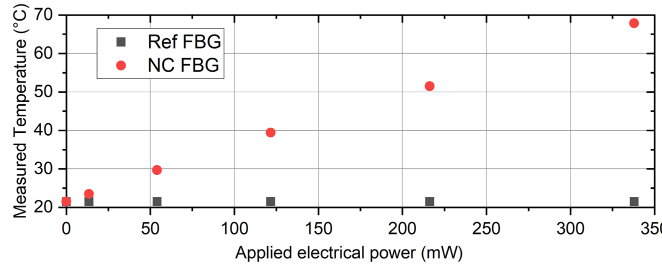

Fig. 9. Temperature evolution of the 14 cm section of the NC fibre with applied electrical power.

Furthermore, the alterations of the electrical resistance of the NC coating, due to local changes in the coating's vicinity, can be measured by directly contacting the coating to a wire along the desired sensing zone. For instance, the temperature dependent resistivity of reduced graphene oxide, reported in films [25], can be used as a sensor. This measurement can be associated to distributed fibre-optic sensing techniques for multi-parameter and multi-sensing approaches using only one optical fibre. Combining both sensing techniques for temperature monitoring using the same optical fibre is also attractive since it increases the data reliability, especially in harsh conditions or during embedding of fibre sensors.

The multimode NC fibre, described in the previous section, was used to test the feasibility of the proposed multi-sensing approach. As pictured in Fig. 10(a), a 1.2-metre-long sample of the NC fibre (hotspot) was placed in a temperature-controlled box and the temperature of the fibre monitored using a Raman-OTDR every 30 seconds, referred to DTS value. The resistance along the NC coating of this fibre sample was simultaneously recorded at 1 Hz. The sample was fixed in a strain-free way to prevent cross-sensitivity to other parameters, such as residual strain or bending, on the resistance measurement. At room temperature, the resistance R_0 was 1.42 MΩ.

First, gradual step increases in temperature from 20–60 °C were set and the measurements taken at steady states. The relative change in resistance, R/R_0, was plotted for several temperatures at steady state on Fig. 10(b), showing a decreasing resistance with increasing temperature as expected [26]. Then, a temperature cycle during which the fibre is heated up to 60 °C (heating phase), then allowed to naturally cool down (cooling phase) to room temperature was set. The resistance value at a given DTS measurement is determined as the average of the last 30 measurements for the values to match. Since the heating process is faster, DTS recordings were performed

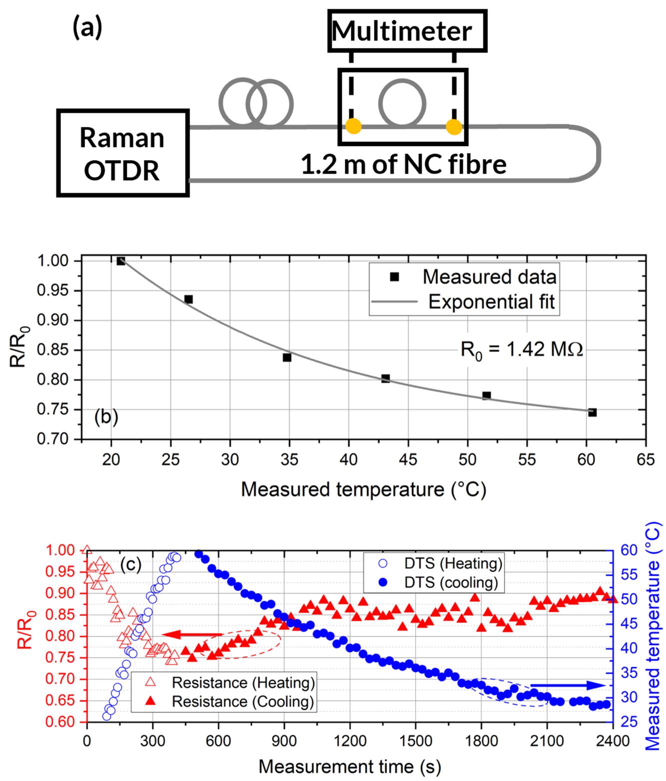

Fig. 10. (a) Experimental scheme to measure temperature both using the optical fibre and the NC coating. Measured relative change in resistance and DTS temperature at (b) steady state and (c) during a heating/cooling temperature cycle.

every 10 seconds during this phase to get a sufficient amount of measurement points. The corresponding resistance-temperature (R-T) plot from the temperature cycle is plotted in Fig. 10(c). As observed, the heat induced response of the resistance of the NC coating followed the same trend as the measured temperature obtained by DTS – the relative change in resistance (R/R_0) decreased to around 0.75 (∼ 60 °C) and gradually increased back to 0.89 (∼ 29 °C, extrapolated from Fig. 10(b)) during the cooling phase. These values are in agreement with the DTS measurements. Hence, the NC fibre can be potentially used to monitor temperature changes during a manufacturing process using two different measurement techniques, with the proviso that the NC fibre is not bent or does not experience residual strain during the measurements.

VI. Outlook and Future Work

Nowadays, optical fibre sensors with nanoparticles are mainly limited to point sensing interrogation schemes, such as interferometry, fibre Bragg gratings, light intensity modulation or fluorescence. Thanks to the simple draw tower fabrication of optical fibres with nanocomposite polymer coating developed in this work, the exceptional electrical, mechanical, thermal and chemical properties of graphene-based materials can now be more easily integrated onto kilometer-long lengths of optical fibres, enabling their deployment in fully distributed fibre-optic sensing systems. Introducing this novel class of NC coating materials to specialty optical fibres and fibre-optic sensing opens many interesting topics for research.

For this work, we mainly focused on evaluating the improvement of the mechanical strength and enhanced sensing performance of a conventional fibre, brought by the inclusion of a small quantity reduced graphene oxide (up to 5 wt%) in its coating, when deployed in two harsh settings (high temperature and moisture). The durability of the NC coated fibres was evaluated by comparing its tensile strength to that of a conventional polyimide-coated fibre after temperature ageing of up to 465 °C. It was shown that the lifetime of polyimide-coated fibres can be enhanced by introducing graphene nanoparticles in the coating.

This increase in durability at higher temperatures can be directly exploited in the development of optical fibre sensors for harsh environments. The feasibility of using NC coated multimode fibres in distributed temperature sensing systems at elevated temperatures was assessed. The results clearly show that the induced microbend losses due to degradation of the PI coating at high temperatures can be mitigated by introducing rGO to the coating, probably due to the higher rigidity of the NC coating compared to pure PI. This results in more reliable measurements using the NC coated fibres since the reduced microbend losses ensures a higher signal to noise ratio in distributed temperature measurements.

When deploying distributed fibre-sensing systems in industrial setting, the entire length of the optical fibre is often encapsulated in a thin-walled metal tube typically made of steel, so-called FIMT (fibre in metal tube) before installation. This confers additional chemical and mechanical protection to the fibre as well as facilitates the installation process of the latter onto the monitored asset. For high temperature measurements, there is an added failure mode to the fibre-optic sensors in FIMTs due to abrasion or adhesion of the fibre to the tube wall. Subsequent cooling of the FIMT may lead to increased attenuation and mechanical tension, leading to temperature inaccuracies in the distributed measurement, ultimately, fibre-optic breaks inside the metal tube. We are currently evaluating the reliability of using the NC fibres, inserted in FIMTs, for temperature monitoring for future field test deployment. So far, the added mechanical strength conferred to the fibre by the NC coating helped keep the integrity of the fibre after several cycling tests, despite increased strain that are induced on the fibre when heated in FIMTs, which generally leads to optical fibre breaks.

The NC coated fibres also show significantly less induced strain by moisture. Tests performed in a climatic chamber on two single-mode fibre samples, with different NC concentration coatings, showed at least a factor 2 lower sensitivity to humidity induced strain, as compared to a conventional PI-coated fibre. This feature can have direct implication in the design of PI-coated fibres to be deployed as sensors in moisture-rich environment, since it can help mitigate the impact of this cross-sensitivity on measurements.

Another interesting property of the NC coated fibres developed in this work are their electrical conductivity. By contacting a wire directly to the coating, long lengths of fibre can be heated by Joule effect. The temperature of a 14 cm sample of this NC fibre could be increased by 50 °C by applying only 350 mW electrical power to the NC coating. This can be used in sensing application requiring active heating of the optical fibre such as measuring flow speeds. The temperature dependent resistivity of reduced graphene oxide in the coating can also be used for locally measuring temperature along a section of the fibre. This measurement can be associated to distributed fibre-optic sensing techniques for multi-parameter and multi-sensing approaches using the same optical fibre. For instance, they can be used to measure temperature on the same optical fibre section for data reliability, especially in harsh conditions where one method could fail or give low signal to noise ratio. Since both sensing methods show dependance to other parameters such as strain, humidity, etc., the collected sensing data can be used to discriminate between measurands while performing multi-parameter sensing with the same fibre. This concept will be tested in a future work.

These fibres are also more thermally conductive, which could be exploited for fast thermal sensing [13], and the increased Young's modulus of the NC coating layers should influence e.g., vibration behaviour of the fibres. All these potential properties can be harnessed to improve the sensing performance of conventional polyimide-coated fibres.

The NC influence on hydrogen permeation and fatigue properties of the fibre are other parameters that deserve to be studied, as well as the incorporation of other 2D-materials in the coating for improved robustness of the fibre to hydrogen-rich environment. Another opportunity is to investigate other nanoparticles and host polymers for unlocking new sensing applications. This vibrant research field offers countless possibilities to the optical fibre sensing community, enabled by the inclusion of graphene-based materials in the fibre coating directly during the fibre draw process.

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References

[1] G. R. Pickrell, Y. Duan, B. Qi, R. G. May, and A. Wang, "Diffusion of water in optical fibres at elevated temperature and pressure," *Proc. SPIE*, vol. 4578, pp. 239–250, 2002.

[2] Z. Huang, G. R. Pickrell, and A. Wang, "Penetration rate of water in sapphire and silica optical fibres at elevated temperature and pressure," *Proc. SPIE*, vol. 43, pp. 1272–1273, 2004.

[3] L. Yu et al., "Observation of temperature dependence of the IR hydroxyl absorption bands in silica optical fibre," *Opt. Fiber Technol.*, vol. 30, pp. 1–7, 2016.

[4] P. Lecomte et al., "High temperature measurements in irradiated environment using Raman fibre-optics distributed temperature sensing," *Proc. SPIE*, vol. 9899, 2016, Art. no. 98991C.

[5] D. D. Lisowski, C. D. Gerardi, and S. W. Lomperski, "Thermal cycling testing of distributed fiber optic temperature sensors for high-temperature applications," in *Proc. 16th Int. Topical Meeting Nucl. React. Thermal Hydraul.*, 2015, pp. 7249–7260.

[6] T. Reinsch and J. Henninges, "Temperature-dependent characterization of optical fibres for distributed temperature sensing in hot geothermal wells," *Meas. Sci. Technol.*, vol. 21, 2010, Art. no. 094022.

[7] M. Lindblom et al., "Refractory lining health monitoring based on Raman optical time domain reflectometry," *Proc. SPIE*, vol. 12643, 2023, Art. no. 1264327.

[8] I. Laarossi, M. Ángeles Quintela-Incera, and J. M. López-Higuera, "Comparative experimental study of a high-temperature Raman-based distributed optical fibre sensor with different special fibres," *Sensors*, vol. 19, 2019, Art. no. 574.

[9] K. Chen et al., "Graphene photonic crystal fibre with strong and tunable light–matter interaction," *Nature Photon.*, vol. 13, pp. 754–759, 2019.

[10] J. Zhu et al., "Multifunctional polyimide/graphene oxide composites via in situ polymerization," *J. Appl. Polym. Sci.*, vol. 131, 2014, Art. no. 40177.

[11] Y. Cui, S. Kundalwal, and S. Kumar, "Gas barrier performance of graphene/polymer nanocomposites," *Carbon*, vol. 98, pp. 313–333, 2016.

[12] A. Urrutia, J. Goicoechea, and F. J. Arregui, "Optical fiber sensors based on nanoparticle-embedded coatings," *J. Sensors*, vol. 2015, pp. 1–18, 2015.

[13] Y. Guo et al., "Kilometers long graphene-coated optical fibres for fast thermal sensing," *Research*, vol. 2021, 2021, Art. no. 5612850.

[14] K. H. Tow, S. Alomari, Å. Claesson, and T. Neves, "Nanocomposite-coated optical fibres for improved distributed sensing performance in harsh environment," in *Proc. 28th Int. Conf. Opt. Fiber Sensors*, 2023, Paper Th5.3.

[15] A. Méndez and T. F. Morse, "Mechanical strength and reliability of glass fibres," in *Specialty Optical Fibres Handbook*. Cambridge, MA, USA: Academic, 2017, pp. 735–781.

[16] P. Stajanca, K. Hicke, and K. Krebber, "Distributed fiberoptic sensor for simultaneous humidity and temperature monitoring based on polyimide-coated optical fibers," *Sensors*, vol. 19, 2019, Art. no. 5279.

[17] T. Neves et al., "Recent advances in the development of a humidity-insensitive optical fiber for large-scale production," in *Proc. 28th Int. Conf. Opt. Fiber Sensors*, 2023, Paper Th6-78.

[18] C. S. Shin and L. W. Chen, "Damage monitoring of composite adhesive joint integrity using conductivity and fiber Bragg grating," *Polymers*, vol. 15, 2023, Art. no. 1575.

[19] S. Y. Chong, J.-R. Lee, C.-Y. Yun, and H. Sohn, "Design of copper/carbon-coated fiber Bragg grating acoustic sensor net for integrated health monitoring of nuclear power plant," *Nucl. Eng. Des.*, vol. 241, pp. 1889–1898, 2011.

[20] A. Nedjalkov et al., "Direct inscription and evaluation of fiber Bragg gratings in carbon-coated optical sensor glass fibers for harsh environment oil and gas applications," *Appl. Opt.*, vol. 57, pp. 7515–7525, 2018.

[21] A. Garcia-Ruiz et al., "Hermetic carbon coatings for electro-thermal all-fiber phase modulators," *J. Lightw. Technol.*, vol. 37, no. 18, pp. 4567–4572, Sep. 2019.

[22] S. Suranshe and A. Patil, "Strategically improving electrical conductivity of reduced graphene oxide through a series of reduction processes," *Mater. Lett.*, vol. 333, 2023, Art. no. 133648.

[23] A. Garcia-Ruiz A et al., "Long-range distributed optical fiber hot-wire anemometer based on chirped-pulse ΦOTDR," *Opt. Exp.*, vol. 26, pp. 463–476, 2018.

[24] N. Simon et al., "Combining passive and active distributed temperature sensing measurements to locate and quantify groundwater discharge variability into a headwater stream," *Hydrol. Earth Syst. Sci.*, vol. 26, pp. 1459–1479, 2022.

[25] P. Sehrawat, A. S. S. Islam, and P. Mishra, "Reduced graphene oxide based temperature sensor: Extraordinary performance governed by lattice dynamics assisted carrier transport," *Sensors Actuators B: Chem.*, vol. 258, pp. 424–435, 2018.

[26] J. Štulík, O. Musil, F. Josefík, and P. Kadlec, "Graphene-based temperature sensors–comparison of the temperature and humidity dependences," *Nanomaterials*, vol. 12, 2022, Art. no. 1594.

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